Reviewer #2

1. (a) Observational data to assess the regional impact of biomass burning plumes originating from Indochina peninsula have been still rare and thus the aircraft observations and the associated model simulations presented in this manuscript are important. However, more clarification is needed to justify some of the conclusions. First, the authors should be able to specify the locations and date of the fires likely affecting the studied events.

R: Figure R1 showed the fire hot spots during the study period between 14-19 and indicated a high dentist of fires frequently occurring during the study period (also, please see the annual fire number variations shown below in Q5). In this study, the biomass burning (BB) event day was on 19 March, and the backward trajectories in the East China Sea (ECS) indicated air masses mainly transported 48-72 and even 96 hrs (15-17 March) ago as shown in Figure R2. According to the backward trajectories in Figure R2, the locations of fire hot spots were distributed randomly in Indochina Peninsula (mainly 18-28 N, 90-110 E; i.e., Myanmar, Laos, Thailand, and Vietnam) as shown in Figure R1.

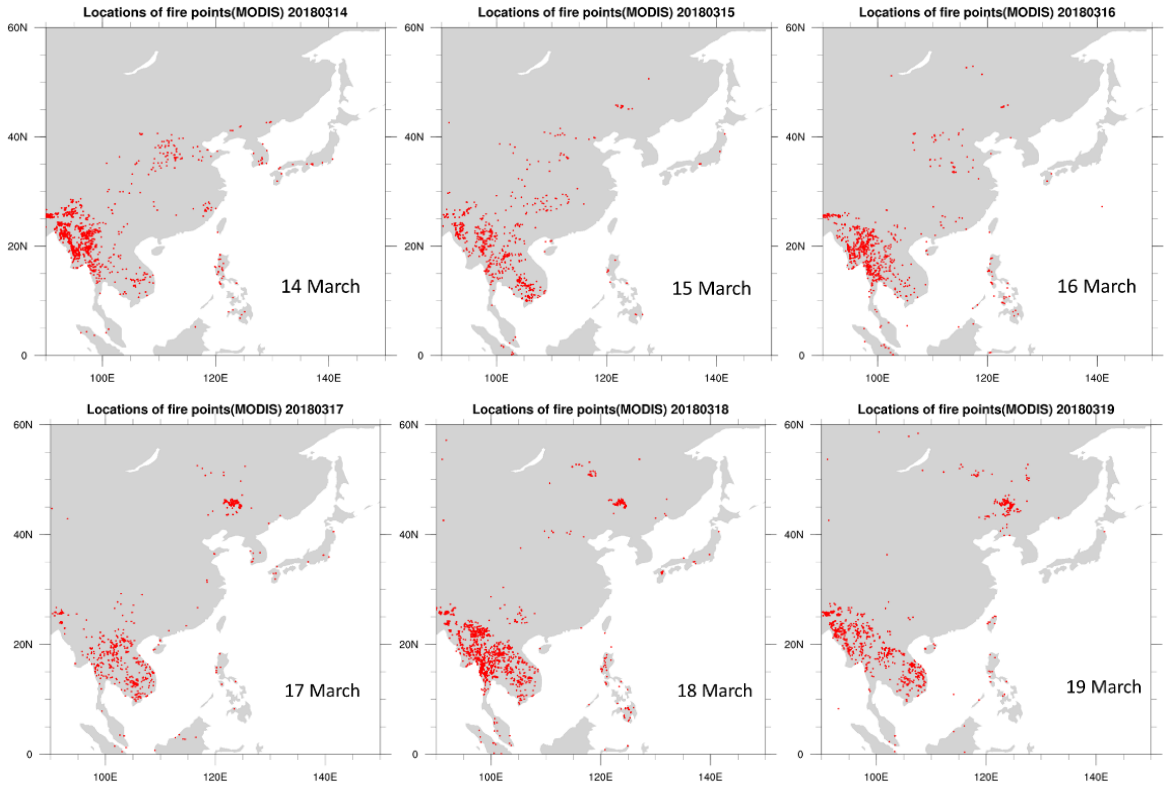


Figure R1 MODIS fire host sports during 14-19 March 2018.

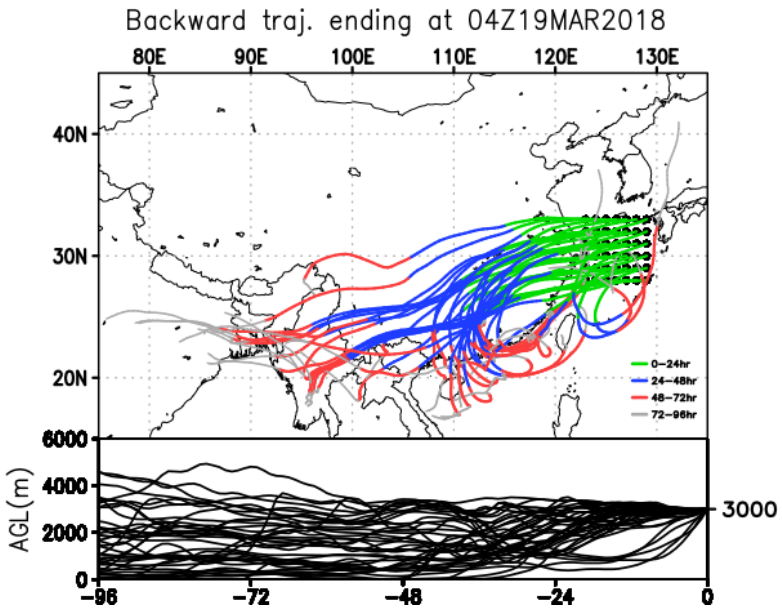
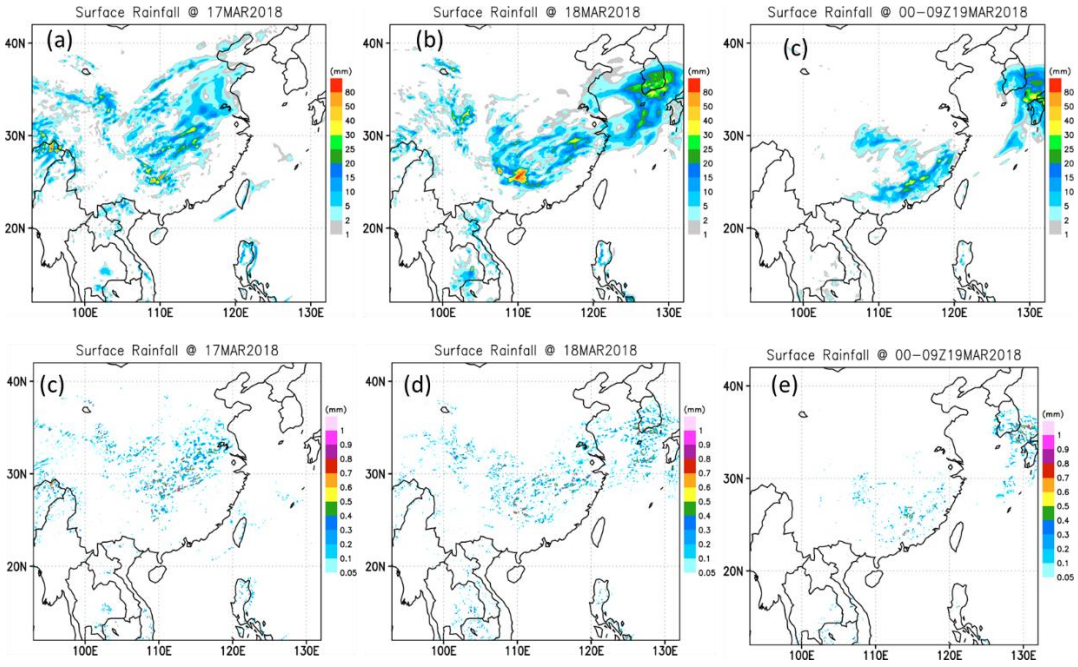


Figure R2 Result of the HYSPLIT model backward trajectory analysis at 3000 meters with multiple points by 1˚X1˚ in the area (122-130˚E, 28-33 ˚N) of East China Sea started at 04 UTC 19 March 2018.

(b) Precipitation and cloud processes during the long-range transport should be mentioned even if negligible, to characterize potential loss of aerosol species and to fully attribute the differences to the emissions

R: Thank you for the comments. Yes, the precipitation and cloud processes should play a role in the loss of aerosols during long-range transport. Figure R3 showed the simulated difference between the control simulation and the aerosol indirect effect turned off on the daily accumulation rainfall between 00:00UTC 17 and 09:00 UTC 19 March 2018. We have found that light rainfall difference (< 1 mm/day) due to wet scavenging along the frontal system (Figure R3 d-f) during the study period. We further presented the simulation of aerosol species (OA, BC) along the aircraft for the simulation only considered direct effect (case ROCD) as shown in Figure 9 in this revision. Most of the time the difference was not significant except for during 03:30-04:20 UTC along the flight where it was located north of 28 N and a frontal cloud band existed as shown in 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)**. (please also see in Q16, Figure R6 in this response)



R3 The simulated daily accumulation rainfall (a-c) and the difference between the wet scavenging turned on and off (d-f) during 17-19 March, 2018.

(c) The degrees of overestimation should be quantitatively assessed and mentioned in the abstract more clearly.

R: The statement about this part has been included in this revision **(L56-58)**

2. Second, details of chemical pathways that enhanced the OH and HO2 levels in the model should be described. This part is purely from model results - to provide associated observational evidence is recommended (also for J values, cloud condensation nuclei, and cloud water).

R: In the RACM model (Stockwell et al. 1997), the production of OH during the day was dominated by $O(^1D) + H_2O$ and $HO_2 + NO$ while $HO_2 + NO$ was dominant in the early morning and late afternoon (Kanaya et al. 2001). In polluted environments, the photolysis of other oxygen-containing species, such as nitrous acid (HONO), formaldehyde (HCHO), and hydrogen peroxide (H_2O_2) can also be important HO_x sources (Seinfeld and Pandis, 2006). In the troposphere, many gas phase species such as VOC, CO, NO_2, and other species are mainly removed through their reaction with OH (sink). Loss of HO_2 was dominated by its reaction with NO and O. Furthermore, the reactions of HO_2 with RO_2, HO_2, and O_3 also played a role (Kanaya et al. 2001; Seinfeld and Pandis, 2006)

Figure R4 indicated the aircraft measurement for the J value (JO^1D) and CCN (Cloud Condensation Nuclei; at a constant instrument supersaturation of 0.38 %) along

the flight on 19 March, 2018. The CCN number concentration (per cm^3), was consistently increased with the aerosol species (such as OA) as the flight passed through the BPTL (2000-4000 m). J value (black solid line) was high as the flight above 4000 m (03-05 UTC). In the contrast, J value decreased as the flight below the BPTL (e.g before 01:00 UTC and after 05 UTC). **(L361-366)** (also see Supplementary S3)

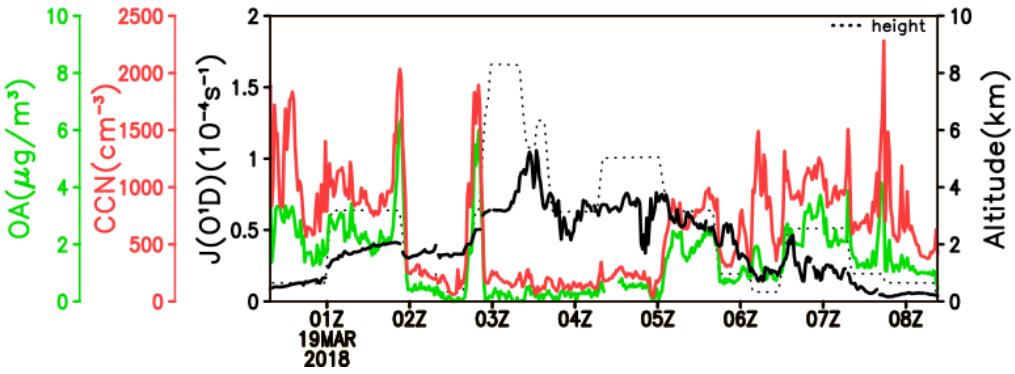


Figure R4 Observed OA concentration (green), J value (O^1D)(black solid) and CCN number (cm^{-3})(red) along with the flight altitude (dot) on 19 March 2018.

References:

Kanaya Y., Y. Sadanaga, K. Nakamura and H. Akimoto, J. Geophys. Res., Behavior of OH and HO2 radicals during the observations at a remote island of Okinawa (ORINO99) field campaign[Atmos.], 2001, 106, 24209-24223.

Seinfeld, J., and S. Pandis (2006), *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, Wiley, New York.

Specific comments.

3. Line 101. Similar to what?

R: Thank you for the comments. This sentence has been amended in this revision. We originally intended to present the emission uncertainties existing pointed out by Shi and Yamaguchi (2014). **(L104-106)**

4. Line 154. The authors state that OH and HO2 are listed in the HALO aircraft data but in fact they were not observed. (only HO2+RO2)

R: Thank you for the comment. Yes, the individual OH or HO_2 is not available in HALO measurement. It is only for modeling output. The HALO data, the total sum of peroxy radicals (RO2*= HO2 + ΣRO2,where R stands for any organic chain) has been measured by the PeRCEAS (Peroxy Radical Chemical Enhancement and Absorption Spectrometer)

instrument. The text has been amended in this revision **(L157)**. For the related response, please see in Q2.

5. Line 178. Is acetone dominant for KET? For example, MACR and MVK from isoprene chemistry could also contribute? Emissions of acetone from anthropogenic and biomass burning should be briefly discussed.

R: In RACM, the "KET" represents acetone and higher saturated ketones (KET). A common technique for reducing the number of VOC species carried in a chemical mechanism is to lump VOCs, molecule by molecule, into surrogate species representing whole classes of compounds with similar structures (Stockwell et al., 2011). The chemistry of KET is treated as a mixture of 50% acetone and 50% methyl ethyl ketone (by mole fraction)(Stockwell et al., 1997). According to Singh et al. (1994), secondary formation from the atmospheric oxidation of precursor hydrocarbons (principally propane, isobutane, and isobutene) provides the single largest source (51%) of acetone. The remainder is attributable to biomass burning (26%), direct biogenic missions(21%), and primary anthropogenic emissions(3%). **(L181-184)**
Also, there is a species "MACR" which represents methacrolein and other unsaturated monoaldehydes in RACM. The species "MVK" is not included in the RACM.

References:

Singh H.B., Hara D.O., Herlth D., Sachse W., Blake D.R., Bradshaw J.D., Kanakidou M., Crutzen P. J., Acetone in the atmosphere: Distribution, sources, and sink., J. Geophys. Res., 99,1805-1819, 1994.

Stockwell, W. R., Lawson, C. V., Saunders, E., and Goliff, W. S.: A review of tropospheric atmospheric chemistry and gas-phase chemical mechanisms for air quality modeling, Atmosphere, 3, 1–32, doi:10.3390/atmos3010001,. 879, 880, 2011

Stockwell, W. R., Kirchner F., Kuhn M.: A new mechanism for regional atmospheric chemistry modeling, J. Geophys. Res., 102, 25847–25879, 1997.

6. Lines 184, 190, and 412. MICS-Asia III and TEDS emissions were used - for which year?

R: The emission for MICS-Asia III was in 2010 and TEDS version for this study was 2013 (as shown in the original version Line 193). **(L190-192; 198)**

7. Line 200. Can the authors describe whether the intensity of biomass burning in Indochina peninsula during this particular period in 2018 was at normal level or not, in comparison to other years?

R: Figure R5 showed the statistic of annual variations of active fire detections from Terra MOIDS satellite in spring (March, April, and May) over Indochina (10˚N to 25 ˚N, 90 ˚E to 110 ˚E) from 2011 to 2020. Each MODIS active fire/thermal hotspot location represents the center of a 1km pixel that is flagged by the algorithm as containing one or more fires within the pixel. Combined (Terra and Aqua) MODIS NRT active fire products (MCD14DL) are processed using the standard MOD14/MYD14 Fire and Thermal Anomalies algorithm (https://www.earthdata.nasa.gov/learn/find-data/near-real-time/firms). Data showed the year 2018 and 2017 were 33% (1/3) lower than other years.

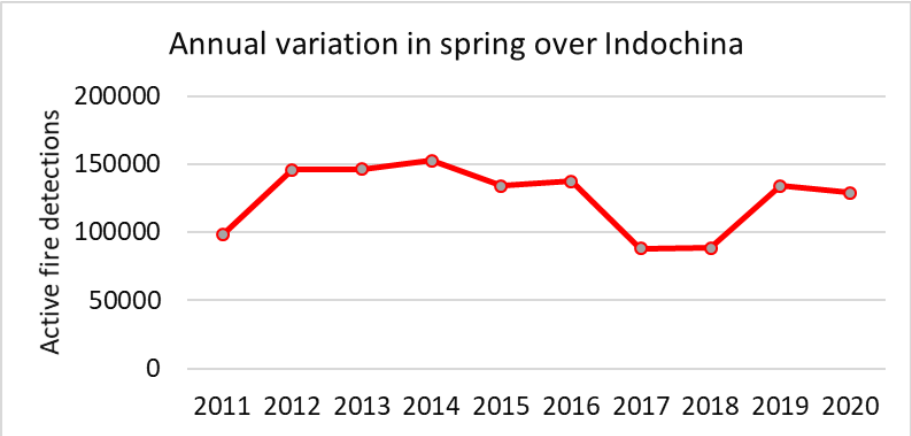


Figure R5: The annual variations of active fire detections from Terra MOIDS satellite in spring (March, April, and May) over Indochina (10˚N to 25 ˚N, 90 ˚E to 110 ˚E) from 2011 to 2020

8. Line 203. It seems that the center of the high pressure system is present over the Japan (Japan Sea), rather than Korea.

R: Text has been amended in this revision. **(L208-209)**

9. Line 269. SO2 enhancement is attributed to Japan - perhaps volcanoes have contributed?

R: During our EMeRGe-Asia campaign period, there was a volcano namely " Kirishimayama" eruption that started on 01 March and stopped on 27 June, 2018. (https://volcano.si.edu/faq/index.cfm?question=eruptionsbyyear&checkyear=2018). Our flight to the north was between 30-32 N (manuscript Figure 6a), i.e. in the south of Japan on 19 March, 2018. In other words, it can not rule out the contribution of this volcano to this high sulfate concentration under favorable weather conditions. However, it is difficult to identify due to unregular release from the volcano and weather conditions.

10. Line 311. Carmichael

R: The text has been amended in this revision**.(L316)**

11. Line 312. Figure 6b indicates biomass burning influence is spread to the north of 30 degN.

R: Thank you for the comments. Text has been amended in this revision. **(L315-321)** However, the ACN still could be around 300ppt or less as the flight at the north of 30 ˚N (during 3:30-4:30 UTC) and could be recognized as the contribution of BB (Förster et al. 2022). In other words, it might still have BB products being transported to the north of 30 ˚N under favorable weather conditions although the ACN concentration was low compared to the south of it at the layer of BPTL(between 2000 and 4000 m).

Ref:

Förster, E., Bönisch, H., Neumaier, M., Obersteiner, F., Zahn, A., Hilboll, A., Kalisz Hedegaard, A. B., Daskalakis, N., Poulidis, A. P., Vrekoussis, M., Lichtenstern, M., and Braesicke, P.: Chemical and dynamical identification of emission outflows during the HALO campaign EMeRGe in Europe and Asia, Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2022-455, in review, 2022.

12. Line 338. As ACN and ACE contain oxygen and nitrogen in their molecules, they are not hydrocarbons.

R: The text has been amended in this revision. **(L347)**

13. Line 350 and 352. Use uppercase 1 for J(O1D).

R: Thanks for the comments. The text has been amended in this revision. **(L358, 361)**

14. Lines 351 and 479. Whether aerosols increase or decrease J(O1D) will be dependent on the assumed single-scattering albedo. Any evidence from direct observations of the J values?

R: Please see the response in Q2 and Figure R4.

15. Line 404. It is better to confirm that the CO hemispheric baseline is not overestimated.

R: Time series of background CO mixing ratios during the 1990s, averaged over the Extratropical Northern Hemisphere was between 100-200 ppb (Wotawa et al. 2001). Furthermore, our study focused on the BB contribution during the event day (19 March 2018) which is a short-term period as presented in this study. The simulated CO

concentration difference with and without BB emission is not related to the global information.

Ref: Wotawa G., Novelli P.C., Trainer M., Granier C.: Inter-annual variability of summertime CO concentrations in the Northern Hemisphere explained by boreal forest fires in North America and Russia: Geophys. Res., Lett., 28, 4575-4578, 2001.

16. Line 416. It is important to confirm that OA and BC have not been removed by wet deposition on the way of transport, to better attribute the model's overestimation to emissions.

R: Thank you for the suggestions. We further checked and identified that OA and BC have been removed by wet scavenging (due to aerosol indirect effect) during 03:30-04:20 UTC. 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. When the simulation only considered the direct effect (case ROCD, purple) below,the overestimations were increased as shown in Figure 9b-c. **(L436-441)**

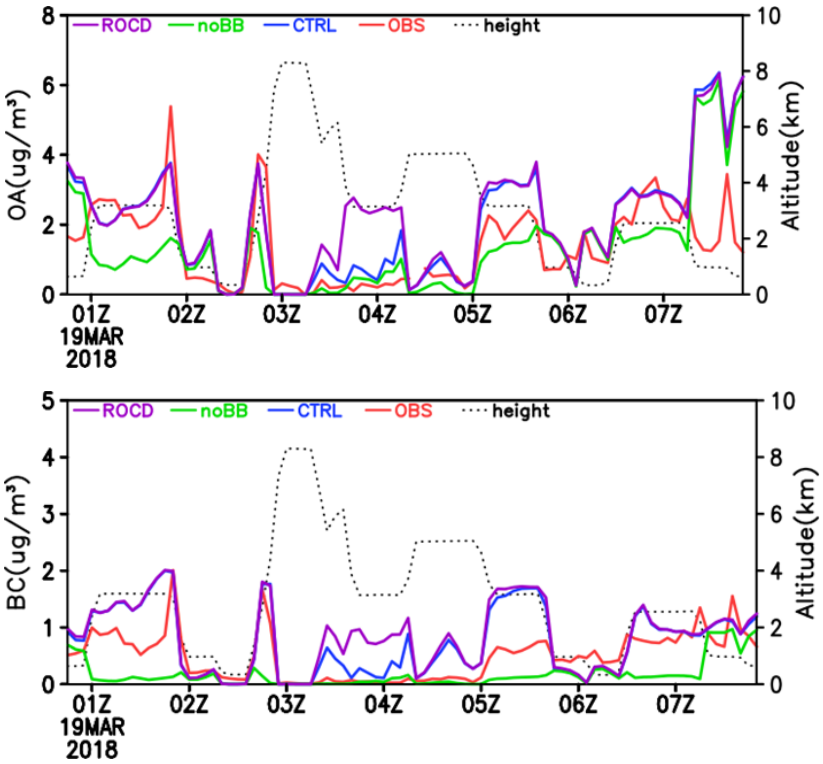


Figure R6 Observed (OBS, red) and simulated concentration (CTRL, blue), and the simulation without indirect effect (ROCD, purple), without BB emission (noBB, green) along with the flight altitude for (a) OA (μg m^{-3}) (b) BC (μg m^{-3}) on 19 March 2018.

17. Line 447. "detraining" is difficult to understand.

R: Thank you for the comments. The text has been dropped in this revision.

18. Line 457. The sentence starting with" The variation trend of PM2.5 ..." needs to be rewritten.

R: Thank you for the comments. This sentence has been rewritten in this revision. **(L476-478)**

19. Line 471. Which processes were responsible for the OH and HO2 enhancement? How well VOCs emissions and chemistry were treated to describe the OH and HO2 budget?

R: In general, the photolysis of ozone followed by the subsequent reaction of $O(^1D)$ with water vapor is the main HOx source during daytime in the clean troposphere. In the troposphere, many gas phase species such as VOC, CO, NO2 and other species are mainly removed through their reaction with OH (sink). HO2 production followed the reaction of peroxy radicals with NO, with additional contributions from formaldehyde photolysis and reactions of OH with CO and formaldehyde. Please also see the response in Q2.

20. Line 513. Any observational evidence of CCN or cloud water enhancement, attributable to the biomass burning plume?

R: This question has been responded in Q2 and Figure R4.

21. Figure 3a, b: As the highest CO area is distant from Indochina peninsula on the day, the authors should be able to state the possible locations and date of fires producing the plumes.

R: To identify the high CO concentration in the South China Sea at 1000 meters in Figures 3a and b, the backward trajectories with multiple points by 1˚X1˚ in the area (110-115˚E, 17.5-22.5 ˚N) in the South China Sea started at 00 UTC 17 March 2018 as shown in Figure R7. The locations of fire hot spots were distributed randomly in Indochina Peninsula as shown in Figure R1 (Q1). The backward trajectories in the South China Sea indicated air masses mainly transported 48-72 and even 96 hrs. In other words, there could be contributed by fires occurring between 100-110 E and 12-20 N (Myanmar, Laos, Thailand, and Vietnam) during 13-15 March 2018.

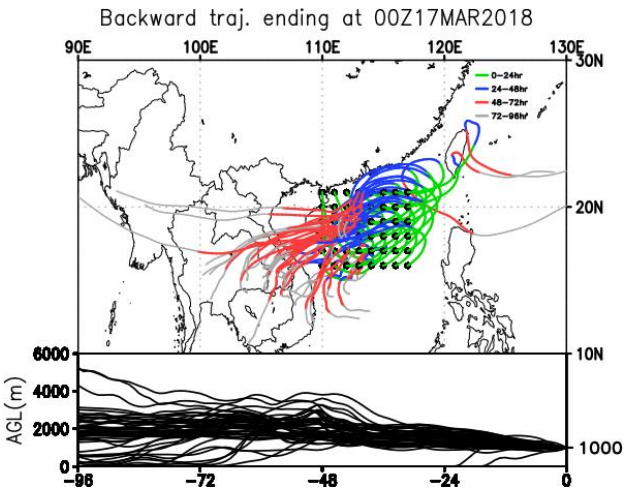


Figure R7 Result of the HYSPLIT model backward trajectory analysis at 1000 meters with multiple points by 1˚X1˚ in the area (110-115˚E, 17.5-22.5 ˚N) of East China Sea started at 00 UTC 17 March 2018