Response to Referee Comments #2 on manuscript egusphere-2024-2064:
"Downward and upward revisions of Chinese emissions of black carbon and CO in bottom-up inventories are still required: an integrated analysis of WRF/CMAQ model and EMeRGe observations in East Asia in spring 2018"

Referee Comment (RC): **General remarks and questions**

This preprint reports aircraft measurements of black carbon (BC), CO, and CO2 measured in the lower troposphere eastward along the Asian continent over the sea and around Japan and the Philippines. Pollution plumes with elevated levels of CO and BC are investigated and the observed concentrations are compared with predictions from the WRF-CMAQ model. Observed to modeled concentration ratios derived from this are proposed as correction factors for emission inventories used in the model. Such an investigation of emission inventories is useful to achieve improved air quality predictions in the regions studied, and the aircraft measurements presented here provide a rare and good opportunity to extend analyses from ground based measurements.

Unfortunately, the presentation of the scientific approach and results in this paper is somewhat confusing and difficult to understand in places. The following aspects are not addressed clearly enough for general understanding and require a more detailed discussion. Major revisions are required before the paper is suitable for publication in ACP.

Author Comment (AC): We appreciate the referee's thorough review and constructive feedback on our manuscript. We have addressed all concerns through substantial revisions, ensuring clearer presentation of the scientific approach and results. The manuscript now provides enhanced clarity, improved methodological descriptions, and a more detailed discussion. Below, we provide detailed responses to each point raised and outline the corresponding revisions. Please note that the structure of the manuscript has been changed based on regions according to the comments of the first referee. Section 3 (Results) now includes 2 subsections: 3.1 for three polluted cases (THL, PHL, JPN) including comparisons of concentrations and residual ratios; 3.2 for CHN case including comparisons of concentrations, residual ratios, and estimate for the emissions, with a separate part for uncertainty assessments. Section titles were also renamed. Revised points are highlighted in the manuscript. Minor textual adjustments and reconstructed text are not highlighted.

RC: (1) To what extent can CO, CO2, and BC be considered as inert tracers that are essentially only subject to transport and deposition after emission on a 5d time scale, as considered in the

backward trajectory calculations. To what extent could other chemical or physical processes influence their concentrations observed on the aircraft?

AC (Revisions made in **Lines 171-179, 206-218**): Note that the time it takes for the air mass to travel from the sources to the observation points for the case studies is less than five days, typically < 40–120 hours from CHN, 0–24 hours from Manila (PHL), 10–120 hours from the broad fire region near THL, 0–36 hours from JPN (mentioned in Sect. 2.3., L196-198). During transport over less than 120 hours, CO, CO_2, and BC remain largely inert. CO undergoes slow oxidation with OH as its atmospheric lifetime is several weeks. As a stable gas, CO_2 is highly inert, particularly when transported at high altitudes without contact to vegetation, with deposition playing a minor role. BC amounts are affected by deposition. VOCs, including biogenic, undergo photochemical oxidation processes involving NO_x, leading to the formation of O_3 and secondary organic aerosols (SOA), which can accelerate BC aging. However, in polluted regions with strong emissions, this effect remains minor (**Sect. 2.2, L171-179**).

Since the flights were conducted away from rain events, the accumulated precipitation along the trajectories (APT), as derived from HYSPLIT rainfall data, helps to exclude data significantly affected by wet deposition. A detailed analysis of APT was conducted to assess the influence of rain on emission characteristics, applying APT criteria for the air masses from CHN and THL (APT 72 h < 1 mm), PHL E-AS-06 and E-AS-10 (APT 24 h = 0 mm), PHL E-AS-03 S2 (APT 10 h = 0 mm), JPN (APT 36 h = 0 mm) based on their travel time to the aircraft (CHN 40–120 h, THL 10–120 h, PHL E-AS-03 S2 0–10 h, E-AS-06 and E-AS-10 0–24 h, JPN 0–36 h; Figure S6, Table S6, S7). The APT 72 h < 1 mm criterion has been previously analyzed and showed no large influence of wet deposition on emission-concentration relationship (Kanaya et al., 2016, 2020), whereas the APT = 0 mm criterion aims to completely exclude the effect of rain. In addition to trajectory calculations, we carefully discuss deposition, dilution, and convection effects, as well as the representativeness of the air masses in the manuscript (**Sect. 2.3, L206-218**).

We provided additional discussions in **Sect. 2.2** and **2.3**.

RC: (2) Which source regions were exactly investigated using the approach with model calculations and flight measurements? Line 108 states: “Pollution plumes from major population centers in Asia were detected during parts of flights.” Which major population centers are specifically meant? Asia is a very large continent.

AC (**L111-113**): We added the specification for Gulf of Thailand, Manila (Philippines), Osaka (Japan), and Central East China including Beijing, Hebei, Henan, Shanghai.

RC: Generalized statements such as “The results suggested that downward and upward revisions of Chinese emissions ... are required” (see abstract) is VERY general and should be formulated more cautiously and refer specifically to the regions or areas investigated.

AC (**L31**): The results suggest that revisions to Chinese emissions inventories are necessary, with a focus on Central East China during the polluted spring season, where our trajectory analysis indicates significant influence. Although our trajectories cover only Central East China for the aircraft observations, combining results from our previous study at Fukue Island with extended temporal and geographical coverage enabled discussion about the national-scale emission estimates. Associated uncertainties in extrapolating our findings to larger scales in terms of seasonal and spatial variations have been included. We added in the abstract where the inventory revision should focus on based on this and our previous studies.

RC: The authors should also consider making the title of the paper more specific.

AC: We made a change to the **title** i.e. “Assessing BC and CO Emissions from China Using EMeRGe Aircraft Observations and WRF/CMAQ Modeling”

RC: Also, to what extent could the findings be limited in time, e.g., only for the season or year investigated? How representative were the environmental conditions (fire activity, traffic, meteorology) prevailing during the flight campaign for the years before and after?

AC (**L456-464, 607-610**): Although the aircraft data for the China case was collected in spring, our estimation method was applied to all of China in 2018, accounting for representative uncertainties, including seasonal variations. Note that the uncertainty associated with representativeness has been discussed in the original text **(Sect. 3.2.4,** L582-585), additional explanation was added at **L609-610)**, supported by our previous year-round investigations (Kanaya et al., 2016, 2020). The environmental conditions during the campaign ensure the captured plumes were representative of dominant emission sources and seasonal patterns. Fire emissions account for only ~10% of anthropogenic emissions in China (7% for BC and 13% for CO, according to GFED and HTAPv2.2z inventories used in CMAQ), the detected pollution plumes by the aircraft predominantly originate from Chinese anthropogenic sources. Trajectory analysis indicates that the observed polluted plumes travelled from Chinese source regions starting around Thursday, 22 Mar 2018, and took 2–4 days to reach the aircraft. Consequently, the probed air masses were influenced by the typical weekday emissions from the transportation sector. The analyzed plume originating from a major population center in China was one of the most significant air pollution transport events recorded also by our long-term ground-based

observations at Fukue Island in western Japan over the past 10 years. The event was regarded as typical regarding its transportation route and negligible deposition loss (**Sect. 3.2.1, L456-464**). The annual and nation-wide emission estimates appear to be extrapolations using a limited set of aircraft observations. This extrapolation is supported by the results from our previous analysis, which used long-term ground-based observations (Kanaya et al., 2020) (**Sect. 3.2.4, L607-609)**. This point has been clarified in the revised text. Our conclusion is obviously limited to the year of observations (2018).

RC: (3) The title of the paper states "Downward and upward revisions … are still required…". Is this statement still valid approximately seven years after the aircraft campaign? Have there been any developments in East Asia since then with regard to biomass burning, industrial emissions, or traffic that would warrant a new investigation?

AC (**L618-621, 622-626**): The estimated emissions and revision suggestion in this study apply to China in 2018, aligning with broader trends indicating a decline in BC emissions since 2010 (Kanaya et al., 2020) and still low levels in 2019–2020 (0.6 Tg yr^{-1}) according to the translation from TCR-2 CO data. However, updated inventories, such as CEDS v2021-02-05 and MEICv1.0 (Zhang et al., 2021), continue to show BC emissions exceeding 1 Tg yr^{-1} from 2018 to 2020, reinforcing the need for a downward revision (**Sect. 3.2.4, L618-621**). The revised manuscript title "Assessing BC and CO Emission from China Using EMeRGe Aircraft Observations and WRF/CMAQ Modeling" does not overstate the results.

Changes since 2018, particularly in biomass burning, may influence emission trends. China's 13th Five-Year Plan for Biomass Energy Development (2016) promotes biomass energy, with further emphasis on biomass heating in the IEA Bioenergy Country Report (2021) and China Energy Transformation Outlook 2023 (COP28, December 2023). Meanwhile, vehicle exhaust remains a major pollution source despite advances in electrification and emissions control. These evolving factors could shift BC, CO, and CO_2 trends in uncertain directions, warranting future investigations. Nevertheless, the inventory revision suggestions for 2018 in this study remain worth considering (**Sect. 3.2.4, L622-626**).

We added a discussion in the last part of Sect. 3.2.4.

RC: (4) There is only very limited information given on the EMERGE campaign and the aircraft measurements. A brief overview should therefore be provided in Section 2.1. The following information would be of interest. Why was springtime chosen for the campaign?

AC (**L106-109**): We provided more detail for the EMeRGe campaign. Springtime in East Asia is characterized by peak emissions from biomass burning in Southeast Asia as well as by continental outflow from China that carries pollution to observable regions.

RC: What altitude range was covered during the measurement flights and what altitude range was specifically evaluated?

AC (**L114-115**): Flight altitudes during the measurements ranged from ground level to approximately 12 km, with ranges around 0.3–2 km were specifically evaluated for designated polluted areas.

RC: Which measuring instruments were used (manufacturer, model) and what are their total measurement errors (precision, accuracy)?

AC (**L117-118, 126-129**): We added more details on measuring instruments in Sect. 2.1.

RC: Which particle sizes were detected by the BC measuring device and how was the photometer signal converted into mass concentration? For which reference conditions (T, p) do the specified mass concentrations apply?

AC (**L118-125**): SP2 detects BC cores with mass-equivalent diameters of 70–500 nm by measuring time-dependent scattering and incandescence signals as particles cross a Gaussian-shaped laser beam (λ=1064 nm) (Schwarz et al., 2006). Avalanche photo-diode detectors capture scattering and incandescence at high and low gain across two wavelength ranges (350–800 nm and 630–880 nm). Particles scatter light based on optical size, while BC-containing particles absorb the laser, heating to ~4000°C and emitting incandescence. The peak intensity of this signal is linearly proportional to BC mass, calibrated using a correction factor (Laborde et al., 2013). BC concentrations were normalized to standard temperature and pressure (STP, T_0 =273.15 K, p_0 =1013.25 hPa). The SP2 incandescence signal was calibrated using size-selected fullerene soot particles at the beginning, during, and end of the campaign (Holanda et al., 2020). We incorporated the explanation into the text and included two references (Schwarz et al., 2006 and Laborde et al., 2013).

RC: Were other compounds (e.g., NOx, VOCs) measured and used in this work, e.g., for the identification of plumes, apart from BC, CO, and CO2?

AC (**L130-131**): Other compounds, including NO_x, OVOC (HCHO) and VOCs, were measured during the campaign but not utilized in this study. While NO_y was initially considered to investigate, its complex mechanisms during transport led to its exclusion.

RC: A PTR-MS instrument is mentioned in line 112. What was measured by this instrument and were the corresponding data used in any way?

AC: The PTR-MS is used to measure VOCs but these measurements not used in this study. We removed this sentence in the revised manuscript.

RC: (5) Section 2.2: which processes (apart from emission, transport, and deposition) play a role for the simulation of BC, CO, and CO_2 on the 5d time scale of the backward trajectories investigated? Table 1 lists various types of emissions. Apart from direct emissions of BC, CO, and CO_2, do other emitted substances, for example biogenic VOCs mentioned in the table, play a role? If so, which chemical processes were involved? Which processes represented in the AERO 6 aerosol module are relevant for the modeled results of BC in this study? What role do the volcanic emissions listed in Table 1 potentially play?

AC (**L151-153, 171-178**): During the transport of about 5 d, CO, CO_2, and BC are effectively inert. CO undergoes slow oxidation with OH, with a lifetime of weeks. VOCs (and biogenic VOCs) undergo photochemical oxidation processes involving NO_x, leading to the formation of O_3 and secondary organic aerosols (SOA), which in turn increases the BC aging, increasing hygroscopicity and enhancing removal through deposition. Although oxidation of biogenic VOCs can be an important source of CO, the plumes analyzed in this study originated from metropolitan areas, suggesting only a minor contribution from biogenic sources. While CO_2 is absorbed by vegetation and marine phytoplankton, the CO_2 levels at altitudes of aircraft observations would be minimally influenced (**Sect. 2.2, L171-173, 175-178**).

In AERO6, BC concentration is unaffected by chemical or heterogeneous reactions, while coagulation alters particle size without affecting BC mass. Dry deposition loss of BC has been tested as negligible during transport over 3 days (Fig. 6, Kanaya et al., 2016) (**Sect. 2.2, L151-153**). Wet deposition was found negligible in cases where the total amount of rain along the trajectory after the emission occurred is less than 1 mm (Kanaya et al., 2016, 2020). The analyzed measurements are in periods which are essentially free of wet deposition as indicated by the accumulated precipitation. Though the wet deposition scheme in the model may need to be tested and revised, this dependency would have had minimal influence on the analysis and conclusions of this study (**Sect. 2.2, L173-175**).

The volcanic entry was **removed from the table**.

RC: (6) For each model component listed in Table 1, references should be provided where a description or corresponding data can be found.

AC: We added the references for each model component in **Table 1**.

RC: (7) In Section 2.4, the general description of the concept of "residual ratios" and "baseline values" is hard to understand and should be rephrased for clarity.

AC (**L223-225** and **261**): We adjusted the text for "residual ratios" (Sect. 2.4) and "baseline value" (Sect. 2.5) for clarification.

Minor Comments

RC: Abbreviations (EMERGE, WRF-CMAQ, GFED, HTAPv2.2z, etc.) should be defined at the beginning of the paper when they are first used.

AC: We have added the abbreviation definitions in the abstract and other relevant sections.

RC: Line 42: Typo "nanometre-sized"

AC (**L44**): It was changed to "nanometer-sized"

RC: Lines 96-97: The sentence is unclear. Which other pollutants (apart from CO, CO2, BC) were investigated?

AC (**L88**): We restructured the paragraph, this sentence became part of the study target (2) statement, and the old phrase was omitted.

RC: Line 105: Typo "Deutches"

AC (**L110**): It was modified to "Deutsches"

RC: What is the meaning of the given quantities (e.g., max = 1.3 mm) for APT3 in lines 340 and 575?

AC (**L197, 208-211, 484-485**): We changed the term to APT 72 h (accumulated precipitation along 72-hour trajectories). Previous studies have shown that a low APT 72 h value (e.g., < 1 mm) indicates weak wet deposition, which does not significantly influence emission-specific

characterization (Kanaya et al., 2016, 2020) (**Sect. 2.3, L210-211**). As the CHN air masses typically travel for 40−120 hours from source to the aircraft, APT 72 h was selected as the empirical criteria that the wet deposition becomes important for the CHN air masses (Kanaya et al., 2016, 2020) (**Sect. 2.3, L197**). We applied APT analysis using the criterion APT 72 h < 1 mm for CHN case, and omitted the specified APT maximum quantities as stated in the unrevised manuscript. Adjustments were made with the term expressions (**Sect. 3.2.2, L484-485**). Besides APT 72 h used for CHN air masses, we used different time range for other cases (**Sect. 2.3, L208-209**).

RC: Table S3 appears to have no use in the paper and should be deleted.

AC (**L507**): Table S3 describes in sufficient detail how the ECLIPSEv6b inventory assigns BC emission factors for each fuel type based on activity levels, abatement, and capacity control in CHN, supporting the discussion of dominant BC-emitting sectors in the original text (**Sect. 3.2.2**, L504-507). We have included the relevant citation for Table S3 (**L507**).