Reply to Anonymous Referee #1

This study combined ozonesonde observations, validated EAC4 reanalysis products, and ground-level measurement data to characterize an ozone stratospheric intrusion (SI) event in June 2013 in 3 cities of China and claimed to have developed a method for quantifying the contribution of SI events to ground-level ozone pollution based on EAC4 reanalysis data. This study is innovative and representative in the field of atmospheric environmental chemistry and atmospheric science as so far, but the following possible doubts remained. I recommend the publication of the manuscript after the authors have properly answered or resolved these possible questions below.

Reply: Thank you very much for your positive and detailed comments. We have carefully considered your suggestions and comments, and made corresponding modifications and explanations.

Major Comment:

1. **Lines 150-151:** "High-level secondary ozone peaks are a characteristic O_3-profile structure associated with tropospheric folding, a major form of SI in the extratropical region" How do you define the range of high-level secondary ozone peaks? It didn't seem to be specific values but a range. Why it is a major form of SI? Here maybe need a simple explanation by 1-2 sentences.

   Reply: Thank you for this comment. Secondary O_3 peak is defined as the abnormal O_3 peak near the tropopause (9-16 km). Here, "secondary" is relative to the first O_3 peak in the stratosphere. We clarified this definition in the revised manuscript. Referring to previous ozonesonde-based observational studies, we claimed that secondary O_3 peak in a height range between 9 and 16 km (i.e., near the tropopause) is a characteristic O_3-profile structure when SI occurs and triggers tropopause folding. Lines 148-150.

2. It's true that Beijing and Changchun are belong to extratropical region, but I'm not sure if Hong Kong is. Maybe it's more subtropical or tropical. If so, can the SI still be determined based on the secondary ozone peaks?

   Reply: Thank you for pointing it out. Hong Kong is a subtropical city. In Hong Kong, we observed a sub-high O_3 layer in the lower free troposphere (4.2–8.1 km height), rather than the upper-level O_3 peak near the tropopause. The sub-high O_3 layer in Hong Kong is a transport result of the extratropical stratospheric O_3 intrusion. However, secondary O_3 peaks in the extratropical region represent the initial and major characteristic of stratospheric intrusion.

3. There are some confusions in **Table 1**: Which statistical comparisons were based on Beijing, Changchun and Hong Kong, and which were based on the 76 cities? This should be described more clearly in the table. In addition, it would be best to give in the table the number of data points N used for correlation analysis, which is an important statistical parameter. Finally, how about the correlation between O_3-Sonde & O_3-Ground? Maybe it has been shown in Zhang et al. 2013, but it's better to be analyzed here by the same statistic method in this study to tell readers the accuracy of ground-level ozone measurements by ozonesondes. In addition to Table 1, I suggest the authors also include scatter plots in the supplement to compare the observation data and ozone products.

Reply: Thank you for picking it up. According to your comments and suggestion, we present the statistical comparisons in the form of scatter plots in this revision, instead of the Table form in the initial manuscript. In the scatter plots, we list the important statistical parameters, including the data points *N*, correlation coefficient *R*, mean absolute bias MAB, root mean square error RMSE, and index of agreement IOA. Besides, we add the correlation statistic between the O_3-Sonde & O_3-Ground using 9 ozonesonde samples in three cities (Beijing, Changchun, and Hong Kong). Although a very limited ozonesonde samples, the O_3-Sonde correlated well with the O_3-Ground (N=9, R=0.981, MAB=3.2 ppbv, RMSE=9.8 ppbv, and IOA=0.994), indicating the accuracy of ozonesondes. Lines 85-87 and Figure 3.

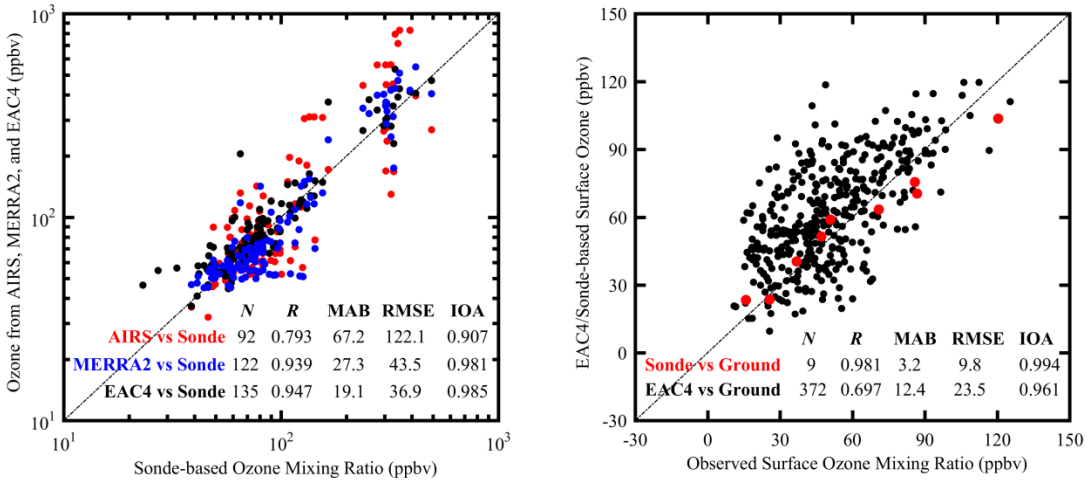

Figure 3. (A) Validation of AIRS, MERRA2, and EAC4 O_3 products with 9 ozonesonde observations from Beijing, Changchun, and Hong Kong. (B) Validation of EAC4/Sonde-based surface O_3 concentrations with ground-based O_3 observations. In (A), AIRS, MERRA2, and EAC4 O_3 data were spatially interpolated to the location of ozonesonde stations. In (B), ground-based O_3 observations across 466 sites in 76 cities were resampled to EAC4 grid (0.75° × 0.75°) for comparison with EAC4-based surface ozone renalysis; ground-based O_3 observations at three neighboring sites (Tiantan site in Beijing, Daishan Park site in Changchun, and Sham Shui Po site in Hong Kong) were used for comparison with Sonde-based surface ozone concentrations. *N*, *R*, MAB, RMSE, and IOA denote the number of statistic samples, correlation coefficient, mean absolute bias, root mean square error, and index of agreement, respectively.

4. **Lines 237-239:** Please include a scatter plot in the supplement for comparing observation data and EAC4 ozone reanalysis product.

Reply: Thanks for this suggestion. As you suggested, we embedded a scatter plot in Figure 3 to compare observation data and other ozone products. Seeing Figure 3 in the revised manuscript.

5. **Section 3.3.** How did you get the contribution of stratospheric intrusions to surface ozone pollution? More specifically, how did you quantitatively distinguish between local photochemical production and stratospheric intrusion? Should there be a description of the relevant calculation process or equations in Section 2 (*Datasets*)?

Reply: Thanks for your suggestions. The ECA4 provide two ozone indicators, the total ozone (O_3) and the stratospheric ozone tracer (O_3S). The stratospheric ozone tracer is a part of total ozone, representing the stratospheric ozone contribution in total ozone. So the contribution fraction is calculated as $CF = 100\% \times O_3S/O_3$. Line 280.

Minor Comment:

1. **Lines 43-45:** When citing multiple studies, could you also specify where the SI events occurred?

   Reply: Thanks for this suggestion. In the revised manuscript, we specified where the SI events occurred. (Lines 40-43).

2. **Lines 58-60:** Any other literate from places other than China?

   Reply: Regarding the opposite conclusions drawn from different chemical tracers, no other literate had been reported from places other than China.

3. **Lines 76-78:** Move the sentence of "Ozonesondes provide … " to Introduction paragraph 2. Just describe the method you used here, with the ozonesondes parameter shown in detail, and if it is refered to Zhang et al. 2013 please please specify this point.

   Reply: Thanks for this suggestion. We moved the mentioned sentence to Introduction paragraph 2 (Lines 46-48). Besides, we specified that the details of the intensive ozonesonde experiment can be found in Zhang et al. (2013). (Lines 79-80)

4. **Lines 155-156:** "Similar sub-high ozone layer (> 80 ppbv) also occurred in the lower troposphere (3.5–6.0 km height) of Hong Kong on June 13." What were the similarities? Was the extreme low humidity and the sub-high ozone layer occurring at the same time? But the humidity in Hong Kong was not extremely low. Why there was no sub-high ozone layer in Beijing and Changchun on June 13 (4.0–6.0 km height also with high-ozone and low-humidity condition)?

   Reply: Thanks for this comment. The sub-high O_3 layer emerged in the lower troposphere (3.5–6.0 km height) of Hong Kong on June 13 is similar to that occurred in the middle troposphere (4.2–8.1 km height) of Changchun on June 12. Both of them are featured with extreme low humidity and high ozone concentration. The relative humidity in Hong Kong declined to 20%, reaching an extreme low value in subtropical coastal region. Although this humidity is not as low as that in Changchun (near zero), but it is lowered from the relatively higher humidity background (80% below and above the sub-high O_3 layer). For a subtropical coastal city such as Hong Kong, its humidity background is significantly higher than that in Changchun (a middle-latitude inland city). In fact, the declining amplitudes of RH in the sub-high O_3 layer are similar in Hong Kong (from 80% declined to 20%) and Changchun (from 60% declined to 0%).

   Why there was no sub-high ozone layer in Beijing and Changchun on June 13? This is a good question. Three-dimensional dynamics associated with upper-level troughs involves stratospheric

dry intrusion (SDI) and warm conveyor belts (WCB) airstreams. The SDI originates in the lower stratosphere on the cold side of the trough (west of the trough axis) and descends behind the cold front, while the WCB originates in the warm sector of the trough (east of the trough axis), ascending rapidly to the mid- and upper troposphere. Given the dynamical characteristics of upper-level trough, the mid-latitude region is mainly controlled by SDI airstream (the dynamical mechanism of upper-level secondary O_3 peaks over Beijing and Changchun). When WCB airstream entangles part of the dry-intrusion air masses in the relatively lower latitudes, it will lead to northward recirculation of pre-intruded O_3 to mid-latitude region, causing the occurrence of sub-high O_3 layer in the middle troposphere (the dynamical mechanism of sub-high O_3 layer over Changchun on June 12). On June 13, both Beijing and Changchun was far away from WCB airstream due to its eastward movement, so no occurrence of sub-high ozone layer.

Lines 155-158: The author observed a sub-high ozone layer in the lower troposphere of Hong Kong on June 13. The vertical profiles of T, RH, and O_3 at that layer are like those around 4 km in the troposphere of Beijing on June 10th. But there is no discussion for the latter one.

Reply: On June 10th, the O_3 concentration showed no peak around 4 km above Beijing (despite slight increase around 4 km but peak around 8 km) although the relative humidity presented a valley. This structure is not similar to that (the co-occurrence of O_3 peak and RH valley) in Changchun (on June 12th) and Hong Kong (on June 13th). We had no enough evidence to attribute the low-humidity layer over Beijing to stratospheric intrusion. So, there is no discussion for this low-humidity layer.

5. **Line 194:** I can not see the trough axis in Fig.3. Could the author highlight them in the figure?

Reply: Thanks for this suggestion. We added the trough axis in the revised manuscript. Figure 4A.

6. **Line 197:** I don't how 2000 km is concluded here.

Reply: Thank you for picking it up. We labeled the SDI-induced O_3-rich belts in Fig. 4A. It stretched from extratropics (40^{o}N) to subtropics (20^{o}N), approximately 2000 km.

7. **Lines 271-273:** It is difficult for me to see how the wind-driven dispersion of the intruded O_3-rich stratospheric air is concluded here. Could you provide more discussion on this?

Reply: Thank you very much for this comment. We made a wrong explanation for the high O_3S concentrations over the Mongolian Plateau. We adjusted the view to examine the three-dimension structure of O_3S, and found a secondary SI emerged over the Mongolian Plateau since June 12. This secondary SI is the reason of high-concentration O_3 over the Mongolian Plateau. However, this elevated O_3 concentrations had no influence on eastern China due to its downwind location. Lines 241-244, and 284-287.

8. The title, abstract, introduction, and summary all mention "fully-validated EAC4 reanalysis", but the text of results is unclear in explaining this process of "fully-validated". What is the definition of fully-validated?

If it is only reliable O_3 correlation validation then being called "fully" is not appropriate even too absolute. It might be that "O_3-validated EAC4 reanalysis" or just "validated EAC4 reanalysis" would be sufficient.

Reply: Thanks for this wonderful suggestion. As your previous suggestion, we added a scatter plot to show the well performance of EAC4 reanalysis. We accepted your suggestion and changed the tile to be "Widespread stratospheric intrusion influence on summer ozone pollution over China revealed by multi-site ozonesonde and validated EAC4 reanalysis". In this new title, we deleted "ground-based measurement" because the ground-based measurement is not as important as multi-site ozonesonde and validated EAC4 reanalysis in this study.

Technical Comment:

1. **Line 44:** SI event should be plural.

   Reply: Thank you for picking it up. We re-wrote the sentences in the revision.

2. **Lines 49-52:** Could you rearrange the citations? Which references correspond to satellite observations, and which are for atmospheric reanalysis and model simulations?

   Reply: Thank you for this suggestion. We rearranged the citations. Lines 50-54.

3. **Line 53:** Which open-source products? Which custom model simulations? And please include references.

   Reply: The open-source products include the freely-available satellite observations and atmospheric reanalysis. The custom model simulations include the specific numerical simulation for SI event. They are already listed by citations in the previous sentences (see reply to Technical Comment # 2). In the revision, we re-wrote the sentences and deleted some redundant description. Lines 50-56.

4. **Line 56:** Could you name a few chemical tracers?

   Reply: Thank you for this suggestion. In fact, we listed two chemical tracers (isotopic sulfur and O_3-CO ratio) in the following sentences. Lines 55-58.

5. **Lines 58-60:** For redibility, I would use "a study using isotopic sulfur (^{35}S)" and "a study using O_3-CO"

   Reply: Thank you for this suggestion. We re-wrote the sentence as your suggestion. Lines 60-62.

6. **1:** The description in Lines 117-131 reads well. However, it is difficult to find the described details in the figure without zooming in. Could you improve Fig.1, for example, by increasing the font size, highlighting the contours for vorticity, and using a different colour for the wind direction? A good figure should be readable at 100% page view.

Reply: A good suggestion!!! We adjusted the Figure 1 in the revision. We adopted a 4×2 subplot matrix to replace the initial 2×4 matrix. The 4×2 matrix allows for better tracking the westward movement of upper-level trough and synoptic change across time. In the new figure, we characterize the geopotential height and upper-level trough by shading, with trough axis highlighted by red dot lines. Only wind direction of jet exceeding 20 m s^{-1} was shown by green arrows. The yellow contours were used to highlight the potential vorticity of 1.5 PVU at 400 hPa.

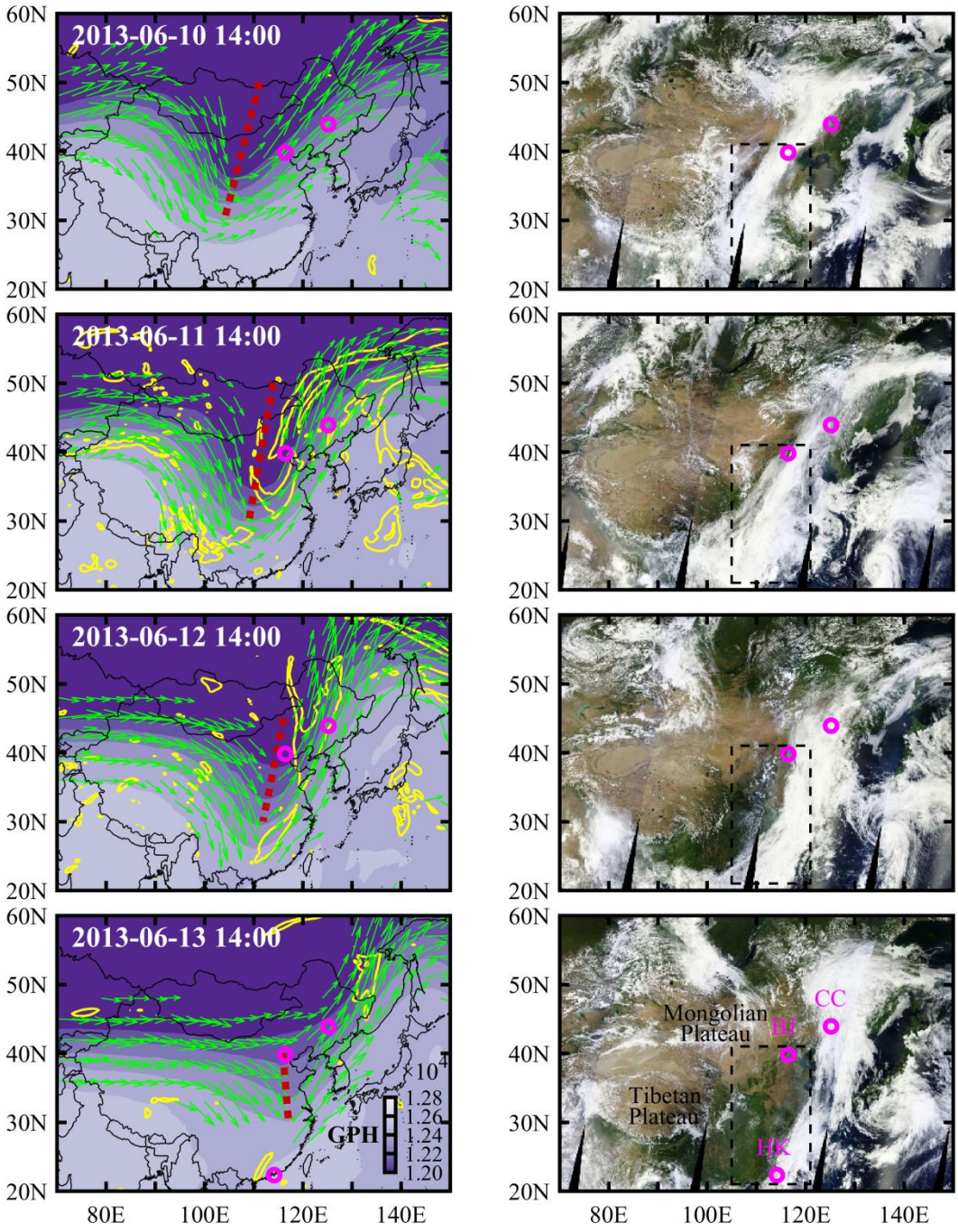

Fig. 1. (A) Horizontal distribution of geopotential height (shading, units in gpm), and wind direction of jet exceeding 20 m s^{-1} (arrows) at 200 hPa, and potential vorticity of 1.5 PVU (yellow contours) at 400 hPa. (B) MODIS satellite cloud images with the dashed box marking eastern China (105 ℮–

121 °E, 21 °N–41 °N). Red dot lines in (A) denote the axis of upper-level trough at 200 hPa. Magenta circles mark the available ozonesondes at different sites (BJ: Beijing, CC: Changchun, and HK: Hong Kong) on different days.

7. **2:** Could you make the font size and legend larger? Please put the location on the left size of each row. The subplot y-axis title (and scale) at the same row in Fig.2 could share the one because you share x-axis title at the same column. The label font is expected to be appropriately larger, so that the legend (how about a row at the top instead?) might more conducive for reading.

   Reply: A good suggestion! As you suggested, we re-plotted the Figure 2. Figure 4 and Figure 5 were also re-plotted.

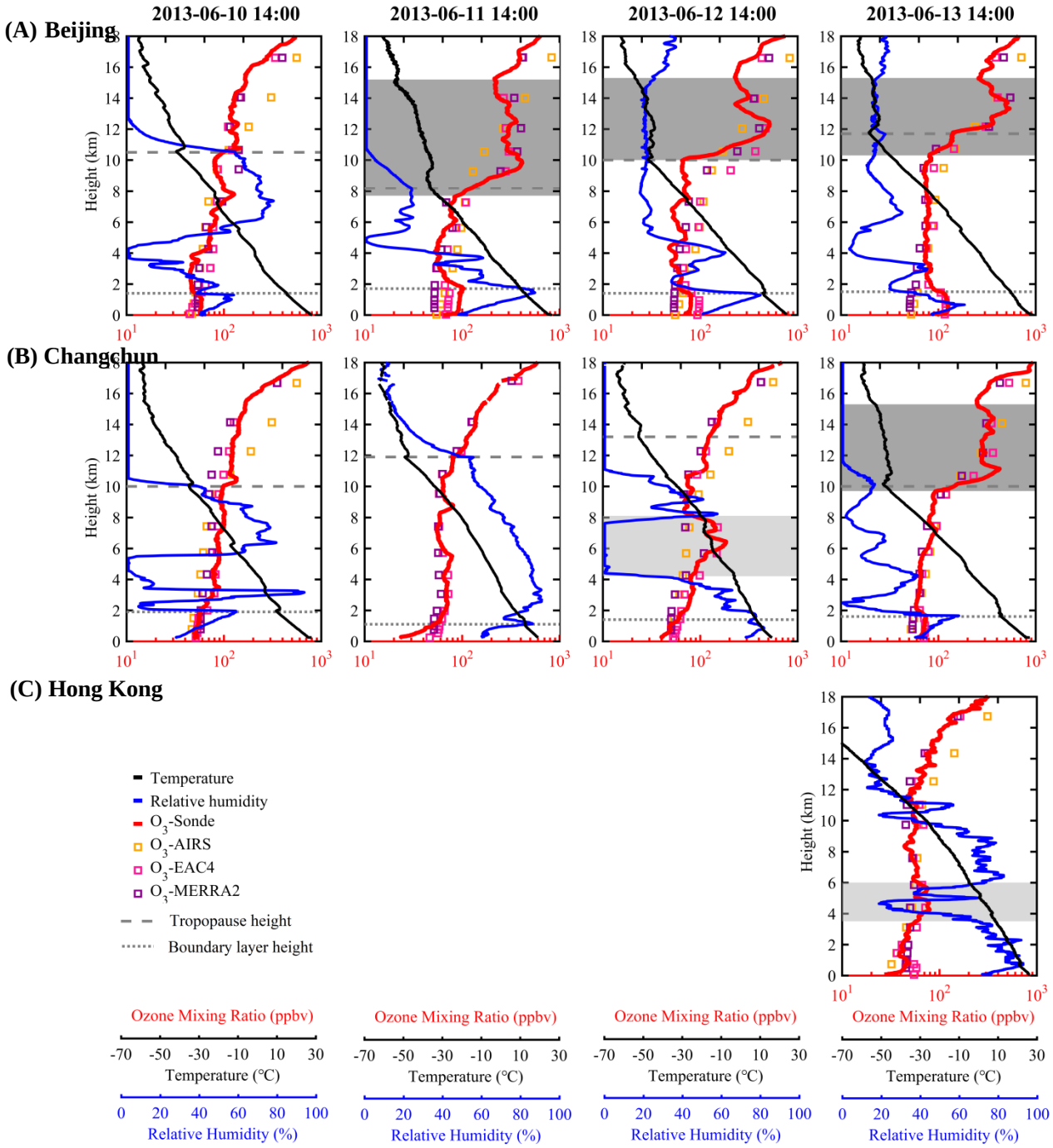

Fig. 2. O_3 vertical distribution over (A) Beijing, (B) Changchun, and (C) Hong Kong derived from ozonesonde and other data sources (including AIRS satellite observation, EAC4 and MERRA2 reanalysis) during 10–13 June 2013. Black and blue lines denote the sonde-based temperature (T) and relative humidity (RH) profiles, respectively. Gray dashed lines represent the thermal tropopause height, and gray dot lines indicate the boundary layer top height. Upper-level secondary O_3 peaks are shaded heavy gray, and SI-induced O_3-rich layer in the troposphere is shaded light gray.

8. **Line 152:** "… sonde-based O_3 profiles …" I noticed you used "ozone" instead of single "O_3" in this paper, shouldn't here the full name of "ozone" be used?

Reply: Thank you for picking it up. In the revision, "O_3" was used uniformly throughout the manuscript, except when it first appeared.

9. **Lines 196:** I am confused by the use of "filament" and "belt" here.

Reply: We re-wrote the sentences to avoid confusion. Lines 209-213: "The stratospheric intrusions developed into elongated (about 2000 km) and slender (about 200 km) streamers with elevated O_3 concentrations exceeding 150 ppbv (referred to as SDI-induced O_3-rich belts) at 400 hPa. On the east of the SDI streamers, the WCB streamers were parallel with anomalously low O_3 concentrations (referred to as WCB-related O_3-poor belts)". Besides, we labeled the SDI-induced O_3-rich belts and WCB-related O_3-poor belts in Fig. 4A.

10. **Lines 279-280:** Could you please label the Taihang Mountains and southern NCP on the figure?

Reply: Thank you for this suggestion. In this revision, we thought it was trivial to highlight the weak O_3S hotspots in the Taihang Mountains and southern NCP. Therefore, we deleted corresponding description in the revised manuscript.

11. **Line 313-314:** "Besides, ground-based stratospheric tracer method had been developed to quantify the stratospheric intrusion contribution over China." How to understand the "ground-based stratospheric tracer method"? "Ground-based" seems to be somewhat of an inconsistency with "" Was it mean ground-based validated stratospheric tracer data? According to the current information, you did not use ground-based data to validate the tracer but only the ozone. Here do need some explanations. Additionally, using "…to quantify the stratospheric intrusion contribution to surface ozone over China" is more explicit.

Reply: Thank you for this good suggestion. We replaced the "ground-based stratospheric tracer method" by "ground-based chemical tracer method" in the revised manuscript. We had introduced the term of "ground-based chemical tracers" in the Introduction. As you suggested in Technical Comment #4, we also listed some ground-based chemical tracers used in previous studies, e.g., cosmogenic sulfur-35 (^{35}S) and the ratio of O_3 to CO.