Responses

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We thank the reviewers for the constructive comments. The feedback has helped us to improve the exposition of the paper. We have carefully addressed all comments (detailed below). The reviewer comments are in black font. Our responses are shown in blue font and excerpts from the revised manuscript are in green font.

Reviewer2

The paper submitted by Cui et al. describe a data base of Source-Receptor-Relationships (SSR) corresponding to tropospheric ozone observations in Western North America. SSR retrieval is based on FLEXPART model V.10 following an approach similar to Cooper et al. 2010 but using more up to date meteorological analysis (ERA 5) and a better seasonal coverage. A data fusion technique has been implemented using the work of Chang et al. (2023) but applied to a more comprehensive ozone data base including new vertical levels and all observations available in Western North America (ozonesonde, lidar, aircraft).

The paper is a brief presentation of the SRR data base with some illustrations of its potential usefulness. More elaborated results will be described in future papers. The main results of the present paper are (i) a description of the FLEXPART model configuration in section 3, (ii) two illustrations of the spatial distributions of SRR for low or high ozone data set in section 4, (iii) while section 5 describes how SRR can be combined with source distributions of ozone precursor. In section 5 the example of the sensitivity to NOx tropical lightning source has been chosen.

I recommend publication of this paper as it is a useful addition to future papers using this SRR data base. However I believe that a short paragraph is missing in section 3 in order to describe differences between this new SSR and the Cooper et al. (2010) 1994-2008 spring time analysis. Such a new short paragraph would be a nice way to discuss the benefits of using ERA 5 and a more comprehensive ozone observation data set.

R2-1) Thanks. In the submitted manuscript, we included in Section 1 a brief explanation about the extension and differences compared with the previous Cooper et al., (2010): "[...] A similar SRR framework was used by Cooper et al. (2010) to explain increased FT ozone concentrations above WNA during April and May from 1995 to 2008. That study used an earlier version of the European Centre for Medium-Range Weather Forecasts (ECMWF) model with a 2° x 2° spatial resolution, generating SRRs up to 16 km above ground level. However, Cooper et al. (2010) focused exclusively on springtime. Chang et al. (2023) demonstrated that positive FT ozone trends over WNA are also present in summer and winter. Therefore, this study extends the analysis of Cooper et al. (2010) by simulating SRRs across all seasons over nearly three decades (1994–74 2021) using an updated version of the ECMWF model. [...]"

In section 3, we also mentioned the advancement of ERA5 data compared with others: "In an independent study, a cross-comparison among three widely used reanalysis datasets, including ERA5, was conducted by Wu et al. (2024). Their study indicated that, when compared to limited observations from field campaigns, the reanalysis datasets exhibited mean wind vector differences ranging from 2 to 4.5 $m \cdot s^{-1}$, with ERA5 showing the closest agreement with observations. Many other studies have evaluated ERA5 from different perspectives, consistently highlighting its strong performance. This further reinforces the reliability of our source-receptor database."

In response to the Reviewer's comment, we added a new paragraph near the end of Section 3 in the revised manuscript to better tie the discussion together:

"The implementation of FLEXPART and the resulting SRRs presented in this database are thus improvements over the previous methodology (Cooper et al., 2010) in three significant ways: i) higher top altitude and vertical layers chosen to discriminate among potential sources aloft, ii) updated meteorological input data at much finer spatial resolution (0.25° x 0.25°), and iii) expanded dataset of field observations, including all four seasons and considerably more ozone observations in the free troposphere. The resultant SRR dataset provides greater potential to conduct a more comprehensive and accurate investigation of ozone trends over a longer time period with enhanced spatiotemporal coverage."

Since long range transport analysis is often viewed as a key component to understand how stratosphere-troposphere exchanges (STE) control ozone increase in the free troposphere, the authors might consider an additional illustration to show how the new SRR could help to address this issue.

R2-2) This is an interesting point, and we discussed it among the author team multiple times while preparing the manuscript. Our ozone data were collected from 900 hPa to 300 hPa, and we have not yet identified an analysis approach which clearly focuses on stratosphere–troposphere exchange (STE). The overall highest 95th percentile of ozone values may be influenced by STE or by transport from the near-surface boundary layer, which requires further analysis. In general, 300 hPa is likely still below the altitude of the most active STE; therefore, the current SRR dataset may be insufficient for fully capturing these processes, but we can suggest that interested users could focus on the 95th percentile of ozone receptors at higher altitudes (e.g., 400–300 hPa) as shown in Figure 1, and select the corresponding SRRs to investigate potential STE processes.

Because it is clear that this topic is of interest to others, we added the related discussion in Section 5:

"In addition, our ozone receptors span from 900 hPa up to 300 hPa. The highest-altitude data (e.g., 400–300 hPa) shown in Figure 1, together with the corresponding SRR values, may help elucidate how stratosphere–troposphere exchange (STE) contributes to ozone increases in the free troposphere. However, 300 hPa is generally still below the altitude of the most active STE; therefore, the current SRR dataset may be insufficient for fully capturing these processes."

Detailed remarks

line 78. CTMs are indeed more complex tools but we can address different question related, especially the complex inetraction between dynamical and photochemical processes

R2-3) Agree. We have added the following sentence to our discussion section:

"We acknowledge that CTMs remain more comprehensive tools for interpreting ozone changes driven by the complex interaction between dynamical and photochemical processes."

line 130 Indeed the measurement error might be minor considering the large data set considered in this work, however Fig. 1 shows also a change an significant increase of data sampling after 2004. I wonder how this feature will be accounted for in the SRR data analysis.

R2-4) That is a good point. Indeed, after 2000, and especially after 2004, we have more ozone observations compared to earlier periods. In our SRR data analysis, we have mentioned that "SRRs are aggregated monthly across various altitudes for cases when ozone values at the receptors are at their low and high percentiles, compared to those over the mid-year period (2004–2014). A more detailed statistical framework is outlined in Ryoo et al. (in preparation) to minimize the influence of varying numbers of receptors across months and years. All subsequent SRR illustrations given here are generated using the same algorithm applied in Figure 3." This type of analysis represents post-processing. Our SRR dataset retains its full level of detail, which supports users to manipulate the data according to their specific needs and to apply different statistical approaches.

line 236 Fig. 2 only show that long range transport change when considering different altitude levels in WNA. Such a very genral picture is difficult to discuss detailed questions as the sensitivity to aircraft emission, biomass burning or STE.

R2-5) We agree that Figure 2 provides a general illustration of the dataset. In Section 5, Discussion and Additional Applications, Figure 5 demonstrates a case where we used our SRR framework to assess the sensitivity of lightning NOx to the ozone receptor space. Ryoo et al. (in preparation) have also applied SRR extensively to investigate the impact of aircraft emissions. In our current manuscript, also in Section 5, we mentioned that "IAGOS profiles (Section 2) have included CO measurements since 2001, and AJAX missions (Section 2) have collocated methane (CH_4) observations."

Thus, in the revised manuscript, we added the following sentences:

"Therefore, the SRR database can be applied to attribute sources contributing to FT CO and CH_4 trends over WNA, as well as to assess the sensitivity of FT ozone over WNA to biomass burning (using CO as a tracer)."

Thank you for the suggestion to include these additional specific possibilities.

line 250 Fig. 3 Is indeed a good illustration to describe the potential of the new data base.

R2-6) Thanks.

Fig. 4: I believe Fig. 4 shows the seasonal sensitivity for the whole tropospheric column, this should be clarified

R2-7) Thanks, yes, in the (submitted) manuscript we have explained (original line 268-269): "Figure 4 shows an example of aggregated analysis of the seasonal patterns at all altitudes for the entire 28-year period."

We slightly modified it to be “[…] all altitudes (surface to 20km) […].”in the paragraph below Figure 3 (current line number 292).

Fig. 5: I am not sure how to interpret the trend in the sum of the residence time. More residence time in the 3-13 km vertical range during the 21th century for the air masses with high ozone ?

R2-8) Thank you for your comments. A slight increase in those SRRs is evident within the 3–13 km layer during the later period (e.g., P2: 2007–2021), indicating that air parcels contributing to high ozone in WNA (e.g., ozone amounts in the 66th–95th percentiles) spent more time at 3–13 km in later years compared to earlier years. Regional emissions at 3–13 km, such as lightning NOx and aircraft, could be potential contributing factors to ozone formation. To improve the clarity of our description we added the following sentence to the revised manuscript, after introducing Figure 5a:

“The positive slope indicates that air parcels which contained ozone amounts in the 66-95th percentiles spent more time at 3-13 km in later years than did equivalent parcels in earlier years.”