We would like to thank the reviewers for their well-considered and constructive comments. We have considered the feedback at length and have revised the paper extensively based on the reviews. Original reviewer comments are in plain text and our responses are in bold italics below.

"Trace gas atmospheric rivers: remote drivers of air pollutions" by Mukesh Rai et al.

General comments:

This manuscript introduces a new tool to characterize the long-range transport (LRT) of trace gases such as O_3, CO, and PAN. The authors extend the concept of atmospheric rivers to define "the trace gas atmospheric rivers (TGARs)" and apply it to the TCR-2 (Tropospheric Chemistry Reanalysis) dataset for the period 2005–2019. They report over 300,000 global TGAR occurrences, claiming this account for up to 60% of total trace gas transport, with most events occurring in midlatitudes. While this concept is novel and has the potential to advance understanding of LRT, the manuscript raises several scientific and interpretational concerns:

Major Comments:

Air pollution levels are shaped by both emissions and meteorology-driven atmospheric transport. Emission trends differ by region and altitude - e.g., surface-level emissions may be decreasing while free-tropospheric concentrations are increasing. TGARs combine the effects of both emissions and atmospheric dynamics, making it difficult to disentangle which factor is driving the observed changes. This raises the question of whether TGARs are an adequate metric for characterizing long-range transport.

Thank you for this important observation. Compared to existing methods used to study long-range transport, the TGAR approach explicitly identifies and quantifies the number of extreme transport events over time (i.e., enhanced transport), enabling the attribution of these events to global trace gas transport. Thus, it could serve as an additional and valuable diagnostic tool for addressing long-term transboundary air pollution issues in air quality studies. To some extent, looking at multiple trace gas species can help with this. The impacts of emissions and transport are partially addressed by the Figure showing maps of trends in concentration alongside trends in IGT for the three different species. However, we concur with the reviewer that disentangling changes in emissions from changes in transport is still challenging.

In the revised manuscript, we now explicitly acknowledge in the conclusions that "Trajectory or tagged simulations would still be required to provide direct insights into air quality management."

TGARs appear closely tied to mid-latitude upper-level jet streams, which enhance wind speeds aloft. However, CO and other pollutants often peak near the surface. Simply multiplying wind speed by trace gas concentration may obscure rather than reveal emission source regions, as high wind regions don't necessarily indicate high recent emissions or source regions.

For example, upper-tropospheric (up to mid-tropospheric O_3, ex O_3 can be strongly influenced by stratosphere–troposphere exchange (STE), especially over the eastern Pacific and Atlantic, leading to variability unrelated to tropospheric anthropogenic sources.

We agree that TGAR, being closely aligned with mid-latitude jet streams, primarily highlight regions of efficient atmospheric transport rather than direct emission sources. It was not our intention to suggest that TGAR should be used to pinpoint emission locations. The intention is to identify regions where long-range transport may enhance pollutant concentrations.

We also acknowledge that upper- and mid-tropospheric O_3, particularly over the eastern Pacific and Atlantic, can be significantly influenced by stratosphere–troposphere exchange (STE), introducing variability that is not related to anthropogenic sources. In this study, we have used vertical integration from 1000 to 300 hPa specifically to help minimize the influence of stratospheric ozone on our results (see line 128-131, equation 1). Nonetheless, stratospheric influence cannot be ignored. We have included the following sentences in the revised introduction: "While the AR framework has been successfully applied to water vapor and aerosols, which predominantly reside in the lower troposphere, many trace gases have important sources both near the surface and in upper air. This vertical complexity can complicate the interpretation of AR based solely on vertically integrated horizontal fluxes. In particular, distinguishing between long-range horizontal transport and downward transport from the stratosphere remains challenging."

The manuscript interprets TGAR trends as emission-driven. For instance, the authors state: "For PAN, the strongest positive trends are observed over the North Atlantic and the Pacific..."

However, similar patterns appear for all trace gases due to their shared alignment with upper-level jets (e.g., North Atlantic and Pacific jet exit regions). These trends are likely to reflect meteorological variability, not emission changes.

We agree that the alignment of TGAR trends across multiple trace gases with upper-level jet structures - particularly over the North Atlantic and Pacific - suggests a strong influence of meteorological variability, rather than emissions alone. It was not our intention to interpret this as emissions-driven. In the revised manuscript, we have attempted to put more emphasis on the role of persistent atmospheric circulation patterns, such as jet stream variability. We have now included a figure that highlights the correlation between the NAO and PNA jet indices and the TGAR frequency in relevant regions. We now also include an explicit statement in the Conclusions section about impacts of changes in large-scale circulation patterns over time on pollution transport.

One major limitation of the TGAR framework is the inability to trace air parcels back to their source regions. Traditional LRT studies use back-trajectory or Lagrangian methods (e.g., footprint analysis or source-receptor-relationship) to identify upwind sources. TGARs, being derived from the vertical integrations of wind and trace gas fields, are sensitive to the height of maxima (e.g., aloft vs. surface) and background meteorology - making source attribution highly uncertain.

Thank you for highlighting this important limitation. We fully agree that, unlike traditional back-trajectory or Lagrangian approaches, the TGAR framework does not provide direct source-receptor linkages. TGARs are intended as a complementary diagnostic framework that highlights regions of enhanced pollutant transport, rather than to explicitly identify upwind sources. In the revised manuscript, we include the following statement in the Conclusions section: "The vertically integrated TGAR approach captures horizontal transport at large scales occurring at all levels in the troposphere. Long-range transport can influence surface air quality when downward transport occurs. However, actual surface-level impacts relevant to human exposure would still need to be evaluated by carefully analyzing local near-surface circulation alongside the TGAR long-range transport. Trajectory or tagged simulations would still be required to provide direct insights into air quality management."

Vertical structure of pollution and transport matters. Transport patterns differ significantly between the surface and free troposphere. For example, recent study shows western North America shows decreasing surface-level emissions and O_3, while free-tropospheric O_3 is increasing (Chang et al., 2023). TGARs based on vertical integration mask this distinction, potentially misrepresenting source–receptor relationships.

We agree that the vertical structure of pollution and transport is extremely important. We had already included a figure (Figure 6 in the original submission, now Figure 3 in the revised manuscript) that shows the relative contribution of the near-surface horizontal transport to the total tropospheric IGT.

We have also added the following statement to the discussion of the Figure that shows trends in tropospheric ozone concentrations: "We acknowledge here that while here we show trends based on a single value for tropospheric ozone, the vertical structure of trends in tropospheric ozone remains an active area of research. For example, it has been shown that between 1995 and 2021, western North America shows decreasing surface-level emissions and O_3***, while free-tropospheric*** O_3 ***is increasing (Chang et al., 2023)."***

TGAR trends may reflect changes in Atmospheric dynamics (e.g., jet strength, variability), anthropogenic emission patterns, stratospheric intrusion events (or stratospheric-tropospheric-exchange events), and photochemical processes during the transport. The TGAR approach makes it difficult to separate and interpret the contributing factors individually. As a result, the drivers of the observed trends remain unclear, which undermines the utility of TGARs for policy-relevant source attribution.

We acknowledge that TGAR trends reflect the combined influence of multiple factors, including atmospheric dynamics, emission patterns, stratospheric intrusions, and photochemical processes. As such, TGARs are not intended to isolate individual drivers but rather to highlight regions of enhanced transport. In the revised manuscript, have attempted to articulate this limitation more clearly in the Conclusions, and emphasize that TGARs are a complementary tool alongside more source-specific approaches when aiming for policy-relevant source attribution

A key goal in studying LRT is identifying which upwind countries or regions contribute to downwind air quality degradation. TGARs do not enable such attribution. Without this, the concept risks being uninformative for regulatory or policy applications.

While TGAR do not directly enable source attribution, they are intended as a complementary tool to highlight regions and times of enhanced transport potential. In the revised manuscript, we emphasize in the Conclusions that TGAR should be used in conjunction with trajectory-based approaches to support more policy-relevant source identification.

In summary, while the TGAR concept is creative, the manuscript lacks physical clarity, robust attributes, and a clear separation of dynamic vs. emission-driven trends. As currently framed, TGARs may obscure key processes and introduce interpretational challenges that outweigh their benefits. A more targeted analysis focusing on meteorology–composition interactions, or incorporating trajectory-based source attribution, would likely be more effective for advancing our understanding of LRT.

Thank you for the feedback. We hope that the revised version is more clear.

Suggested Alternatives and Improvements:

- Separate vertically resolved TGAR analysis (e.g., surface vs. mid- and upper troposphere) or clearly justify why a vertically integrated approach offers greater insight or benefit to understand LRT of air pollution.

We do include a figure (Figure 6 in the original submission, now Figure 3 in the revised manuscript) that shows the relative contribution of the near-surface horizontal transport to the total tropospheric IGT. We are not convinced of the value of a vertically resolved TGAR analysis, since long-range pollution transport pathways can involve vertical as well as horizontal motion along the way. For example, pollution from East Asia can be lofted by warm conveyer belts, undergo rapid transport across the Pacific in the free troposphere and then descend over the Western US. It is not clear how separating surface vs mid- and upper troposphere would be useful in capturing this sort of process.

• Correlate trace gas variability (CO, O_3) with jet stream indices or wind anomalies.

Thank you for this suggestion. We have now included a figure showing time series of TGAR frequency anomalies with NAO and PNA jet indices.

• Avoid multiplying trace gas concentrations by wind speed without identifying specific receptor regions and providing a clear physical justification for the resulting values and their interpretation.

It was not clear to us what is meant here. Further clarification from the reviewer could be helpful.

Minor comments:

Overall, the manuscript lacks sufficient references in both the introduction and main text. Key background literature related to long-range transport, meteorology–chemistry interactions, and similar analytical approaches should be cited to provide context and support for the study. Furthermore, the authors often include previous findings not to support their results, but as additional background information, which at times feels disjointed or unrelated to the main analysis. These descriptions would be more appropriately placed in the introduction.

We have extensively revised the introduction to include more key references to background literature and alternative approaches. We revised the structure more broadly and tried to ensure that background information is appropriately placed in the introduction, and that citations in the main text directly support the analysis and interpretation of our results.

The manuscript should clearly explain the practical value of ARs, TGARs, or AARs in the context of trace gas transport, and why this can be used as compelling metrics for LRT. ARs are well established for their role in extreme weather prediction, but it is unclear whether a similar framework is needed for pollutant transport. The meteorological influence on pollutant transport is already well known, and wind alone is not sufficient to explain pollutant behavior - particularly for secondary pollutants like O_3. Other factors such as relative humidity and precipitation, which are tightly coupled with AR dynamics, are also critical to consider.

We have attempted to better articulate the practical value of these metrics in identifying coherent, large-scale transport events that can significantly influence downwind pollutant levels. We also acknowledge that pollutant behavior, particularly for secondary species like O_3, is influenced by more than just wind - factors such as relative humidity, precipitation, and chemistry play important roles. In the revised Conclusions section, we have included language to convey the added value of the TGAR framework as a complementary-not standalone-tool for understanding long-range transport.

While the TGAR approach visualizes column-integrated trace gas concentrations, it does not resolve where pollutants originate. This leaves questions of source attribution unanswered. Which countries or regions are responsible for the observed elevated concentrations of CO, O_3, and PAN? Without this clarity, the utility of TGARs in informing policy or mitigation strategies remains limited.

We agree that TGAR, as a column-integrated diagnostic, do not provide direct source attribution and therefore cannot identify specific countries or regions responsible for elevated pollutant levels. In the revised manuscript, we acknowledge this constraint and emphasize that TGAR are intended to complement rather than replace-traditional source attribution methods such as back-trajectory analysis. We also now clarify that while TGAR help identify regions and time periods of enhanced long-range transport, they must be used in conjunction with source-resolving tools to inform policy or mitigation strategies effectively.

Strong wind is often associated with lower trace gas concentrations due to dispersion. Vertical integration of concentration does not distinguish between surface-level and free-tropospheric contributions, making it difficult to assess actual surface-level impacts relevant to human exposure or regulatory control.

We agree that strong winds can lead to dispersion and dilution of trace gases, and that vertically integrated concentrations may obscure distinctions between surface-level and free-tropospheric contributions. In the revised manuscript, we include the following statement in the Conclusions: "The vertically integrated TGAR approach captures horizontal transport at large scales occurring at all levels in the troposphere. Long-range transport can influence surface air quality when downward transport occurs. However, actual surface-level impacts relevant to human exposure would still need to be evaluated by carefully analyzing local near-surface circulation alongside the TGAR long-range transport."

Figures - in the current form, many of the figures are difficult to interpret. In particular, the maps are not clearly visible, and wind vectors are overly dense, which reduces readability and visual effectiveness.

Thank you for your feedback regarding the figures. In the revised manuscript, have improved the quality and visibility of the maps. We have also reduced the density of wind vectors to avoid visual disorder and enhance interpretability.

The introduction frames long-range transport as a hemispheric phenomenon, but the body of the manuscript focuses primarily on midlatitude transport. This inconsistency should be clarified. If the study targets midlatitude dynamics, the framing and scope should be adjusted accordingly.

Thank you for this feedback. The TGAR approach is intended to provide "an objective and globally applicable approach to quantify long-term variations in the frequency, intensity, and spatial characteristics of extreme transport events". We now include this statement upfront in

the abstract and we have extensively revised the introduction based on comments from both reviewers.

The TGAR framework does not account for ozone mixing or stratospheric intrusions, which can play a major role in upper-tropospheric O_3 variability. This limitation should be acknowledged, and the authors should discuss how TGARs differ from or complement known STE processes. Please clarify.

We agree that the TGAR framework does not explicitly account for ozone mixing or stratosphere-troposphere exchange (STE), both of which can significantly influence upper-tropospheric O_3 variability. We have acknowledged in the paper that "in certain cases, these analyses may be complicated by contributions from stratospheric inflows." TGAR analysis of multiple species can help to distinguish between the role of stratospheric intrusions (which will increase tropospheric O_3, but will not increase CO or PAN) and horizontal transport.

In the detection method, the criterion requiring directionality to be less than 45 degrees needs justification. Why was this threshold chosen, and how does it impact the identification of TGAR events?

Thank you for pointing this out. We now include the following statement: "The 45-degree directionality threshold was chosen to ensure that the identified TGAR events represent coherent, directionally consistent transport patterns, which are characteristic of atmospheric river-like structures."

The reported number of TGAR events (~300,000) seems heavily dependent on the temporal resolution of the input data (0, 6, 12, 18 UTC). While the fact that TGARs occur "frequently" is meaningful, the actual number/count alone does not provide much insight, in my opinion. This could vary significantly with hourly or daily datasets. A more robust validation would involve case studies, e.g., identifying high pollution episodes and evaluating the role and frequency of TGARs during those events. This would ground the TGAR framework in real-world air quality applications and improve its interpretability.

This is a fair comment. We agree that the reported number of TGAR events is sensitive to the temporal resolution of the input data, and that the count alone may not fully convey the relevance of these events. The revised manuscript now puts less emphasis on the absolute number of events. We now include a subsection with a case study that links a TGAR event to a documented high-pollution episode.

The abstract currently lacks clear takeaway messages, and the overall benefit of applying the TGAR concept to LRT analysis remains vague. The authors should clearly state what new insight TGAR provides and how it improves upon existing approaches in LRT characterization.

We have rewritten the abstract in light of comments from both reviewers.

Reference

Chang, K.-L., Cooper, O. R., Rodriguez, G., Iraci, L. T., Yates, E. L., Johnson, M. S., et al. (2023). Diverging ozone trends above western North America: Boundary layer decreases versus free tropospheric increases. Journal of Geophysical Research: Atmospheres, 128, e2022JD038090. https://doi.org/10.1029/2022JD038090