

Dear Editors and Reviewers,

Thank you very much for your careful review on our manuscript egosphere-2024-930. We appreciate very much your encouraging comments and constructive suggestions on improving our manuscript. We have accordingly made the careful and substantial revisions. The revised portions are marked up in the revised manuscript. Please find our point to point responses to the reviewers' comments as follows:

Responses to the reviewer #2

[This study investigates the impacts of stratospheric ozone intrusion on tropospheric ozone in central and eastern China, using air quality observations, global ozone (re)analysis data, and customized Lagrangian simulations. The results are interesting and the manuscript is well organized in general, however, there are still some problems need to be improved before a consideration for publication.]

Response 1: Many thanks for the encouraging comments and helpful suggestions on our manuscript. Following the reviewer's suggestions and comments, we have accordingly made careful revisions. Please find our point-to-point responses as follows:

[Abstract L16: SIs]

Response 2: Thank you for pointing out the printing errors. It has been corrected to SIs in the revised manuscript.

[Data: Why the former ERA-Interim data was used in this study, instead of the more advanced ERA5 or MERRA2?]

Response 3: We appreciate the reviewer's suggestions. In the revised manuscript (Lines 117-120), we have added accordingly the following discussions:

In this study, ERA data are solely used for large-scale atmospheric circulation analysis, as detailed in Section 3.5. Stratospheric intrusions are primarily controlled by large-scale circulation, and our analysis using ERA-Interim data at 0.75-degree resolution effectively addresses the scientific issue.

Nevertheless, future studies could utilize more advanced ERA5 or MERRA2 data to explore more scientific insights.

[Model configuration: Why the year 2019 was chosen to simulate? As I know, the SI's contribution to tropospheric ozone in China is significantly larger than normal years. Anyway, to discuss the SI's contribution with simulations only in one year will lead to large uncertainties due to the interannual differences.]

Response 4: Thanks for the reviewer's comments. In response to the comments, we have accordingly added the following discussions in the revised manuscript:

The year 2019 was chosen in this study as a sequel of our previous studies (Meng et.al. 2022a, b) on the influence of stratosphere-to-troposphere transport on summertime surface O₃ changes in 2019 and a typhoon-induced SI event in North China Plain in a series of research (Lines 102-105).

This study only in one year could lead to uncertainties due to the interannual differences. Further study with multi-years global simulations combining with diverse observation data could comprehensively understand the impacts of stratospheric intrusions on tropospheric ozone in China, encompassing multi-year climate, interannual and seasonal variations.

By the way, we have modified the title to "Tracing the origins of stratospheric ozone intrusions: direct vs. indirect pathways and their impacts on Central and Eastern China in spring-summer 2019".

References

- Meng, K., Zhao, T., Xu, X., Hu, Y., Zhao, Y., Zhang, L., Pang, Y., Ma, X., Bai, Y., Zhao, Y., and Zhen, S.: Anomalous surface O₃ changes in North China Plain during the northwestward movement of a landing typhoon, *Sci. Total Environ.*, 820, 153196, <https://doi.org/10.1016/j.scitotenv.2022.153196>, 2022a.
- Meng, K., Zhao, T., Xu, X., Zhang, Z., Bai, Y., Hu, Y., Zhao, Y., Zhang, X., and Xin, Y.: Influence of stratosphere-to-troposphere transport on summertime surface O₃ changes in North China Plain in 2019, *Atmos. Res.*, 276, 106271, <https://doi.org/10.1016/j.atmosres.2022.106271>, 2022b.

[It is better to describe briefly about the domain-filling technology here.]

Response 5: Thanks for the reviewer's suggestions; we have supplemented a detailed description of the domain-filling technique in Section 2.2 in the revised manuscript (Lines 143-150) as below:

In our FLEXPART simulation, we employed domain-filling technology (Chen et al., 2012; Drumond et al., 2010) to simulate the stratospheric ozone tracer, ensuring that only air masses released from the stratosphere

were considered. During the initial stages of simulation, particles in the troposphere were filtered out, while stratospheric particles were assigned mass according to the formula: $M_{O_3} = M_{air} \cdot P \cdot C \cdot 48/29$, where M_{air} denotes the air mass of the particle, P is the potential vorticity, $C=60 \times 10^{-9}$ pvu⁻¹ is the ozone/PV scaling factor (Stohl et al., 2000, 2005), and the factor 48/29 is applied to convert volume to mass mixing ratio. Throughout the integration, particles were advected from the stratosphere into the troposphere driven by GFS wind fields.

References

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmospheric Chemistry & Physics, 5, 2461-2474, 2005.

Stohl, A., Spichtinger-Rakowsky, N., Bonasoni, P., Feldmann, H., Memmesheimer, M., Scheel, H. E., Trickl, T., Hübener, S., Ringer, W., and Mandl, M.: The influence of stratospheric intrusions on alpine ozone concentrations, Atmos. Environ., 34, 1323-1354, [https://doi.org/10.1016/S1352-2310\(99\)00320-9](https://doi.org/10.1016/S1352-2310(99)00320-9), 2000.

[Figure 1: The colormap used in this figure should be updated to make the signals on a-b and k-m clearer.]

Response 6: We have replaced the colormap in Figure 1.

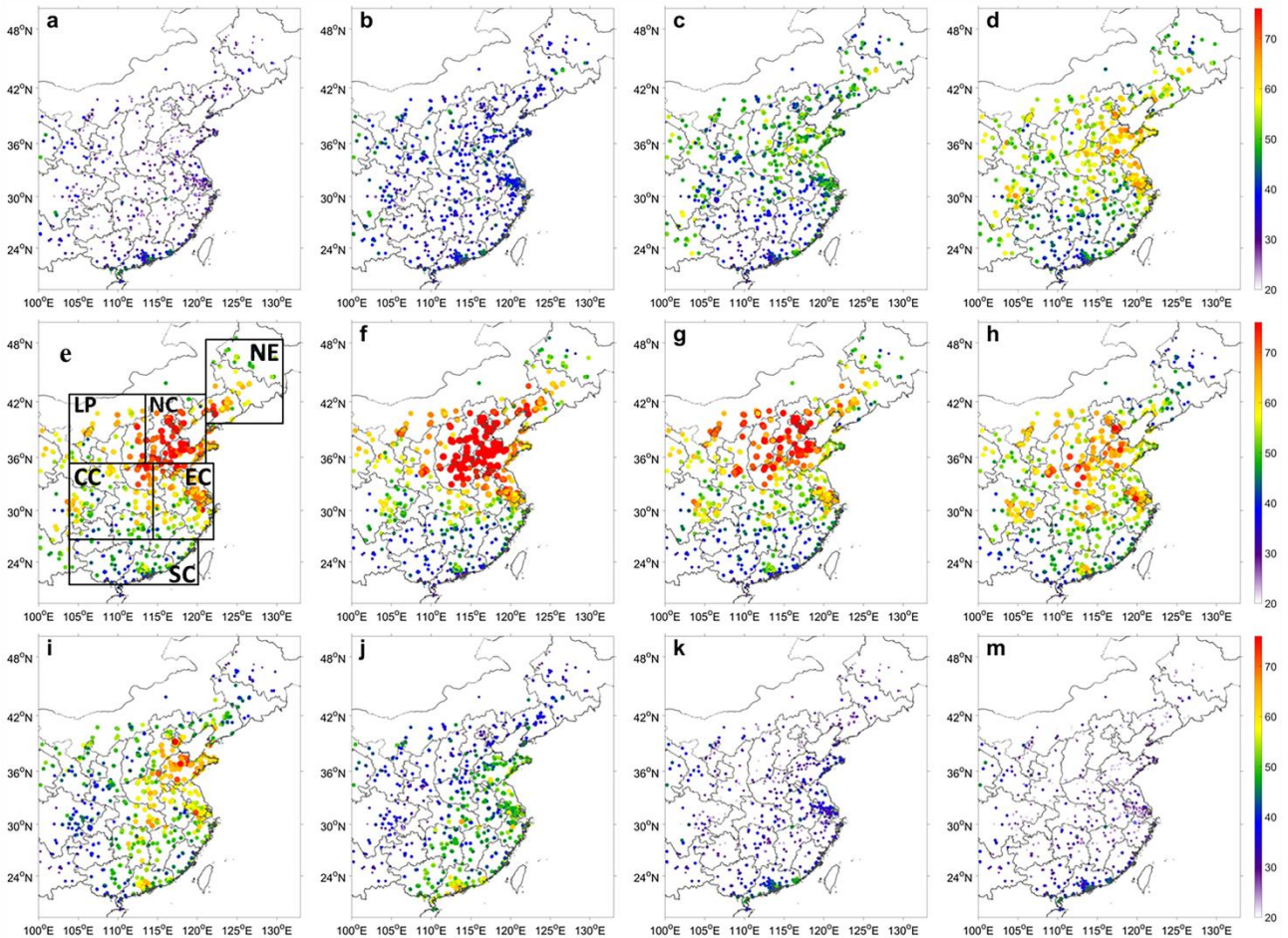


Figure 1: Distributions of the observed near-surface O₃ concentrations (ppbv) over CEC for (a-l) January to December during 2015-2019 with the sub-regions NE (NorthEast China); NC (North China); LP (Loess Plateau); CC (Central China); EC (East China); SC (Southern China).

[Figure 2: The vertical ozone gradients also represent the position of the tropopause. Seen from this figure, the 1.5 PVU may not a proper choice for all the selected regions.]

Response 7: Thank you for your detailed comments on our manuscript. We agree with the reviewer’s opinion that tropopause height varies significantly with latitudes and seasons. Indeed, a fixed threshold of 1.5 PVU may not sufficiently resolve the STE processes across over such a broad region in China. Therefore, based on the tropopause distribution characteristics outlined in Kunz et al. (2006) and Chen et al. (1995), we have introduced latitude-variable potential vorticity (PV) thresholds with 1.5 PVU north of 30°N and 3.0 PVU south of 30°N to define the dynamical tropopause and assesses the stratospheric origin of air masses over the study region in China in the revised manuscript. This updated approach confines ozone exclusively to stratospheric origins, crossing the tropopause into the troposphere. We have modified Section 2 “Data and

Methods” in the revised manuscript (Lines 143-176) to introduce this updated approach with latitude-variable potential vorticity (PV) thresholds as follows:

2.2 Model configuration

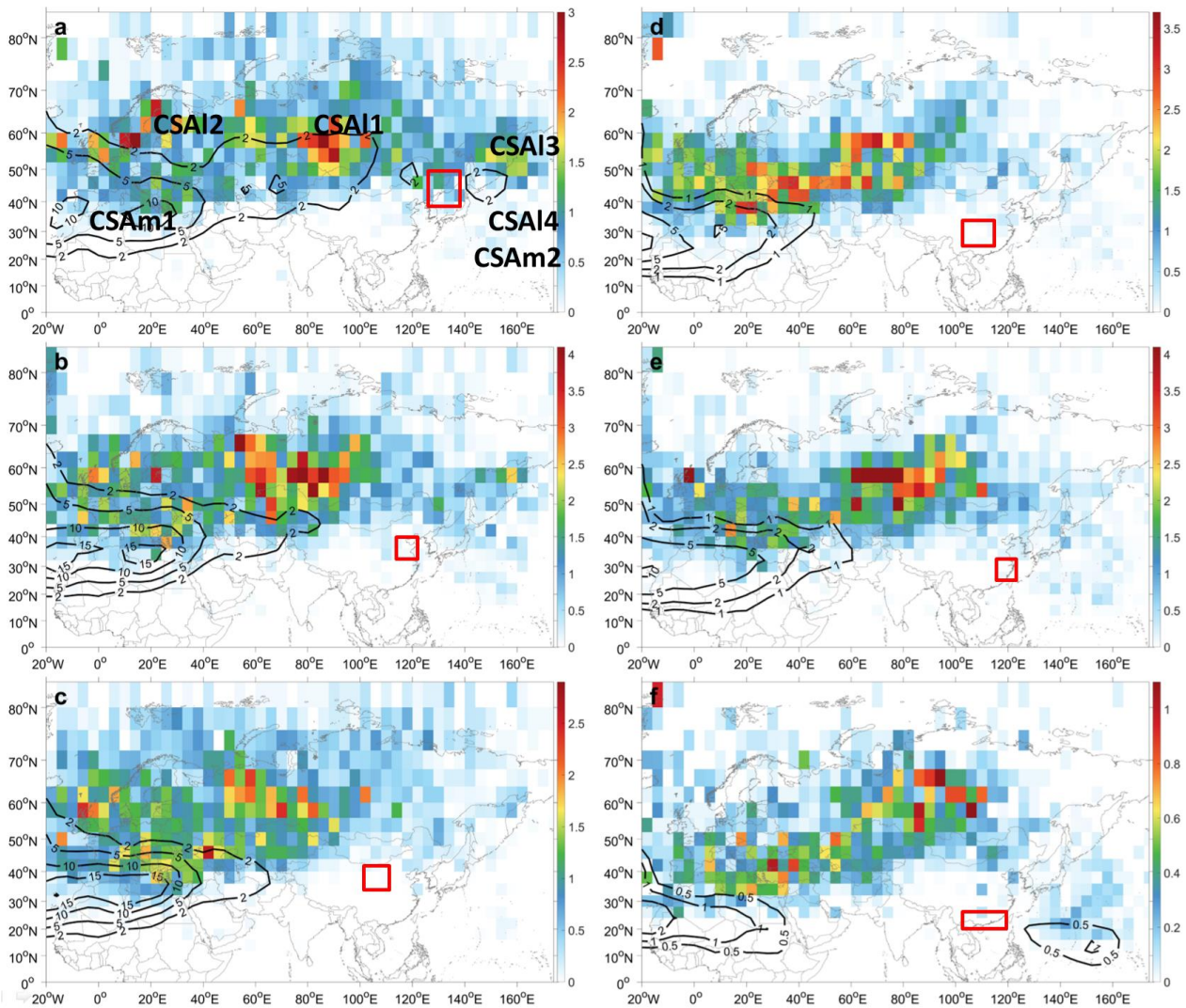
We conducted a daily rolling simulation with 15-day forward trajectories of over 500,000 air particles released from the stratosphere over the Eurasian region (-20°W–180°E) between May 1 and August 31, 2019. In our FLEXPART simulation, we employed domain-filling technology (Chen et al., 2012; Drumond et al., 2010) to simulate the stratospheric ozone tracer, ensuring that only air masses released from the stratosphere were considered. During the initial stages of simulation, particles in the troposphere were filtered out, while stratospheric particles were assigned mass according to the formula: $M_{(O_3)} = M_{air} \cdot P \cdot C \cdot 48/29$, where M_{air} denotes the air mass of the particle, P is the potential vorticity, $C = 60 \times 10^{-9} \text{ pvu}^{-1}$ is the ozone/PV scaling factor (Stohl et al., 2000, 2005), and the factor 48/29 is applied to convert volume to mass mixing ratio. Throughout the integration, particles were advected from the stratosphere into the troposphere driven by GFS wind fields. In mid-latitudes, the potential vorticity (PV) is commonly regarded as an indicator of the dynamical tropopause (Bellevue et al., 2007). PV values ranging from 1.0 or 1.6 PVU (Stohl et al., 2000) to 3.5 PVU (Hoerling et al., 1991) are typically used (Akritidis et al., 2016, 2018, 2021; Hoskins and Berrisford, 1988; Skerlak et al., 2014; Sprenger et al., 2003; Stohl et al., 2003). Gerasopoulos et al. (2006), for instance, used a PV greater than 1.5 PVU in backward trajectories to track air masses of stratospheric origin. In this study, we defined the dynamic tropopause as a PV of 1.5 PVU and stratospheric air as a PV greater than 1.5 PVU. The model output included three-dimensional ozone concentrations and position information of particles at 3-hour intervals, a horizontal resolution of $0.3^\circ \times 0.3^\circ$, and 17 vertical layers spaced 500 m apart.

2.3 CSA identification and CSA impact index

We employed the three-dimensional trajectory information obtained from the FLEXPART simulation to identify particles that reached the middle and lower troposphere within each sub-region of CEC, either directly or indirectly from the stratosphere. Unique IDs were then assigned to these trajectories/particles, enabling seamless tracking during subsequent analyses. By assimilating the meteorological field with the trajectory information and applying the distance weighting method, we determined the spatial location of each trajectory as it crossed the tropopause. The PV values, interpolated from meteorological reanalysis

data, were used to determine the dynamical tropopause. Specifically, we adopted latitude-dependent PV thresholds with 1.5 PVU north of 30°N and 3.0 PVU south of 30°N to define the dynamical tropopause and assesses the stratospheric origin of air masses over the study regions in China. This approach constrained ozone to originate exclusively from the stratosphere, crossing the tropopause into the troposphere. This methodology enabled us to identify the critical source areas (CSAs) of SI, which are the zones where stratospheric air masses penetrate into the troposphere by crossing the tropopause. They are characterized by a high proportion of gridded particles/trajectories information within these spatial locations.

Following the reviewer's suggestion, sensitivity tests of with different PV thresholds to track stratospheric origins have been conducted performed in order to obtain reliable results over China, newly proposing the latitude-dependent PV thresholds for this study (New Figure 7). Compared to the original 1.5-PVU threshold, the latitude-dependent PV thresholds present reasonable performance with slight variations in the CSAII index over the sub-region of South China (Fig. 7f), influenced by the SI source above the northwest Pacific. The latitude-variable PV thresholds (New Figure 7) show less deviation from the 1.5-PVU threshold over otherthe rest sub-regions in CEC, confirming the robustness of our simulations and CSAII index algorithm.



New Figure 7: Distributions of CSAII for ISI intruding the lower troposphere (color shaded) and middle troposphere (black lines) of over the regions a) NE, b) NC, c) LP, d) CC, e) EC and f) SC from May to August in 2019. Each sub-region's position has been indicated with the red square.

References:

Chen P .Isentropic cross-tropopause mass exchange in the extratropics[J].Journal of Geophysical Research Atmospheres, 1995, 1001(D8).DOI:10.1029/95JD01264.

Kunz A , Konopka P ,R. Müller,et al.Dynamical tropopause based on isentropic potential vorticity gradients[J].Journal of Geophysical Research Atmospheres, 2011, 116(D1):- .DOI:10.1029/2010JD014343.

[3.2: Why the 2 and 10 days forward trajectories can indicate the direct and indirect transports? The reason should be well explained and the sensitivity of the results to the selected length should be tested.]

Response 8: In response to the reviewer's suggestion, we have clarified our definitions of direct and indirect stratospheric intrusions. Previous studies (Eisele et al.; 1999; Meng et al., 2022b; and Zanis et al., 1999) proposed that stratospheric air can be transported to the lower troposphere directly through rapid vertical movement (lasting less than 2 days in the troposphere) or indirectly from thousands of kilometers away through a phased tilted mode (resulting in longer tropospheric residence times, such as > 10 days). We have expanded upon these concepts to distinguish between direct and indirect stratospheric intrusions, where direct intrusions involve stratospheric air from specific sources reaching mid or lower troposphere regions within approximately two days, while indirect intrusions extend this period to around ten days. Notably, both mid and lower tropospheric regions are considered endpoints for stratospheric air transport in our definitions, with transit times set at two and ten days for these regions, respectively.

In the revised manuscript (Lines 214-221), we have added the above sentences.

References

- Eisele, H., Scheel, H. E., Sladkovic, R., and Trickl, T.: High-Resolution Lidar Measurements of Stratosphere–Troposphere Exchange, *Journal of the Atmospheric Sciences*, 56, 319-330, 10.1175/1520-0469(1999)056<0319:HRLMOS>2.0.CO;2, 1999.
- Meng, K., Zhao, T., Xu, X., Zhang, Z., Bai, Y., Hu, Y., Zhao, Y., Zhang, X., and Xin, Y.: Influence of stratosphere-to-troposphere transport on summertime surface O₃ changes in North China Plain in 2019, *Atmos. Res.*, 276, 106271, <https://doi.org/10.1016/j.atmosres.2022.106271>, 2022b.
- Zanis, P., SCHUEPBACH, E., GÄGGELER, H. W., HÜBENER, S., and TOBLER, L.: Factors controlling beryllium-7 at Jungfraujoch in Switzerland, *Tellus B*, 51, 789-805, <https://doi.org/10.1034/j.1600-0889.1999.t01-3-00004.x>, 1999.

[Figure 3 and 4: How the contributions of stratospheric ozone directly/indirectly transported to the troposphere were estimated? As an important result of the paper, detailed description of the methods should be clearly described.]

Response 9: Thanks for the reviewer's suggestions. The contributions presented in this study originate from ozone concentration calculations at various altitudes within the troposphere obtained from our FLEXPART simulation. The calculation of concentration within each model grid is achieved by sampling tracer mass fractions of all particles within the grid cell and dividing by the grid cell volume:

$$C = 1/V \cdot \sum_{i=1}^N (m_i f_i)$$

Here, V represents the volume of the grid cell, m_i denotes the mass of particle i , N is the total number of particles, and f_i represents the mass fraction contributed by particle i to the respective grid cell. The mass fraction f_i is computed using a uniform kernel with grid distances Δx and Δy in the longitude-latitude output grid. We have incorporated the above methodology description into the revised manuscript (Lines 157-163).

Assessing stratospheric intrusions and their transport poses inherent challenges. In the revised manuscript, we validate the reliability of our FLEXPART simulations by comparing SI ozone and TOST ozone concentrations averaged monthly across the mid and lower troposphere layers. TOST ozone data are derived from global ozonesonde observations and Lagrangian models, as evaluated for reliability in Liu et al. (2013a). Concentrations of TOST and SI ozone in each sub-region are detailed in Table S1 in the revised supplement. SI ozone concentrations in each layer of each sub-region are notably lower than TOST values but exhibit similar temporal and regional trends. These comparative findings underscore the credibility of our simulation methodology.

[Figure 7: It is better to mark the location of each region.]

Response 10: As requested by the reviewer, the position of each sub-region has been marked in the new Figure 7 (please see response 7).

[P12 L305: Is the CSAm1 related to the southern Asia High or collocated with the subtropical jet?]

Response 11: We appreciate the reviewer's discussion. As depicted in Figure 9, CSAm1 corresponds to the subtropical jet over Europe and North Africa. Accordingly, we have updated the following description at lines 404-406: "it is evident that the CSAs and CSAm1s of ISI are intricately linked with the pattern of atmospheric fluctuations and are located within the subtropical jet in the UTLS."

[Figure 9: The colormap should be improved for this figure to make the information more visible.]

Response 12: Thanks for your careful review. We have improved Figure 9 to enhance the visibility of the information.

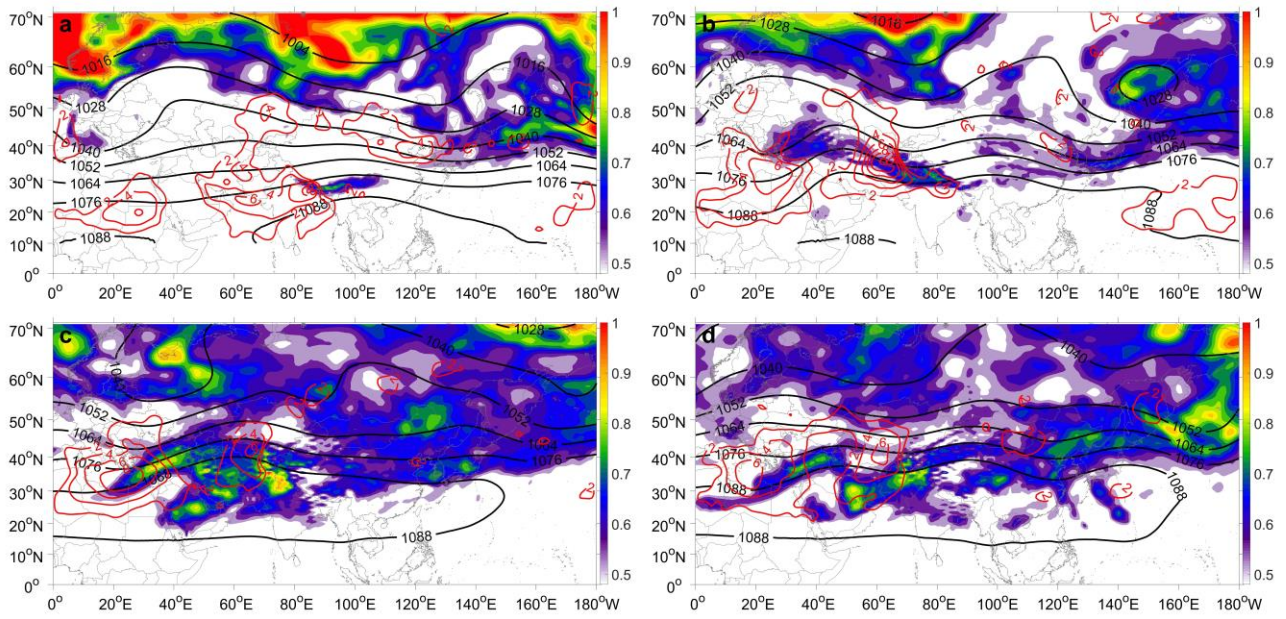


Figure 9: 400 hPa PV (PVU, color contours), geopotential height (gpm, black lines), and vertical velocity (>0 hPa s⁻¹, red lines) averaged at heights of 150–400 hPa in May (a), June (b), July (c) and August (d) in 2019.