This study compared the radiative effects of anthropogenic aerosols between western hemisphere and eastern hemisphere in the recent decades and their impacts on the atmospheric circulation. It is an interesting topic, as the interhemispheric contrast of anthropogenic aerosols were often focused in previous studies but the eastern-western hemispheric contrast of anthropogenic aerosols gains much less attention despite this may be the dominant change of anthropogenic aerosols in the NH during the last four decades.

Response:

Thanks for the precious summary of the key point of our study, which emphasizes on the zonal contrast of aerosol forcings.

The analysis is solid, but some descriptions are not very accurate and some mechanisms are still unclear. Moderate revisions are needed to address these concerns.

Response:

Thanks for the valuable feedback. We further clarified our discussions in the manuscript and have included more dynamical analysis to the manuscript in this round of revision:

 The cross-equatorial atmospheric energy transport due to radiative forcing occurring at mid-to-high latitudes leads to the Hadley Cell shifting northward to balance the interhemispheric energy imbalance (Fig 7).

- Both atmospheric and oceanic meridional energy transport over the North Atlantic contributes to the remote warming over the North Pacific (Fig 10).
- Analysis of the cloud droplet concentration to show the response to aerosol indirect forcings (Fig 4).
- The additivity and sensitivity test of the two sets of regional forcings experiments (EastFF and WestFF). (Fig. 3)

Please check the detailed responses to the specific comments below.

Detailed comments:

1. Page 2 L23-26: I don't think you can say the SAT gradient can be used as a predictive metric of NH jet changes, as they are just the same thing based on the thermal wind relationship. Both of them are forced by other factors.

Response:

Thanks for the correction. By introducing the "rule-of-thumb" indicator, we argue that the latitudinal gradient of FSNTOA, not SAT gradient, a measurable quantity, can be considered as an indicator of the circulation shifts in NH.

By showing the diagnostic geostrophic winds and the corresponding SAT gradient in Figure 7 (now Figure 8), we just want to explain that the jet stream shifts are largely consistent with the simulated SAT gradient.

We modify the text in the abstract as follow:

"This leads to a counter-clockwise anomaly of zonal mean stream function over the tropics (i.e. a northward shift of Hadley cell) and stronger equatorward shift of the Northern Hemisphere (NH) jet stream, consistent with the thermal wind argument of surface air temperature (SAT) gradient. Furthermore, the consistent relationship between the jet stream shift and the Top-of-Atmosphere net solar flux (FSNTOA) gradient suggests that the latter can be considered as a rule-of-thumb indicator."

2. Page 2 L28-29: First, this sentence seems problematic; Second, when you say "dominating role of WH forcing", which aspects do you point to? Based on your results, at least for the jet shift, it is more caused by EH forcing.

Response:

Thanks for the suggestion. We want to emphasize the dominating role of WH forcing in driving NH SST responses. As is pointed out by the reviewer, we realize that our sentence may bring confusion. We modified this paragraph to describe our conclusions more clearly:

"Two sets of regional FF simulations (Fix_EastFF1920 and Fix_WestFF1920) are performed to separate the roles of East versus West aerosol forcings, which had clearly opposite trends in the last 40 years. We find that the WH aerosol reduction dominated the simulated warming over NH mid-to-high latitudes."

3. L38-40, IPCC AR6 shows it is 1.1C

Response:

Thanks for the correction. We have changed the description from 1C to 1.1C.

4. L40-42: Dong and Mcphaden (2017) also shows a good example of how the GHGs and internal variability, such as IPO, shapes the global warming at the decadal time scale.

Lu Dong and Michael J McPhaden 2017 Environ. Res. Lett. 12 034011.

Response:

Thanks for the suggestion. We added the reference to the first paragraph of the Introduction section.

 5. L53-55: Similar conclusions have been obtained in earlier studies, such as Salzmann (2016). Salzmann, M. Global warming without global mean precipitation increase? Sci. Adv. 2, e1501572 (2016).

Response:

Thanks for the suggestion, except that Salzmann's study is about global mean precipitation, rather than the precipitation extremes under various intensities as examined in Lin et al. (2018).

We added the suggested references to the manuscript.

6. L44-61: Although there are many differences between GHGs and aerosols as you mentioned here, some other studies found some climate impacts caused by GHG and aerosols can be very similar. For example, Xie et al. (2013) showed the SST and ocean precipitation response patterns are very similar in both GHGs and aerosols. Recently,

Song et al. (2021) also shows both GHG and aerosols modulate the seasonal delay of tropical rainfall in a similar way, i.e., by modulating the atmospheric column humidity. This similarity between the two forcings should also be mentioned.

Xie, SP., Lu, B. & Xiang, B. Similar spatial patterns of climate responses to aerosol and greenhouse gas changes. Nature Geosci 6, 828–832 (2013).

Song, F., Leung, L.R., Lu, J. et al. Emergence of seasonal delay of tropical rainfall during 1979–2019. Nat. Clim. Chang. 11, 605–612 (2021).

Response:

Thanks for the suggestion. We add such discussion on the similar forced responses to the introduction.

"However, despite the subtle differences between GHGs and aerosols, other studies found similar climate responses to GHGs and aerosols. Xie et al. (2014) found that the regional ocean temperature and precipitation in response to GHGs and aerosols are similar, suggesting the importance of the spatial distribution of radiative changes. Song et al. (2021) show that increasing GHGs and decreasing aerosols in the recent decades both delay rainfall by inducing a moister atmosphere. Both the differences and similarities between GHGs- and aerosol-induced climate responses indicate the complexity and importance of the temporal and spatial distribution of AA forcings. " 7. L59-61: As mentioned above, Song et al. (2021) found the recent decreases of aerosols, combined with the increased GHGs, contribute significantly to the seasonal delay of tropical rainfall. As the decreased aerosol and increased GHG will continue in the future, the seasonal delay of tropical rainfall is expected to amplify in the future.

Response:

Thanks for the suggestion. We have added the following to the text:

"The FF-related aerosols are projected to further decrease in future decades (Andreae et al., 2005; Zheng et al., 2020), even for Asian regions, with more strict air quality measures in developing nations. The future decline of FF aerosol will lead to further unmasking and warming in addition to GHG-induced global warming (Xu et al., 2015; Wang et al., 2018; Lelieveld et al., 2019; Allen et al., 2020) and have consequences for heat extremes (Zhao et al., 2019; Xu et al., 2020) and humidity and precipitation (Song et al., 2021)."

8. L69: for the first reference: Who?

Response:

Thanks for pointing out the editing issue. Should be "Allen et al., 2014"

9. L78: tropics->tropical

Response:

Thanks for the correction. We have fixed this issue.

10. L166-167: Fix_FF1920 have 20 members, but here Fix_EastFF1920 and Fix_WestFF1920 only contain 10 members, is there any sensitivity of the results to the member numbers? For example, if you also only use 10 members of Fix_FF1920, could you obtain the similar results?

Response:

Thanks for the suggestion. We conducted sensitivity tests to see if ten ensemble members are sufficient enough to smooth out the randomly generated internal variabilities in the model. We calculated the Tripole Index for the Interdecadal Pacific Oscillation (TPI; Henley et al., 2015) based on each individual simulation and the multi-member-mean (MMM) results for each experiment, which is shown in Fig. R1.

Although we still see some small variations in the MMM TPI index of Fix_EastFF1920 and FixWestFF1920, the majority of the internal variability is successfully smoothed out. There is no doubt that the more ensemble members exit, the less model-generated internal variability will exist in MMM results, but considering that Fix_FF1920 and the two regional experiments show the similar magnitude of variation, we believe that 10 members are acceptable for the purpose of separating externally driven responses and the randomly distributed variability.

Henley, B.J., Gergis, J., Karoly, D.J., Power, S.B., Kennedy, J., Folland, C.K., (2015). A Tripole Index for the Interdecadal Pacific Oscillation. Clim. Dyn. 45 (11–12), 3077–3090, <u>http://dx.doi.org/10.1007/s00382-015-2525-1</u>.



Fig R1:

Model-generated TPI index from (a) ALL, (b) Fix_EastFF1920, (c) Fix_FF1920, and (d) Fix_WestFF1920. The grey lines represent each ensemble member; the black curve represents the TPI index calculated from the ensemble average. Note that ALL has 40 members; Fix_FF1920 has 20 members; Fix_EastFF1920 and Fix_WestFF1920 have 10 members, respectively.

Another relevant question is that you should also show whether the trend of many variables you focused here in the Fix_FF1920 is roughly the sum of Fix_EastFF1920 and Fix_WestFF1920.

Response:

Thanks for the suggestion. We conducted the additivity test as suggested on the burden of sulfate aerosol and surface temperature (Fig. R2).

For the column burden of sulfate, the results in response to FF resemble the sum of EastFF and WestFF (SUM).

For the surface temperature (TS), FF-induced TS trends are also similar to the sum of EastFF and WestFF, except for the central Pacific and part of the Arctic region. The possible reason for the slightly larger warming in SUM may be related to the residue of internal variability (as shown in Figure R1 above) due to the limited ensemble sizes, the responses to the unfixed regions (such as the Arabian Peninsula and Africa), or the nonlinearity issues due to model subtraction. Overall, we argue that the sum of the two regional experiments (EastFF and WestFF) show very similar responses compared with FF responses and thus are capable of separating the East versus West aerosol forcings.

We add the following discussion to the Method section:

"An additivity test is conducted to evaluate whether the summation of EastFF and WestFF can roughly reproduce FF. The SO4 column burden (BURDENSO4) and surface temperature (TS) in response to FF and EastFF + WestFF (SUM hereafter) are shown in Fig. 3. The FF-induced SO4 column burden resembles the sum of the SO4 burden from SUM. The TS responses are also very similar between FF and SUM, except for the central Pacific and part of the Arctic region. The warmer patterns over the central Pacific in SUM compared to FF is possibly related to the TS responses to remote forcings beyond the two regions in consideration here (e.g., Arabian Peninsula, South America, and Africa), the residues effects of internal variability even after ensemble average due to limited ensemble sizes. Overall, the sum of two sets of regional fixed single forcing experiments well represent the major patterns of FF aerosol induced response, and thus the two new sets of simulations here are capable of separating the East versus West aerosol forcings."



Fig R2 (the new Fig. 3):

Left column: the 40-year trend of (top) Sulfate column burden (kg/km2/decade), and (bottom) surface air temperature in response to FF.

Right column: So as to the left column for the summation of EastFF and WestFF.

11. L201-202: We know the decreased trend of FF-related aerosols in the North America and Europe is due to the clean air acts and increased trend of FF-related aerosol in the India and China is due to the economic development, but what's the reason of the stronger increasing trend of BB-related POM over the northeastern Asia?

Response:

Thanks for the question. Based on Deser et al. (2020) and Lamarque et al. (2010), the BB-related POM over northeastern Asia is due to the increasing biomass combustion from forest fires.

Deser, C., et al. (2020). Isolating the Evolving Contributions of Anthropogenic Aerosols and Greenhouse Gases: A New CESM1 Large Ensemble Community Resource, Journal of Climate, 33(18), 7835-7858.

Lamarque, J.-F., et al. (2010). Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017–7039,

https://doi.org/10.5194/acp-10-7017-2010

12. L217: removing "1"

Response:

Thanks for the correction. We have fixed this issue.

13. L241: and FF-> FF and.

Response:

Thanks for the correction. We have fixed this issue.

14. L238-240: Could you explain a little bit more about how the indirect effects of aerosols (i.e., cloud droplets number and cloud lifetimes are enhanced) could expand the affected regions? Do you mean the cloud formed in the emission region can be transported to other places?

Response:

Thanks for the question. Reviewer #1 also touched on this issue.

We include the cloud number concentrations in Fig. 3 (the new Fig. 4) and see significant cloud droplet changes over the subtropical Pacific regions due to the aerosol emission over Asia. We also see some cloud droplet changes over the North Pacific region due to aerosol reduction from North America. Therefore, we argue that the indirect aerosol effects extend beyond the emission domain and well into the ocean.

Although the results above indicate that aerosol indirect forcing (fast response) contributes to the tropical Pacific cloud changes, we also agree with reviewer #1 that cloud fraction changes are partially driven by the SST and circulation responses (so-called slow response). For example, the eastern subtropical Pacific in the Southern Hemisphere shows cloud fraction changes without much CDNC change.

15. L244: should be decrease of CLDTOT rather than increase in response to WestFF based on Fig. 3?

Response:

Thanks for the question, but actually, there is an increase of CLDTOT over the <u>Pacific warm pool region</u> in Fig 3



Fig. R3 (the new Fig. 4)

The 40-year trend in cloud droplet number concentration (CDNUMC) and total cloud fraction (CLDTOT) in response to FF, EastFF, and WestFF.

16. Figure 3. You mentioned that regions passing the 95% significance is dotted, but I didn't see any dots there. You may also need to do the significance test in Fig. 2 and many other figures.

Response:

Thanks for pointing out the editing mistake. We now added the significance test to the horizontal pattern figures (the new Fig. 4 and Fig. 10).

For the vertical pattern figures, we have masked insignificant regions to be white.

17. Fig. 5: What did you do when you say "smoothed in 30 degrees of latitudinal range" **Response:**

We mean we smooth the latitudinal profile using a moving average with a 30DegLat window. We modified the sentence to clarify our method:

"The trend and gradient lines are smoothed using the moving average method (with 30 degrees of latitudinal range sampling window)."

18. would you like to mention how the increased FSNTOA gradient drives the equatorward shift of the NH jet stream?

Response:

Thanks for the suggestion. By introducing the "rule-of-thumb" indicator, we argue that the latitudinal gradient of FSNTOA, a measurable quantity, can be considered as an indicator of the circulation shifts in NH. We do not intend to claim that the circulation changes are simply driven by the FSNTOA gradient. We further clarify our point in the manuscript to avoid misleading.

However, since reviewer #1 also suggested adding some dynamic analysis, we now include a new subsection discussing the relationship between the shift of circulation and the cross-equatorial atmospheric energy transport (AET0), specifically for the Hadley Cell shift. Due to the length of the manuscript, we did not include further diagnostics on the NH jet streams. More detailed analyses on jet streams will be presented in a future study, as mentioned in the Conclusion.

The heavily modified Section 3.3 portion related to meridional energy transport now reads:

"Previous studies have explored the tropospheric circulation responses to inter-hemispheric (meridional) forcing gradient due to anthropogenic aerosols – more reflecting aerosols over NH compared to SH will lead to an equatorward shift of NH Hadley circulation and NH westerly wind (e.g., Hwang et al., 2013; Hilgenbrink et al., 2018). Meanwhile, recent studies also put effort into how the west-east contrast effects of aerosol induce circulation changes (Wang et al., 2015; Kang et al., 2021). However, from 1980 to 2020, NH anthropogenic aerosol forcing (Sect. 3.1) is highly heterogeneous, with both strong zonal contrasts and subtle latitudinal differences (Fig. 4), further compounding the forcing-response relationship (Shindell and Faluvegi, 2009; Persad and Caldeira, 2018). Next, we analyze the aerosol-induced tropospheric responses in terms of zonal average, both globally and regionally, for the EH and WH portions (marked as red boxes in Fig. 4a).

Figure 6a–c shows the decadal trend of global Zonal Mean Meridional overturning Stream Function (ZMMSF) in response to FF, EastFF, and WestFF during 1980–2020. The ZMMSF, in response to FF, features a counter-clockwise Hadley Cell anomaly (shown in blue) over the tropics, which indicates a northward shift of the Hadley Cell into NH. The northward shift of Hadley Cell also clearly occurs in response to WestFF, but not to EastFF, indicating that the shift of Hadley Cell is mainly due to the WestFF. The global mean ZMMSF shifts in our results are consistent with previous studies (Xu et al., 2015; Allen and. Ajoku, 2016; Amaya et al., 2018; Shen et al., 2018) focusing on the inter-hemispheric forcing gradient. That is, the tropical circulation always tends to move towards a warmer hemisphere with larger positive forcing.

To further diagnose why EastFF and WestFF induce distinct changes of the Hadley Cell, we calculated the zonal, column integrated meridional energy transport in response to aerosol forcings, shown in Fig. 7b-d. The Atmospheric Energy Transport (AET) is calculated based on the:

$$\frac{\partial}{\partial \Phi} F_a = R_{TOA} - Q, \qquad (2)$$

Where Φ is latitude, F_a (a function of latitude and longitude) is the meridional energy flux, R_{TOA} is the net radiative flux at the top-of-atmosphere (downward positive), and Q is the net downward energy flux at the surface. Q includes shortwave radiation, longwave radiation, sensible heat flux, and latent heat flux. AET is then obtained by integrating the energy flux from south to north:

$$AET(\Phi) = 2\pi a^2 \int_{-\pi/2}^{\Phi} \cos \Phi' \left(R_{TOA} - Q\right) d\Phi'$$
(3)

Where a is the Earth radius. The oceanic energy transport (OET) is calculated based on the: The oceanic energy transport (OET) is calculated based on the:

$$\frac{\partial}{\partial \Phi} F_o = Q \tag{4}$$

The positive radiative forcing in NH extratropics from WestFF induces a strong negative AET at the equator (Fig. 7d), which leads to the northward shifts of Hadley Cell and ITCZ to balance the interhemispheric difference in radiative forcing. Previous studies demonstrated that cooling NH leads to a southward shift of ITCZ (Broccoli et al., 2006, Kang et al., 2021), and the mechanism is consistent with what we find here. On the other hand, the EastFF introduces a strong negative radiative forcing close to the NH tropics and a weak positive forcing in NH extratropics; as a result, the AET has much smaller trends at all latitudes compared to WestFF (Fig. 7c vs. Fig. 7d). Therefore, the Hadley Cell does not shift significantly in response to EastFF. The AET changes in response to the total FF (Fig. 7b) closely resemble that in response to WestFF, again confirming the dominant

role of WestFF in driving the Hadley Cell. OET at the equator in response to all three cases shows a near-zero trend, so it does not contribute much to the shift of Hadley Cell. "



Fig R4 (added as the new Figure 7)

(a) The 40-year climatology of Northward energy transport (Pwatt) is calculated based on ALL experiments ensemble average. (b–d) The decadal trend of northward energy transport in response to FF, EastFF, and WestFF (Pwatt/decade), which are obtained by subtracting the fixed single forcing experiments from the ALL experiment.

The dashed blue lines represent the oceanic energy transport; the solid red lines represent the atmospheric energy transport.

19. L371: below 35N? seems problematic. Suggest changing to southward of 35N Response:

Thanks for the suggestion. We now change all "below X °N" to "southward of X °N", and all "above X° N" to "northward of X °N".

20. Fig. 6: Here, why do you only focus on the EH, rather than Global in previous figures? Could you explain it a little bit?

Response:

Thanks for the question. We include Fig. 6 (now Fig. 8) to compare geostrophic wind theory (upper row) and the shift of NH jet stream (lower row). Since we already show the global jet stream in Fig. 5 (now Fig. 6), we show EH jet and geostrophic wind in Fig. 6 as an example to avoid duplicated panels. Actually, the consistency between geostrophic wind and jet stream holds true in all cases of ALL, EH, and WH. We mentioned this in the text:

"The derived Ug patterns always resemble the simulated U pattern in EH (Fig. 8d–f), WH, and Global (not shown), revealing the strong correlation between tropospheric circulation changes and the tropospheric temperature changes (and thus the geopotential height changes)."

21. L394: sometimes using FSDS, but in other cases, you use FSNTOA. Please justify your choices.

Response:

Thanks for the suggestion. This is due to the similar rationale of Comment/Response 20 above. We previously used FSDS for the shift of Hadley Cell (previous Fig. 5; has been removed in the new Fig. 6 now) and FSNTOA for the jet stream (the new Figure 6). Both FSDS and FSNTOA are treated as the "rule-of-thumb" indicators of the circulation shift. In Fig. 5 (the new Fig. 6), we compare these two and find that these two gradients are pretty similar.

As suggested by the reviewer, and considering the similarity between FSNTOA and FSDS gradient, we now use FSNTOA as the rule-of-thumb indicator throughout the manuscript.

22. L418: references are needed here.

Response:

Thanks for the suggestion. We have added references to the text.

"which is mentioned by previous studies that demonstrate the north Pacific cooling due to Asia aerosol emissions (Dong et al., 2014; Takahashi and Watanabe, 2016; Smith et al., 2016)."

Takahashi C, Watanabe M (2016) Pacific trade winds accelerated by aerosol forcing over the past two decades. Nat Clim Chang 6:. https://doi.org/10.1038/nclimate2996

Dong L, Zhou T, Chen X (2014) Changes of Pacific decadal variability in the twentieth century driven by internal variability, greenhouse gases, and aerosols. Geophys Res Lett. https://doi.org/10.1002/2014GL062269

Smith DM, Booth BBB, Dunstone NJ, et al. (2016) Role of volcanic and anthropogenic aerosols in the recent global surface warming slowdown. Nat Clim Chang 6:936–940. https://doi.org/10.1038/nclimate3058

23. L430: induce es->induces a

Response:

Thanks for the correction. We have fixed this issue.

24. L468: other ver?

Response:

Thanks for the correction. It should be "over". We modified this sentence and fixed the error.