**Response to Reviewers**

**Reviewer #2**:

“The manuscript presents a study of where emissions from NOx and primary aerosols are modified sequentially to improve AOD predictions over eastern Asia using TROPOMI NO2 data and AOD products from multiple geostationary satellites (including GEMS). This is done for two periods, one with resulting increasing of emissions and another with decreases due to COVID lock-down conditions. This study represents a great contribution to the field and it’s within the scope of the journal. The manuscript is well written and referenced.

One of my major concerns is that I think organic aerosols are being treated as primary aerosols which is a misconception. This likely results in an overprediction of the contribution of primary aerosols. More discussion on the topic and caution on how this data might be used needs to be included as is likely that changes attributed to primary PM emissions should really be attributed to changes to precursor gases other than NOx. This needs to be addressed throughout the manuscript.

Another concern is that when reading the title and abstract it gives the impression this study is using GEMS trace gas data which is not the case as the only GEMS product being used is the AOD one after being fused with a few other datasets. I would encourage the authors to rephrase the title and abstract to avoid giving these expectations, as there are high expectations from the community about studies assimilating trace gas retrievals from GEMS.

Additional comments line by line can be found below.”

**Authors’ response:** We appreciate your time and concern devoted to reviewing this manuscript. Please find our responses to your comments below:

1. One of my major concerns is that I think organic aerosols are being treated as primary aerosols which is a misconception. This likely results in an overprediction of the contribution of primary aerosols. More discussion on the topic and caution on how this data might be used needs to be included as is likely that changes attributed to primary PM emissions should really be attributed to changes to precursor gases other than NOx. This needs to be addressed throughout the manuscript.

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| Author’s response | Thanks for providing good discussion points that need to be enhanced. In response, we have made multiple updates throughout the manuscript, which will comprehensively address your major concern and minor points as below. |

1. Another concern is that when reading the title and abstract it gives the impression this study is using GEMS trace gas data which is not the case as the only GEMS product being used is the AOD one after being fused with a few other datasets. I would encourage the authors to rephrase the title and abstract to avoid giving these expectations, as there are high expectations from the community about studies assimilating trace gas retrievals from GEMS.

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| Author’s response | We agree that the title and abstract might give wrong impression or expectation to the community, and we have made updates in these to prevent such a situation. |
| Changes in manuscript | * Title: Satellite-based, top-down approach for the adjustment of aerosol precursor emissions over East Asia: TROPOMI product, and the Geostationary Environment Monitoring Spectrometer (GEMS) data fusion product and its proxy * Line 19: “… using a series of GEMS data fusion product and its proxy data, TROPOMI data, and CTM-based inverse modeling techniques ….” * Lines 33-34: “… supported by TROPOMI and GEMS-involved data fusion products …” |

**Minor points:**

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| Reviewer’s comment | Author’s response | Changes in manuscript |
| 238-241. Please provide additional information with respect to iteration procedure. Is this iterating from month to month? Or is this an iteration within the same month to find convergence? Also clarify if F and the jacobian matrices are recomputed after each iteration. Eqn 3 can generate negative values, so also provide information on how that was handled. | Thanks for bringing up these discussion points, and we agree that these need more detailed descriptions.  During inverse modeling, we performed iteration within each month to achieve convergence. The DDM-3D-derived sensitivities and their Jacobian matrices following each iteration, as you have already mentioned.  Regarding Eq. 3, we are aware that the equation can derive negative values. Among several confirmed approaches to constrain the negative a posteriori (Bergamaschi et al., 2009; Corazza et al., 2011; Souri et al., 2018; Vojta et al., 2022), and we employed Souri et al.’s (2018) approach towards optimizing log() instead of to add a hidden constraint.  The discussions above were added to the manuscript accordingly. | * Lines 242-244: “We iterated Eq. 3 two times within each month to attain convergence, and and were updated after each iteration. It should be noted that we derived log() instead of to constrain negative a posetriori values, the details of which are described in Souri et al. (2018).” |
| 242-256. What prior emissions are used when doing the primary PM emission estimations? Line 251 says that PM adjustments are applied to the NOx-constrained emissions but is not clear if the NOx constrained emissions are used as the prior for the PM emission estimation or not. | Yes, as you pointed out, during the primary PM emissions adjustment, the a priori emissions refer to the NOx-constrained emissions. We agree that this should be clarified, and we have made changes in the manuscript accordingly. In addition, we further enhanced the details of the primary PM emissions adjustment process. | * Lines 249-254: “To adjust the primary PM emissions, we applied analytical inversion described in Eqs. 2 and 3 to the emissions of 19 primary PM species predefined as contributors to the AOD in the 6th generation CMAQ aerosol module (AERO6) (Simon, 2015) … is a priori primary PM emissions (in the NOx-constrained emissions inventory obtained earlier) … was set as 100% (Crippa et al., 2019).” * Lines 261-264: “In this approach, represents the sensitivity of the total primary PM emissions with regard to changes in the AOD … the loadings of such species over vast areas in East Asia in a top-down manner.” |
| 242-256. Aerosols in east Asia are mostly secondary unless coming from biomass burning or dust events. But this approach is scaling primary PM emissions. This caveat and discussion on the limitations of this approach needs to be discussed in the text. If NOx constrained emissions were used as prior for the PM emission constraints this reduces the problem only partially as discrepancies in AOD could be attributed to emissions from other precursors such as VOCs, SO2 and NH3. | Thanks for providing a valid discussion point. As mentioned in your comment, our NOx and PM emissions adjustments do not consider the precursors of other secondary inorganic aerosols (i.e., SO2 and ammonia for sulfate and ammonium aerosols, respectively) and of secondary organic aerosols either. We agree that this limitation needs further elaboration with more details, and we have updated the manuscript accordingly. | * Table S2 and caption: “The list of the primary PM species included the KORUS-AQ emission inventory, and the corresponding pollutants simulated in CMAQ version 5.2 and measured at the Korean supersites.” * Table 2 and caption: “Concentrations (μg/m3) and compositions (%) of surface PM2.5 and its components in Korea … Others: the summation of unknown (undefined) PM2.5 species.” * Lines 404-415: “… the remaining portion (46.74% on average) was mostly comprised of primary PM (36.32% on average) and some unknown (undefined) aerosols (Table 2). As both the contributions of primary and secondary aerosols to aerosol loadings were significant, we considered … that employs more comprehensive sets of top-down constraints (e.g., observational references for SO2 and ammonia loadings in the troposphere).” |
| 250-252 If I’m reading the text correctly, NOx emissions were estimated at a monthly scale and primary PM emissions at a daily scale? Can you elaborate these different timescales were chosen? | Yes, as mentioned already in your comment, NOx emissions were adjusted at a monthly scale and primary PM emissions were adjusted at a daily scale.  While the temporal resolutions of those AOD products afforded by the geostationary platforms were sufficiently fine to be used in the daily emissions adjustments, the temporal resolution of TROPOMI NO2 columns was too coarse to be utilized in daily NOx emissions adjustment (given zero to one valid snapshot of columnar NO2 over the modeling domain per day). We expect that this limitation could possibly be resolved by employing GEMS tropospheric NO2 columns in inverse modeling in the follow-up study.  Based on your suggestion, we have enhanced the description for the use of different time scales accordingly. | * Lines 270-273: “It should be noted that NOx emissions were adjusted monthly, due to the relatively coarse temporal resolution of TROPOMI NO2 columns (providing zero to one valid snapshot of columnar NO2 per day over the modeling domain), while primary PM emissions were adjusted daily by using the AOD products at sufficiently fine temporal resolutions afforded by geostationary platforms.” |
| Section 2.5.1. Can you clarify how NOx and primary aerosol emissions are scaled spatially? Are different correction factors derived for each grid cell? Is there any spatial correlation used within neighboring cells? | Yes, as mentioned in your comment, we applied the adjustment ratio (for scaling the a priori to the extent of the a posteriori) to each grid cell. We first regridded the observation references, the spatial resolutions of which vary with different instruments, to CMAQ’s modeling grids (27 km × 27 km) by using the distance-weighted mean of those grid-based references with a radius of 0.25° (approximately 27 km); in this way, the resultant each grid-based adjustment ratio from the inversion will be already collocated with each of CMAQ’s grids.  Based on your suggestion, we have enhanced related descriptions in the manuscript. | * Lines 170-172: “To ensure consistency in the horizontal spacings between the TROPOMI NO2 columns and CMAQ’s modeling grids, we regridded the TROPOMI NO2 columns into 27 km × 27 km grids by using distance-weighted mean of those observation references with a radius of 0.25° (approximately 27 km).” * Lines 199-200: “The consistency in the grid spacings among AHI AOD, GOCI-AHI AOD, and CMAQ’s modeling grids was ensured in the same approach described in Section 2.2 above.” |
| Section 2.5.2. Why not apply the same approach as in section 2.5.1 for NOx emissions on 2022? Eqn 5 might only be valid is meteorological conditions were consistent for both years. Unless there is a very good reason for doing this, I would suggest using the same approach for consistency. | Thanks for acknowledging the valid discussion point. The rationale for employing the basic mass balance approach for the 2022 NOx emissions was due to the limited timeline afforded for this study. As the period of interest, i.e., 2022, was relatively recent compared to 2019, we had to process the most recent satellite data in a concurrent manner as soon as the datasets were made available for use like a relay race. We hope this explanation clarifies our reasoning. | - |
| Are AERONET sites considered over the whole domain or only over Korea? | We used all AERONET sites available for the entire domain. To clarify this, we have updated Figure 1 to depict the locations of AERONET sites, as well as other ground-based in-situ measurement sites. | * Figure 1 and caption: “Modeling domain and the locations of the ground-based in-situ measurement sites used for model evaluation.” |
| How is organic aerosol included in this summation of lumped species? Organic aerosols are a mixture or primary and secondary aerosols, with a big fraction of it being secondary for anthropogenic pollution other than biomass burning (e.g., see papers from Jose Jimenez group at CU-Boulder), and thus if organic aerosol is being considered as primary this is a strong misconception that needs to be addressed. Additionally, sampling of organic aerosol is a difficult undertaking, and it is been found that routine measurements as those used in the Korean sites might underpredict organic aerosol as compared to the more research grade measurements (like those from an High res -time of flight – aerosol mass spectrometer). You can refer to KORUS-AQ measurements for insights on this. | Thanks for pointing out the insufficient description for the lumped PM species, which may misinform readers regarding the presence of organic constituents.  We believe that the updates made in the manuscript in response to your earlier comment on Lines 242-256 above (“Aerosols in east Asia ...”) can partially address the concerns in this comment. The measurements made at Korean supersites, other than OC, do not consider organic aerosols as a target, which leaves concerns mentioned in your comment (e.g., underpredicted loadings of organic aerosols). |  |
| How is dust being measured? If it’s through ions, generally only a small fraction of the total mass concentration is captured. | No direct dust measurement was available at Korean supersites, and we apologize for the misinformation.  The updates made in the manuscript for your pervious comment include the corrections made for this comment. | * Table 2 and caption * Table S2 and caption * Lines 312-313: “… the lumped summation of the primary PM species listed in Table S2, and the rest remaining undefined.” |
| It would be great if the emission changes could be aggregated on a per country or per region basis, as emissions generally are based on what’s reported by each country, which will help inform the teams producing those emissions. Also, evaluation against NO2 surface measurements is only done in Korea, so knowing what emissions changes were found here would help the interpretation. | Thanks for providing us with a great discussion point, and we have been considering it as one of our focuses in follow-up studies.  Once we secure a sufficient amount of ground-based in-situ measurements available across other subdomains of interest (e.g., Mongolia, Russia and Japan) for model evaluation (which will determine whether the top-down inversion and the corresponding changes in the bottom-up estimates of subdomain-specific emissions were valid or not), we will be able to perform such country-, region-, and province-specific assessments of bottom-up emissions.  In addition, we enhanced the interpretations regarding the changes in emissions their subsequent impact on model performances in both Korea and the NCP region in China. These updates in the manuscript will be used for addressing several other comments of yours below. | * Lines 324-326: “Then we evaluated … in Korea and the NCP region in a time series.” * Lines 331-337: “However, in the NCP region … not as effective in reducing the model biases in the NCP region as it was in Korea.” * Lines 355-357: “In brief, the model's initial underestimation of AOD was mitigated by the NOx emissions adjustment, which led to increased NOx emissions, and then by the subsequent primary PM emissions adjustment, which resulted in overall increases in primary PM emissions.” * Lines 364-369: “Despite the success of the sequential adjustments … … region-specific tactics for adjusting the bottom-up estimates of gas-phase air pollutant emissions in future studies.” * Lines 440-450: “For example, in MAM 2019 … was considered to better capture the high AOD peaks across the southeast China in a spatiotemporally more frequent and continuous manner, was more effective in resolving the model’s initial AOD underestimation.” |
| Figure 2 and 3. Shouldn’t columns b) and c) be the same plots in both figures as is the same base year and same emissions? They look quite different in both figures. | Yes, the columns b) and c) in Figure 2 and those in Figure 3 are based on the same base year and emissions to each other’s.  To ensure the consistency during the spatial comparisons (CMAQ AOD versus AHI AOD, and CMAQ AOD versus GOCI-AHI AOD), we temporally collocated CMAQ AOD to AHI AOD and GOCI-AHI AOD each because each has different acquisition time per valid AOD retrieval.  To clarify this, we have made updates in the manuscript. | * Figure 2 caption: “Note that CMAQ-simulated AODs were temporally collocated to the AHI AOD.” * Figure 3 caption: “Note that CMAQ-simulated AODs were temporally collocated to the GOCI-AHI AOD.” |
| Figure 2-4. There still seems to be a substantial gap for AOD after the inversions. Thus, I would encourage the authors to discuss potential reasons for this behavior. One might be related to the approach of only scaling primary PM, while most of the aerosol might be from secondary origin. It was not clear to me how emissions were modified spatially, so depending on how’s that done that could be another potential reason. | Thanks for the detailed concerns, and we agree that such limitations of this study (i.e., the precursors of other secondary aerosols than nitrate remaining unadjusted, and consequent impact on the model performances) need further elaboration.  For example, we found that the increase in model bias after the NOx emissions adjustment in the NCP region for certain seasons needs further discussions. In short, since the model once experienced severe AOD underestimation, the overall “increases” in NOx emissions (regardless of the improvement or degrade in the corresponding model accuracies) helped the model mitigate the AOD underestimation.  We believe that the updates made in the manuscript in response to your earlier comment above (“It would be great if the emission changes …”) can address the need for further details. |  |
| As mentioned above, it looks like organic aerosol is being considered as primary aerosol which is generally not the case. Thus, some of the conclusions derived here might not be accurate. I think there needs to be text suggesting that is likely that the corrections to primary PM emissions might be overpredicted as they are compensating for changes that might need to be made to precursor gases other than NOx. |
| 444-445. This is stating that things improved due to GEMS, which in my opinion is not clear from these results as multiple other datasets are being used. To make this point more clear you would have to add an additional test where GEMS is not used and compare it to the one with GEMS for the same period. | Thanks for pointing it out, and we have noticed that the statement and its nuance, which seem to be specifically highlighting the utility of the GEMS-involved products, does not fit into context of the paragraph and may mislead readers. In response, we have made updates in the manuscript.  To better support the updated conclusion, it was desirable to either 1) compare the amount of information (i.e., the number of AOD records) afforded by 2022 GEMS AOD (2019 AHI AOD was the proxy of it earlier) versus that afforded by 2022 GEMS-AMI-GOCI-2 AOD (2019 GOCI-AHI AOD was the proxy), or 2) compare those afforded by 2019 GOCI-AHI AOD and 2022 GEMS-AMI-GOCI-2 AOD each other.  Unfortunately, neither approach was available for this study. The 2019 GOCI-AHI AOD product used earlier was served as a prototype for the development of the 2022 GEMS-AMI-GOCI-2 AOD product (the production of the GOCI-AHI AOD product has been discontinued, and it is currently only available for research purposes for the year 2019). Also, the GEMS AOD product and its algorithms are currently on their development stages (2-D rendered products are available for the general public) according to the data provider (NIER). | * Lines 527-529: “The enhanced observation quality and quantity afforded by the GEMS-involved synergistic product and its proxy appeared to be beneficial to capturing the spatiotemporal variations in the emissions of the aerosol precursors.” * Lines 463-465: “Note that the GOCI-AHI AOD product used earlier was served as a prototype … discontinued, and it is currently only available for research purposes for the year 2019.” |

**References**

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