All reviewer comments are copied and pasted below unedited in normal text, while our responses are given in blue text.

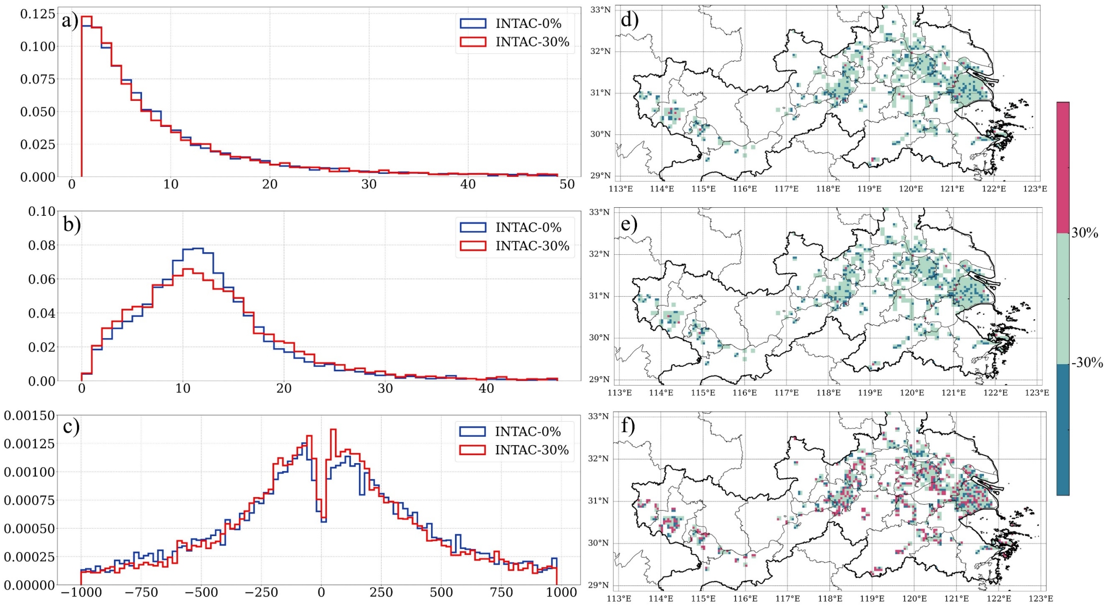
This study introduces a novel mass-balance based analytical framework designed to rapidly and flexibly quantify NO_x emission fluxes with high spatial and temporal resolution, using daily observations from TROPOMI across three rapidly changing regions in eastern China. The study effectively quantifies source attribution by revealing unprecedented insights into NO_x/NO_2 emission ratios across five industrial sources. The results also indicate significant discrepancies between observed emissions and those predicted by the MEIC a priori inventory. Furthermore, the paper emphasizes the substantial NO_x emissions linked to small and medium industrial and residential activities in regions adjacent to rivers. This paper stands as a well-executed and valuable piece of research which is overall well-written. The methodology is emerging and innovative, and the results contribute valuable insights into the emissions landscape, there are certain critical details that require clarification. I look forward to re-reviewing the paper in the next round, provided that the following comments can be addressed.

Major comments:

The paper presents fitted alpha1-alpha3 values, but it is important to assess how sensitive these values are to variations in the initial assumptions about NO_x emissions. Did the authors conduct a sensitivity analysis to determine if the alpha values remain stable under different scenarios? This information would provide insight into the reliability of the emissions estimates and the overall methodology.

To address your concern about the robustness of our approach, we conducted sensitivity analyses to verify that the fitted coefficients (alpha1-alpha3) are relatively stable even with imperfect assumptions surrounding the a priori NO_x emissions used in the first step.

The priori emissions dataset (INTAC) utilized in this study was developed by the MEIC team in collaboration with various authoritative research institutions. We consider it the most suitable choice for priori datasets over China. The uncertainties in INTAC arise from its integration process. Relatively low uncertainties are exhibited by NO_x emissions of INTAC, benefiting from well-established estimates for large-scale combustion sources. Given this, we assume that the uncertainty is less than 30%. In this study, we selected Region 2 and tested what impacts would occur if the prior emissions were to have uncertainties near the extreme upper and lower bounds of their ±30% uncertainty range. The coefficients were refit using new priori emissions values ($INTAC_{30\%}$ Case) while maintaining the same values from TROPOMI. The results are presented in Response Figure 1. It is observed that for most grids (over 60%), the NO_x/NO_2 ratios and lifetimes exhibit significant robustness following the 30% perturbation of INTAC as the priori. However, the transport parameter demonstrates less robustness, with approximately 40% of grids showing a lower ratio than the 30% perturbation, meaning slightly over half of the total grids may suffer from a non-linear response. As illustrated in Response Table 1, the 20th and 80th percentile ranges of NO_x/NO_2 are from 2.8 to 14.6 and from 2.7 to 15.0 in the $INTAC_{0\%}$ and $INTAC_{30\%}$ cases, respectively. The corresponding ranges for lifetime (days) are from 0.29 to 0.68 and from 0.27 to 0.73. The differences in transport (km) are slightly larger than those of the other coefficients, ranging from -323. to 351. and from -289. to 314. Our findings indicate that the variations in the fitted coefficients are relatively minor, thereby confirming the robustness of our method. And any slightly larger changes in the transport term are buffered by the non-transport terms.



Response Figure 1. The distributions of three key coefficients obtained from $INTAC_{0\%}$ and $INTAC_{30\%}$: a) NO_x/NO_2, b) Lifetime [hours], c) Transport [km]; The ratio of ($INTAC_{30\%}$-$INTAC_{0\%}$)/$INTAC_{0\%}$ is also displayed on: d) NO_x/NO_2, e) Lifetime [hours], f) Transport [km];

Response Table 1. The 20th, 50th,80th percentile ranges of three coefficients from $INTAC_{0\%}$ and $INTAC_{30\%}$.

NO_x/NO_2	$INTAC_{0\%}$	$INTAC_{30\%}$
20%	2.76	2.68
50%	6.30	6.24
80%	14.6	14.9
Lifetime (days)	$INTAC_{0\%}$	$INTAC_{30\%}$
20%	0.29	0.27
50%	0.48	0.48
80%	0.68	0.73
Transport (km)	$INTAC_{0\%}$	$INTAC_{30\%}$
20%	-323.	-289.
50%	18.1	16.3
80%	351.	314.

We hope this clarifies the issue and demonstrates the reliability of our approach. Please let us know if you have any further questions or require additional details.

In Section 2.1, line 110, why did you choose to exclude column NO_2 has a climatology smaller than 1.43×10^{15} molec/cm^2. How was this number determined?

The TROPOMI NO_2 retrievals were analyzed with a background noise cutoff, as suggested by previous studies (Qu et al., 2021; Tack et al., 2021), which is computed to be approximately 1.43×10^{15} molecules/cm^2, based on a combination of both a constant term on the uncertainty as well as a term which scales with the total column loading impacting the uncertainty (in urban and industrial types of areas). The resulting solutions are inherently self-constrained within a specific range due to the physical constraints on the acceptable values of thermodynamic and chemical parameters used to drive the model. Additionally, the TROPOMI NO_2 retrievals were adjusted with a 40% increase to approximate findings from ground-based Max-DOAS studies suggesting that TROPOMI NO_2 values are underestimated. Conversely, a 40% decrease was applied to approximate studies indicating that co-emitted black carbon (BC), dust, and other absorbing aerosols cause TROPOMI NO_2 values to be overestimated. Furthermore, TROPOMI NO_2 retrievals were also evaluated with a lower error cutoff for background noise, set at (1×10^{15} molecules/cm^2). These constraints filter out TROPOMI NO_2 values that are not physically realistic given the in-situ environmental conditions. We err on the side of being conservative, and would rather only work with data that we consider sufficiently trustworthy. Consequently, the uncertainty in the computed emissions output is consistently observed to be narrower than the uncertainties in the TROPOMI NO_2 inputs.

In Section 3.1, Figure 5 plots the distribution of monthly NO_x/NO_2 over grids from different sources, how many facilities were counted for each emission source?

Thank you for your insightful comment regarding the number of facilities counted for each emission source in Section 3.1, Figure 5.

In our analysis, we utilized location data from pollutant sources provided by the Pollutant Discharge Permit Management Information Platform of the Ministry of Ecology and Environment to quantify the distribution of monthly NO_x/NO_2 over grids of five different sources. Specifically, the number of facilities counted for each emission source is as follows: 1) Power Plants: We included data from 163 power plants, which were identified as significant point sources of NO_x emissions (Beirle et al., 2021). 2) Industrial Facilities: This category encompasses 109 steel and iron factories and 212 cement factories. 3) Residential and Commercial: This category includes emissions from 71 biomass burning facilities and 243 heat and production supply facilities, with data aggregated from urban and suburban areas (Jones et al., 2023).

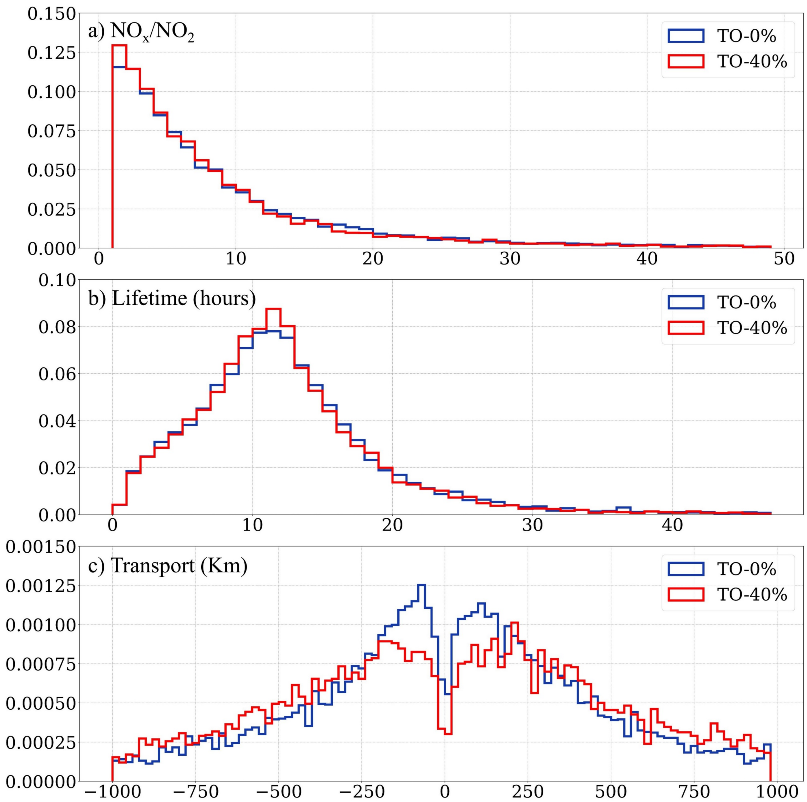
Response Table 2. The number of facilities counted for each emission source in Section 3.1, Figure 5.

	Power Plants	Steel and Iron Factories	Cement Factories	Biomass Burning	Heat Production and supply
Number	163	109	212	71	243

In Section 3.2, a key aspect to consider is the performance of the fitting process and the resulting errors in each parameter, as this is essential for assessing the reliability of the MCMFE framework. Given that the fitting incorporates all observations, does it remain unbiased across different months and grid cells?

In addition to the sensitivity analysis of the initial assumptions regarding NO_x emissions, a robustness test of the mass-conserving flexible emissions inversion was also conducted. We have conducted a comprehensive sensitivity analysis to assess the robustness of our methods. This analysis is done in much greater depth and detail in a a separate manuscript that which is currently under review in Remote Sensing of Environment (http://ssrn.com/abstract=4984749). In this manuscript under review at the present time, we have assumed an additional case in which the TROPOMI NO_2 observed column values include uncertainties at the extreme upper and lower bounds of their ±40% uncertainty range respectively, and use these to re-compute the fitting factors and subsequent emissions. When the TROPOMI NO_2 column values were adjusted, all related factors were simultaneously modified. This set of uncertainty simulations was uniformly applied as the $TO_{40\%}$ case, where the NO_2 columns were multiplied by random perturbations ranging from 0.6 to 1.4. The coefficients were refit using these new values from TROPOMI and the same values from both INTAC and meteorology over the entire domain included in this work. The results are provided in Response Figure 2 and Response Table 3.

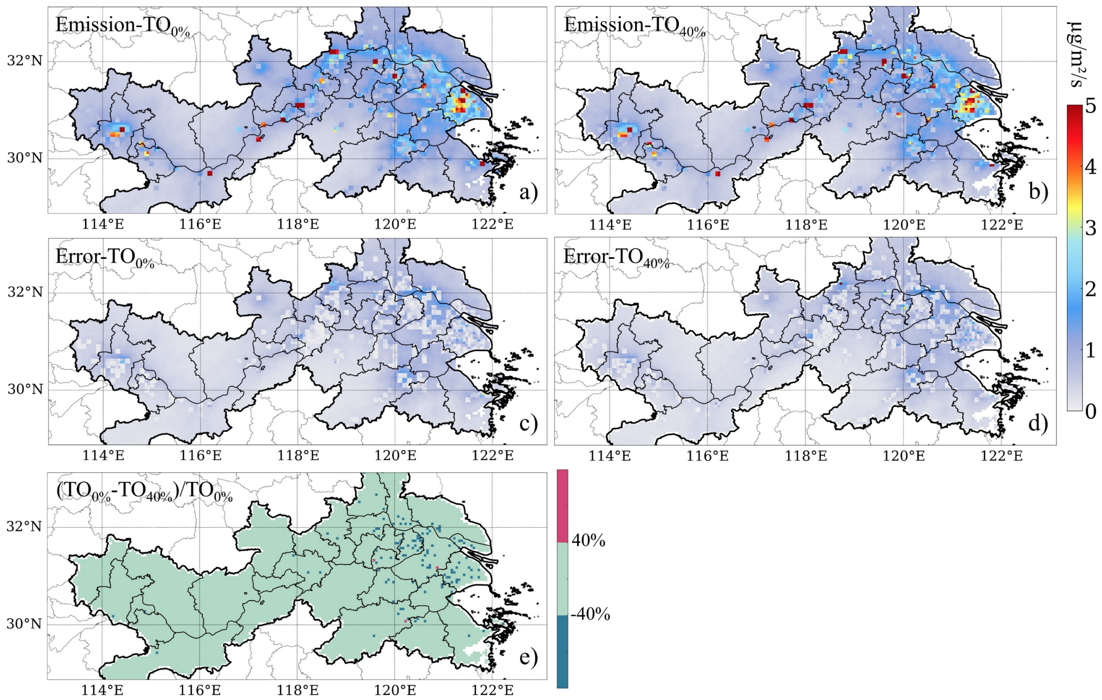
The first point to note is that the refit values of the chemical and thermodynamic terms yield uncertainty ranges that are smaller than their respective perturbations, indicating that the fits are stable to the uncertainty perturbation. At the 40% uncertainty level, there is a reduction in short-term transport and an increase in long-term transport. This again is consistent with the uncertainty in the a priori emissions estimate. However, accounting for the net uncertainties (i.e., including the buffering effects of the chemical and thermodynamic terms with the more uncertain transport term) our overall findings demonstrate that the mass-conserving flexible emissions inversion method provides robust inversion results (as illustrated in Response Figure 3). This is especially so when compared to the traditional wind speed and concentration gradient method, as observed in the transport term being the least stable to the uncertainty perturbation.



Response Figure 2. The distributions of three key coefficients obtained from $TO_{0\%}$ and $TO_{40\%}$: a) NO_x/NO_2, b) Lifetime [hours], c) Transport [km]

Response Table 3. The 20th, 50th,80th percentile ranges of three coefficients from $TO_{0\%}$ and $TO_{40\%}$.

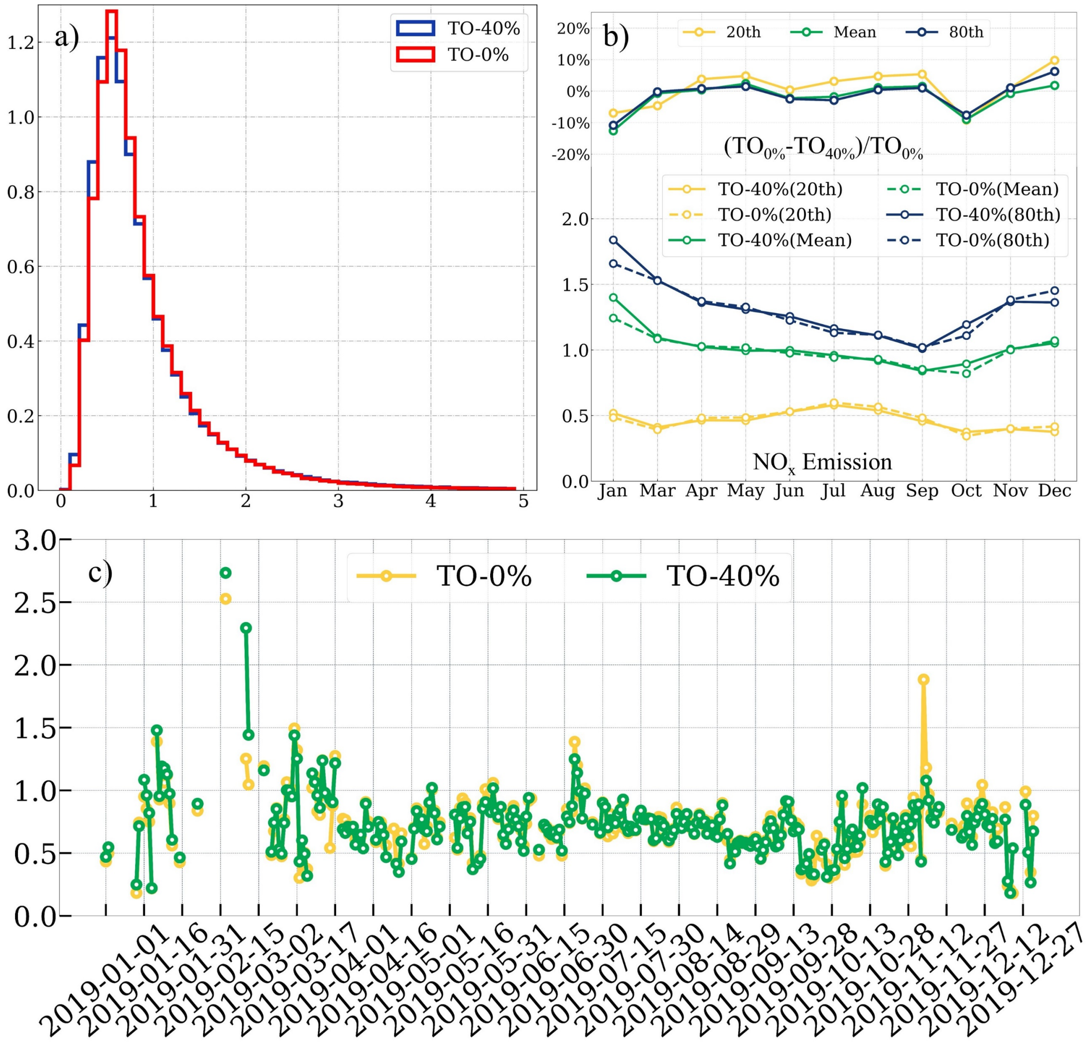
NO_x/NO_2	$TO_{0\%}$	$TO_{40\%}$
20%	2.76	2.63
50%	6.30	6.08
80%	14.6	13.4
Lifetime (days)	$TO_{0\%}$	$TO_{40\%}$
20%	0.29	0.30
50%	0.48	0.48
80%	0.68	0.68
Transport (km)	$TO_{0\%}$	$TO_{40\%}$
20%	-323.	-403.
50%	18.1	24.0
80%	351.	416.



Response Figure 3. The TROPOMI NO_2 column data is perturbated by a random factor 40% herein called [$TO_{40\%}$] to represent its range of uncertainty. The results are displayed for annual mean of emissions of a) $TO_{0\%}$, b) $TO_{40\%}$; and annual mean of error of c) $TO_{0\%}$, d) $TO_{40\%}$; and the annual mean of e) $(TO_{0\%}-TO_{40\%})/TO_{0\%}$

It was observed that 93% of the daily grid cells exhibited a ratio of [($TO_{0\%}$-$TO_{40\%}$)/$TO_{40\%}$] within ±40%. The spatial and temporal 20th and 80th percentile ranges of NO_x emissions for the $TO_{0\%}$ and $TO_{40\%}$ scenarios were 0.47 to 1.25 μg/m²/s and 0.45 to 1.25 μg/m²/s, respectively.

Overall, the day-by-day and grid-by-grid NO_x emission ranges are quite similar in both cases, as illustrated in Response Figure 4. On a month-by-month basis, the deviations for nearly all months are within ±10% (February is excluded due to missing TROPOMI data in 2019 for region 2). January, October, and December exhibit the largest deviations, with $TO_{40\%}$ values consistently lower than $TO_{0\%}$ across all percentile ranges in January and October. Furthermore, the day-by-day spatial median values across different cases exhibit only slight variations. These findings indicate that changes in the driving factors (α_1, α_2 and α_3) between the different NO_2 column loading scenarios are generally smooth and consistent, providing redundancy and being significantly influenced by the a priori emissions used in the fitting process. The constraints on the physically realistic values of α_1 and α_2, along with the constant use of INTAC, create a negative feedback loop affecting the relationship between NO_2 column changes and the final emissions products. This is consistent with the observed computed emissions and their differences.



Response Figure 4. a) The PDF of NO_x emissions [µg/m²/s] over all individual days and grids of $TO_{0\%}$ and $TO_{40\%}$; b) The 20th, mean and 80th values across different months of $TO_{0\%}$ and $TO_{40\%}$; c) The time series of the spatial median values of $TO_{0\%}$ and $TO_{40\%}$ for whole year.

In Section 3.4, Figure 8 plots have the unit of (Kton yr^{-1} $cell^{-1}$), however, the size of a cell is not mentioned in the context.

Thank you for your observation regarding the units and cell sizes in Section 3.4, Figure 8. The units of (Kt yr^{-1} $cell^{-1}$) represent the emissions per cell per year. We acknowledge that the size of each cell was not explicitly mentioned in the context. The actual grid is given to be 0.05°x0.05°, which means that the cells in our grid have varying areas due to differences in latitude and longitude, which affects the computation of total emissions of the whole year.

To address this, we have now included a detailed description of the grid cell sizes in the revised manuscript. “The grid cells are defined by a latitude-longitude grid with a resolution of 0.05ºx0.05º, meaning that area of each cell varies with latitude. This variation is accounted for in emission calculations to ensure accurate representation of emissions per unit area.”

Did the authors use MEIC emissions data or assume monthly invariant emissions?

We have reorganized the paragraph of section 2.3 in the following way to be clearer. The fact is that the MEIC emissions data themselves assume monthly invariant emissions, so we decided to not impose additional variation. Please see the details below:

“The assumptions regarding NO_x emission datasets in the initial step applied are harmonized using multi-source heterogenous data, developed by the MEIC (Multi-resolution Emission Inventory for China) team (Huang et al., 2012, 2021; Kang et al., 2016; Liu et al., 2016; Zheng et al., 2021; Zhou et al., 2017, 2021), in collaboration with various scientific research institutions. This dataset is referred to as the high-resolution INTegrated emission inventory of Air pollutants for China (INTAC), which is highlighted in purple in Figure 3. The original INTAC emissions are quantified in units of Mg/grid/month, with a temporal resolution of one month and a spatial resolution of 0.1° x 0.1°, for the year 2017. It is important to note that higher resolution inventories, such as the 1-km resolution inventory developed by the MEIC team at Tsinghua University, is also available (Zheng et al., 2021). However, the 1-km inventory is for 2013, and the INTAC inventory we utilized offers the highest resolution available which is the closest temporal match to 2019 TROPOMI data. This dataset covers mainland China and includes emissions from eight sectors: power, industry, residential, transportation, agriculture, solvent use, shipping, and open biomass burning(Wu et al., 2024). To align the resolution of the original INTAC Inventory with that of TROPOMI grids, we undertake several processing steps: 1) The units are converted from Mg/grid/month to $\mu g/m^2/s$ as the first step, due to the varying areas of each longitude-latitude grid. 2) Next, the INTAC inventory is adjusted to a 0.05°x0.05° grid using the nearest neighbor method. 3) Finally, we assume that the monthly emissions remain constant on a day-to-day basis. To ensure that the values used do not fall within the error range of the TROPOMI sensor (i.e., noise), values below 0.2 $\mu g/m^2/s$ are designated as NaN and are not considered further in this study."

Minor comments:

Line 215 and line 338, "densitiy" should be "density".

Thank you, we have made this modification.

Line 127, "boarder" should be "border".

Thank you, it has been modified.

Line 232, typo error "heat productin and supportion". Line 234, "form" should be "from".

Thank you for helping us improve this!

Line 262, typo error "reated".

Thank you, it has been modified.

Line 283, typo error "Qingdai".

Thank you for catching this. We have fixed it.

Line 363, typo error "differnces".

Thank you again.

Line 289 and 424: However -> Moreover

Thank you, it has been modified.

Line 360, 380 and 388: differences -> the differences

This has been updated.

Line 18: Kton/year -> Kt/year. Please ensure that units are consistently presented and aligns with standard publication practices.

All units are not consistent. Thank you for your help with our communication.

Please use hyphen (-) in the words separated by two lines or change the format.

Thank you, it has been modified.