Response to reviews

We thank all the reviewers for spending time on the paper improvement. Below, we provide a point-by-point response to the comments and we list the related changes made to the manuscript. The original review text is indicated in italic blue font and the response in regular black font. New text appearing in the revised paper is indicated by a red color.

Reviewer Mclinden,

This is a well written paper, and easy to follow. But it seems odd to me that conclusion (anthropogenic SO_2 emissions in India are only half what we thought) is really just mentioned in passing. Also, there is no validation of the results... comparisons are made with databases that differ by this factor of two, but that is it. The modifications to the flux-divergence methodology as previously employed are such that they require validation in their own right. Furthermore, when one obtains results that contradict previous studies it behooves the authors to provide some rationale as to why this is, and what could be behind the bias.

Information on the Indian SO_2 emissions is sparse, which limits the validation of the emissions obtained. We also do not have access to the in-situ measurements of India SO_2 surface concentrations for recent years. Therefore, we limited our comparisons to bottom-up inventories and the satellite-based emission estimates of Fioletov et al. (2023). Since our resulting emissions are about 50% lower than those of previous results, i.e. EDGAR v8, CAMS-GLOB-ANT v5.3 and estimation of Fioletov et al. (2023), we provide additional evidence to support our results in the revised manuscript.

In the bottom-up emission inventories, i.e. the recent versions of EDGAR and CAMS-GLOB-ANT, the country-total Indian SO_2 anthropogenic emissions are in the range of 10-14 Tg/year. According to the CAMS-GLOB-ANT v5.3 inventory India emitted approximately 11 Tg/year in 2023. However, we found that the CAMS model-simulated SO_2 columns, which are driven by the CAMS-GLOB-ANT v5.3, are much higher by a factor of 2 than TROPOMI SO_2 measurements in recent years (see the comparison in 2023 in Fig. 9 and below). Considering the good data quality of TROPOMI observations (Theys et al., 2021; De Smedt et al., 2021) and assuming the CAMS model has a good performance, we attribute the higher simulation results for a large part to the overestimation of the model input emissions.

It is important to note that if we use a constant SO_2 lifetime of 6 hours, this will result in an SO_2 emission total of approximately 12.0 Tg/year for India, aligning well with the estimated emissions of bottom-up inventories. This information is now added in the revised manuscript from line 445. However, instead of taking a fixed lifetime, we have made an extra effort to derive the local SO_2 lifetime. We notice that the factors determining the SO_2 lifetime can vary dramatically both spatially and temporally, leading to a significant variability of the lifetime. Previous studies indicated a wide range of SO_2 lifetimes (about 0.6-2.6 days in Lee et al., 2011 and references therein) rather than a fixed value. Therefore, we assume that a constant lifetime is not the best choice to calculate emissions for the whole India.

We add the discussion started from line 445 in the revised manuscript:

We calculate Indian SO_2 emissions to be 5.2 Tg $year^{-1}$ using the SO_2 local lifetime, and 12.0 Tg $year^{-1}$ using a fixed 6-hour lifetime. The country-total emission obtained with a local lifetime are about 50% lower than the reported emissions in the most used bottom-up inventories, i.e. CAMS-GLOB-ANT and EDGAR. The CAMS-GLOB-ANT v5.3 inventory estimates that India emitted 11 Tg $year^{-1}$ SO_2 in 2023. However, the CAMS model simulated SO_2 densities, driven by CAMS-GLOB-ANT v5.3, are much higher by a factor of 2 than the TROPOMI measurements (see the comparison for 2023 in Fig. 9). Considering the good data quality of the TROPOMI observations (Theys et al., 2021; De Smedt et al., 2021) and assuming the CAMS model has a good performance, we attribute the higher simulation results mainly to a positive bias in the emissions that were input to the model.

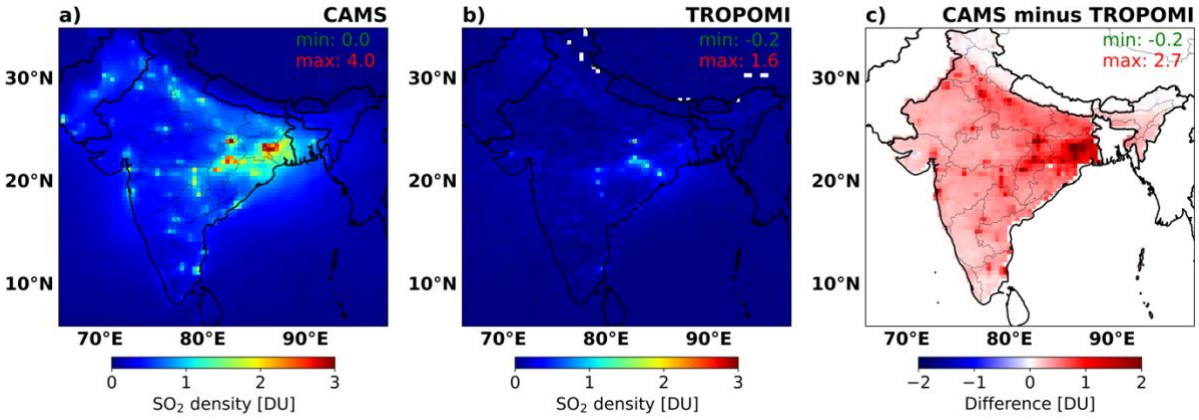

Figure 9. Indian SO_2 vertical column densities (VCDs) averaged in 2023 from (a) CAMS global composition forecast dataset (at about the overpass time of 6 UTC), and (b) TROPOMI Level-2 COBRA dataset. We integrate the TROPOMI observations to a resolution of 0.4° × 0.4°, the same as the CAMS datasets. (c) is the difference obtained by subtracting (b) from (a). The data of the

same days are used for comparison. The CAMS SO_2 density with total cloud coverage larger than 30% are excluded from the averaging.

While I am not convinced that the flux-divergence method is the best approach for SO_2 emissions as its strength is for extended area sources (e.g., urban NOx emissions), and SO_2 comes from a collection of point sources (albeit occasionally in fairly close proximity), it is worthwhile to explore its effectiveness. Retrieving emissions on a grid smears out the emissions which usually can be geolocated to within a couple km, and makes getting the total for a facility sometimes challenging since one must figure out which grid boxes need to be summed over.

The divergence method is fast and no model or other prior knowledge is needed for the emission derivation. The method does not need clearly isolated plumes, and it works also for areas with a mix of emissions (highways, towns, industry, power plants close together). The daily estimates from the divergence method and a longer time period of observations are needed. Up to now, the flux-divergence method has been proved to be effective for point source of NO_x (Beirle et al., 2019), CH_4 (Liu et al., 2021) and CO_2 (Hakkarainen et al., 2022). In our study we explore the feasibility for SO_2 emissions and have discussed the spreading effect from the divergence method. One of our major efforts is to reduce the spreading caused by the divergence calculation. |With this improvement, the determination of the location of the SO_2 point sources improved.

The authors attempt to calculate the lifetime considering simple physical (dry deposition) and first order chemical (via SO_2 + OH) loss, and these individual lifetime are combined. As an aside, why is only dry deposition included? Adding it would not impact my argument below, but it seems like wet deposition should be considered to be consistent with the approach chosen by the authors. Dry deposition lifetime were quite long, >60 hours, and thus do not have a huge impact on the combined lifetime. Chemical lifetime were calculated using an OH field (τ = 1/[k [OH]) from the ECMWF CAMS forecast model, and are roughly 20 hours, varying in space and time. The combined lifetime is something like 15 hours.

In our study, we calculate the monthly mean SO_2 effective lifetime under the PBL height, as the major amount of SO_2 is concentrated inside the PBL. As a major oxidant in the troposphere (Crutzen, 1973; Logan et al., 1981), the OH concentration is high in India (Lelieveld et al., 2004; Lelieveld et al., 2016).Therefore, we assume the OH oxidation plays the most important role in determining the SO_2 lifetime. The SO_2 dry deposition, which occurs at the same time, has indeed a secondary influence on the lifetime (Chin et al., 1996). The SO_2 wet deposition

and other chemical reactions occurring in the cloud's droplets are less important (Smith and Jeffrey, 1975; Chin et al., 1996; Qu et al., 2019) compared to the OH oxidation and SO_2 dry deposition in terms of the monthly mean lifetime, especially since we use only cloud-free observations. In this case, we suggest that the SO_2 monthly mean lifetime will not change significantly even we involve the wet deposition and other chemical reactions. (See the answer of Point 2 below for more information.)

My concerns regarding this lifetime issue are laid out here:

1.Using the OH field from ECMWF CAMS is not appropriate as the spatial resolution is 0.4 x 0.4 degrees (~40 km), and thus represent an average OH value, with half or more of the averaging area (on average; including the upwind portion) representing background values and chemistry. What is relevant is OH in the plume where the bulk of the SO_2 is, and OH at the plume core, plume edge, and background will all be much different. Using different models with comparable resolution to look at differences does not help in this regard. If one wants to use model OH then something like a plume-following chemical box model is the most appropriate choice.

Just as the reviewer mentioned, the variation of the lifetime influencing factors should be considered. That is why we calculate the lifetime on each grid-cell. Solving the varied lifetime both in and out of the plume would be the way forward in the future. In this paper, we have made a first step by using CAMS output, which is indeed coarse but is already showing that concentrations and lifetimes can vary spatially and temporally and differ from simplified constant lifetime assumptions. We think it is a good start to derive the non-constant local lifetime for emission estimation.

Additionally, the OH changing within the plume is not fully ignored in our method although the resolution of the model grid is relatively coarse. To support this statement, we calculate the distance that is possible for SO_2 transportation according to the simple exponential decay function Eq. (1) and distance calculation function Eq. (2)

$$C = C_0 e^{-\frac{t}{\tau}}, \quad (1)$$

$$S = W\ t \quad (2)$$

In the given equations, C represents the SO_2 concentration after it has decayed over a time t, given a lifetime τ and an initial concentration C_0. S denotes the distance travelled by the plume

over the time *t* at wind speed *W*. If we assume the initial concentration C_0 is 1 and *C* is 10%, the possible distance at different wind speeds and lifetime conditions are listed below.

	τ=6h	τ=15h
W=1m/s	S=49.8 km	S=124.3 km
W=2m/s	S=99.6 km	S=248.7 km
W=3m/s	S=149.4 km	S=373.0 km
W=4m/s	S=199.2 km	S=497.4 km

Based on the table, we know that the SO_2 plume can cover more than two model grids with a resolution of 0.4º (about 40 km) even under very low wind conditions (2 m/s) and a short SO_2 lifetime (6 hours). Therefore, the OH change along the plume is considered on the resolution of our grid.

2.In previous applications of the flux-divergence method, the lifetime was derived using the data itself (including and especially in the original formulation by Beirle et al. (2019) and this seems like the obvious method to employ here. Complications such as non-linear chemistry will then be accounted for. There is sufficient signal to tease out some spatial and seasonal differences from the TROPOMI data itself. At the very least the authors should have validated their calculated lifetimes using the data itself!

In the background, without local emissions, we expect that the negative values from the flux divergence are cancelled out by the positive lifetime term. Therefore, the lifetime is optimised when this cancellation occurs in the divergence data. Figure S8 with fixed 6h lifetime seems to suggest a higher positive background as compared to Figure 6 with local, which indicates that the local lifetime is more realistic.

We further checked the averaged lifetime by deriving a new monthly mean SO_2 lifetime ($\bar{\tau}$) from the CAMS model by considering all SO_2 consuming processes and all kinds of sink according to Eq. (3),

$$\bar{\tau} = \frac{C}{E}, \tag{3}$$

With *C* being the SO_2 concentration and *E* the SO_2 emission rate. We sum *M* and *E* for each month covering entire India to derive a monthly mean $\bar{\tau}$ averaged for the whole India. Fig. 2

shows the monthly $\bar{\tau}$ in 2019-2020 and 2022-2023 based on the CAMS model. The lowest lifetime is in summer, around 9.5 hours on average, while the longest lifetime is in winter, around 25.5 hours on average. The lifetime in spring and autumn is comparable, around 19 hours on average. The noticeable monthly/seasonal variation of lifetime align well with our calculations based on the OH oxidation and SO_2 dry deposition. This indicates that our calculated SO_2 lifetime will not change significantly even if wet deposition and other chemical reactions are considered now. Additionally, it is important to note that this model-intrinsic SO_2 lifetime of each month consistently exceeds 7 hours. It is evidence that a fixed lifetime is not the best choice for the emission estimation for the whole India.

In addition, we set a closed loop validation to validate the calculated SO_2 lifetime from CAMS model in Sec. 3.4 in the paper. This validation shows the emissions derived from the SO_2 lifetimes are comparable with the model input emissions, indicating that the lifetime we derive are in line with the model. Note that we account for the high uncertainty of the lifetime in the estimated error of the final emissions.

We add section 3.2.4 in the revised manuscript:

The monthly mean SO_2 effective lifetime is calculated based on OH oxidation and the SO_2 dry deposition. We assume negligible influence on lifetime from SO_2 wet deposition and other chemical reactions occurring in the cloud's droplets in terms of monthly mean lifetime, especially since we use only cloud-free observations. To show this, we derive a new monthly mean SO_2 lifetime ($\bar{\tau}$) from the CAMS model by considering all SO_2 producing processes and all kinds of sink according to Eq. (6),

$$\bar{\tau} = \frac{C}{E}, \tag{6}$$

with C being the total SO_2 concentration and E the total SO_2 emissions. We sum both the concentrations and the emissions of the model for each month covering the entire India to derive a monthly mean averaged C and E for the whole India. Fig. 2 shows the monthly $\bar{\tau}$ in 2019-2020 and 2022-2023 based on the CAMS model. This model-intrinsic SO_2 lifetime of each month consistently exceeds 7 hours. The lowest lifetime is in summer, around 9.5 hours on average, while the longest lifetime is in winter, around 25.5 hours on average. The lifetime in spring and autumn is comparable, around 19 hours on average. Note that the CAMS model

includes both dry and wet deposition of SO_2. The noticeable monthly/seasonal variation of lifetime align well with our calculations based on the OH oxidation and SO_2 dry deposition, indicating our calculated SO_2 lifetime will not change significantly even if wet deposition and other chemical reactions are considered. At the same time, we see a large variation both spatially as in the average from month to month. Therefore, we will use the monthly-averaged local lifetime from here on.

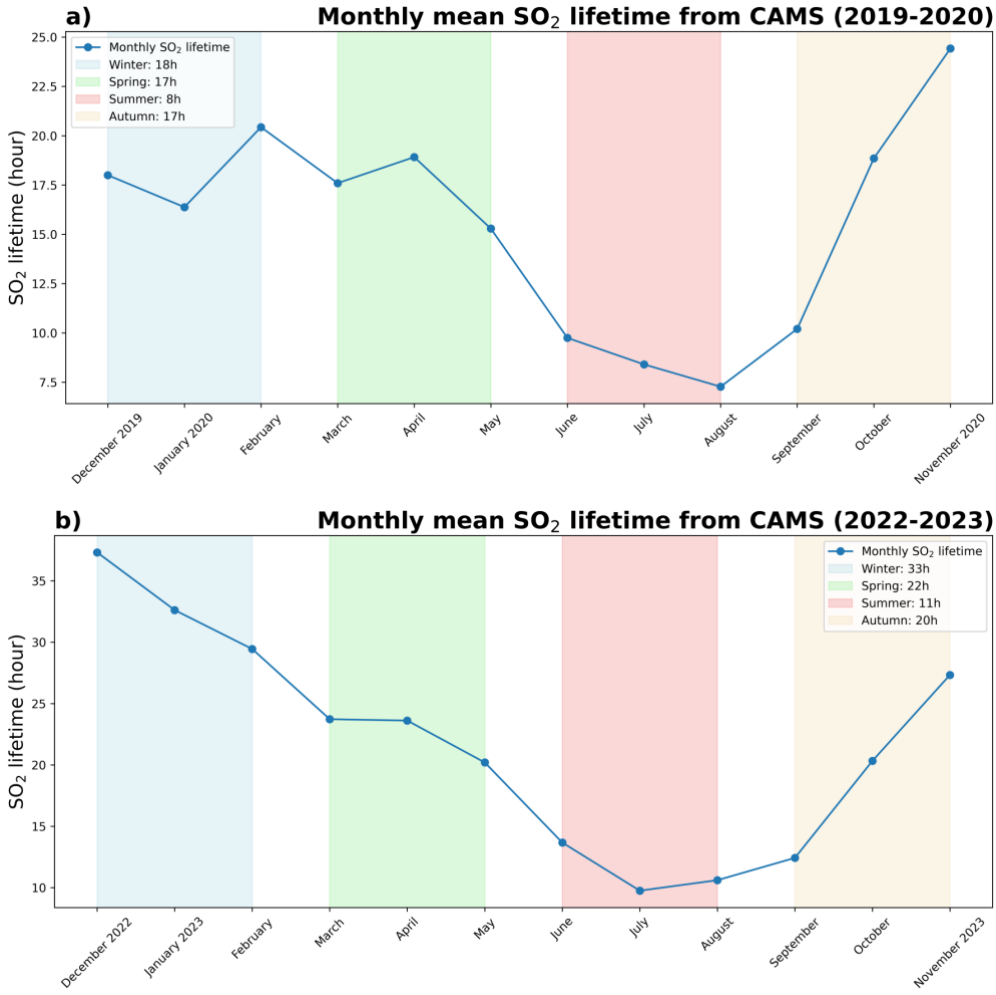

Figure 2. Monthly averaged SO_2 lifetime in India for (a) 2019-2020 and (b) 2022-2023. The lifetime is calculated by accounting for all SO_2 producing processes and all kinds of sink in the CAMS model.

3.A simple analysis of the plumes themselves in the satellite data suggests that the effective lifetime is shorter than 15 hours. The authors do not ever show the actual TROPOMI SO2 data in their paper (there is one panel in the supplement), which seems strange considering it is the basis for the emissions calculations. Shown below in Figure 1 is an OMI SO2 VCD averaged over 2014-2017 (this is all I had handy; TROPOMI will look similar, but hot spots will appear sharper due to its higher spatial resolution). The dots are the larger SO2 sources. These are unpublished, diagnostic figures from the same EMG (exponentially modified Gaussian) method as published in Fioletov et al. (2016, 2018), McLinden et al. (2016) and later papers by that group. The left panel is the mean VCD (minus a slowly varying background bias, an artefact of the method, as discussed in the references above). The right is the reconstruction of the satellite data assuming a 6 hour effective lifetime.

The TROPOMI SO_2 observations are shown in the new Figure 9b of the updated manuscript.

It is important to mention the purpose of this study, which is to derive a SO_2 emission inventory covering the whole India. The lifetime we use is the effective lifetime averaged for each grid cell of the area instead of averaged within the plume of the individual source. The EMG method is also relying on an effective lifetime since the lifetime within the plume is considered constant and averaged over a certain time period. Both effective lifetimes are not necessarily defined in the same way and both are approximations of the real lifetime.

In the EMG method the effective lifetime is derived under the assumption that the SO_2 lifetime is constant within a SO_2 plume (Fioletov et al., 2015). However, the factors influencing the SO_2 lifetimes, such as OH concentration, can vary significantly along the plume. For example, Krol et al. (2024) found OH concentration is lowest near the source and gradually increase with distance. They found the lifetime of NO_2 and NO_x around the studied coal-fired power station are longest near the source and gradually shorten with increasing distance. Although this study focuses on NO_x instead of SO_2, it provides insights that the lifetime varies spatially due to the spatial variation of influencing factors. Since both NO_2 and SO_2 are converted through reactions with OH, this study provides information also relevant for SO_2.

4.For EMG methods similar to that employed in my point 3, de Foy et al. (2015) argued that there is a dispersion component to the effective lifetime, and it reduces the overall lifetime relative to the chemical+physical lifetime. In my understanding of the flux divergence method, diffusion is not accounted for and so it seems reasonable that the effective lifetime appropriate

for this method may also have a dispersive component. Further, in the case of an isolated point source, as it the case for many here, the two methods are virtually the same with the main difference being that the EMG method of Fioletov et al. assumes a plume shape, and the flux divergence method does not constrain the plume, but derives emissions on a grid. If this case, the physical interpretation and value of the effective lifetime should be very similar, and I would argue that my Figure 1 above is the most compelling argument for a shorter lifetime. See Vindhyachal in particular (the most northerly isolated hotspot). If the physical interpretation of the lifetime is not the same, then one would need to look at how the different methods perform when emissions are well know, such as from CEMS or other direct measurements. The EMG method of Fioletov et al has been extensively validated against such measurements in the US, Europe, and for volcanic sources as well.

The divergence method is based on local mass balance and therefore it includes diffusion. The dispersive component in the divergence method leads to spreading of the signal (noise on the divergence) but it doesn't affect its lifetime calculation. By averaging over a longer time period, the effect is strongly reduced. In future studies we hope also to have results for the US or Europe, allowing a more extensive validation for point sources.

5.Application to India is more difficult since there is little reference data to validate against. In such cases it is useful to apply the method to other locations where such data is available. For example, in the US, there is CEMS data to compare against. The authors even mention it can applied anywhere. If one wants their results to be received with confidence, this is an essential step.

The high and evenly distributed SO_2 point-source emissions in India (Fioletov et al., 2023) provides an ideal base for developing and applying the divergence method. This is because the divergence method particularly works well for identifying strong point-source emissions, like Indian SO_2 emissions. We also did the test for Europe and US. The emission maps of these clean regions mix with more noise compared to the India case, making it not a good a base for improving the divergence method.

The Indian high but not clear SO_2 emissions itself is also an important reason why we focus on India. The Indian SO_2 emissions change rapidly. This study aims to provide an updated SO_2

budget for India for recent years. In future research, we certainly would like to also focus on regions where more reference data is available, like China.

Additional comments:

Presumably there is nothing that unusual about Indian OH levels (the lifetime as derived here, to first order only depends on k and [OH]), and thus had the authors preformed a global analysis they would have found similar results (ie, ~a factor of two smaller emissions as compared to current, best emissions values) everywhere. Even just for India this has large implications, but for the globe this would lead us to rethink the SO_2 budget.

In the recent ATMOS conference in 2024, we learned that the latest global top-down SO_2 emission estimation from Adrian Jost and Steffen Beirle in Max Planck Institute for Chemistry is 30% lower than the estimation from Fioletov et al. (2023). The online abstract might be available after August 30, 2024. In the EGU conference 2024, the Indian researcher Pramod Kumar working at Laboratoire des Sciences du Climat et l'Environnement, France estimates that Indian SO_2 emissions are approximately 5.0 Tg $year^{-1}$ in recent years. The work is now on writing. We notice the previous studies seldom provide conclusions for the present or future SO_2 emissions in India. And this study aims to provide more information of the potential Indian SO_2 emissions in recent years, like what the other researchers are also doing now. Since (1) the climate zone of India is very different from other regions in the world, (2) bottom-up emissions have a high uncertainty in India, and we have estimated a high uncertainty on our own emissions, we will not draw any conclusion about the SO_2 budget of the entire world.

Line 382: "But we see a noticeable smoothing effect and an overall positive bias on emissions estimated with a fixed 6-hour lifetime compared to the emissions estimated with a local, variable lifetime, especially around the source location"

But given the colour-scale, and comparing Figure S8 with Figure 5, this is exactly what one would expect when you ~double the emissions... there should be more red because all values are larger for the 6 hour lifetime, and thus it would appear to be smoother. The only way this line of reasoning could be reliably used is to compare an isolated source, and then normalize them their peak value. Eyeballing Vindhyachal, the large, isolated point source in my Figure 2, below, the effect, even with the 2x emissions, appears marginal. Bigger picture, this type of analysis is indirect at best... again, why not use the actual data to deduce the lifetime?

We have made efforts to ensure that the emission signal remains concentrated at the source rather than spreading to the surrounding areas. Therefore, after filtering the noise, we expect the emission signal can represent the point source emissions without large spreading. We have indeed considered the effect of the color scale. Therefore, as recommended by the reviewer, we have changed the color bar range in Fig. 6 and Fig. S8 in the revised manuscript by adapting the color range to the maximum value. The general positive bias still exists in the "6-hour" emission map after adapting the color bar (see the comparison figure of a zoom-in area below). The 6-hour lifetime is deduced from the actual measurement (Fioletov et al., 2015).

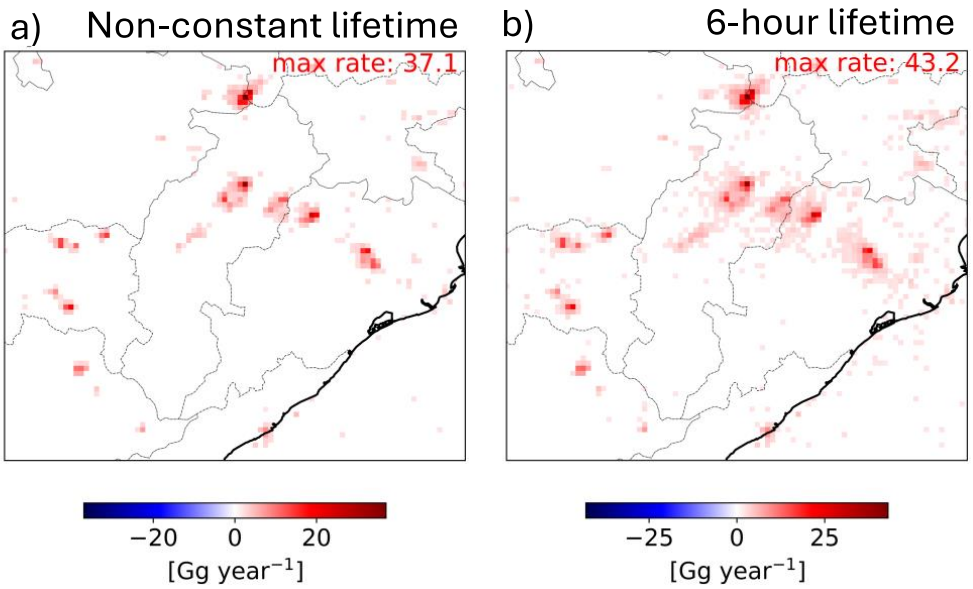

Figure R1. SO_2 emissions in a zoom-in area with dense SO_2 point sources in India. (a) is the same as Fig. 6d in the paper. It represents the SO_2 emissions calculated based on non-constant SO_2 lifetime (b) the same is Fig. S8d in the supplementary information. It represents the SO_2 emissions calculated based on a constant 6-hour lifetime.

Reviewer 2,

This paper uses a flux-divergence method to estimate monthly SO_2 emissions in India based on satellite observations. The paper is well organized but I have a little concern about the calculation of SO_2 lifetimes, which is crucial to the final emission estimation. The authors declaimed that the satellite overpasses at noon time and they used SO_2 under cloud-free conditions, so they considered only the gas phase loss of SO_2. It may be true that SO_2 is mainly removed through the reaction with OH as the satellite overpasses, but SO_2 lifetime is as long as tens of hours, during this time, SO_2 is removed mostly by the liquid phase or the heterogeneous reactions, both of which is faster and more efficient than the gas phase reaction. So I'm not sure if it is appropriate to consider only gas phase loss of SO_2 in the SO_2 lifetime estimation.

In our study, we calculate the monthly mean SO_2 effective lifetime under the PBL height, as the major amount of SO_2 is concentrated below the PBL. As a major oxidant in troposphere (Crutzen, 1973; Logan et al., 1981), the OH concentration is high in India (Lelieveld et al., 2004; Lelieveld et al., 2016).We therefore assume the OH oxidation plays the most important role in determining the SO_2 lifetime. The SO_2 dry deposition, which occurs at the same time, has indeed a secondary influence on the lifetime (Chin et al., 1996). The SO_2 wet deposition and other chemical reactions occurring in the cloud's droplets are less important (Smith and Jeffrey, 1975; Chin et al., 1996; Qu et al., 2019) compared to the OH oxidation and SO_2 dry deposition in terms of the monthly mean lifetime, especially since we use only cloud-free observations. In this case, we suggest that the SO_2 monthly mean lifetime will not change significantly even we involve the wet deposition and other chemical reactions.

To show this, we derive a new monthly mean SO_2 lifetime ($\bar{\tau}$) from the CAMS model by considering all SO_2 producing processes and all kinds of sink according to Eq. 1,

$$\bar{\tau} = \frac{C}{E}, \quad (1)$$

With C being the SO_2 air mass and E the SO_2 emission rate. We sum C and E for each month covering the entire India to derive a monthly mean τ averaged for the whole India. Fig. 2 shows the monthly $\bar{\tau}$ in 2019-2020 and 2022-2023 based on the CAMS model. The lowest lifetime is in summer, around 9.5 hours on average, while the longest lifetime is in winter, around 25.5

hours on average. The lifetime in spring and autumn is comparable, around 19 hours on average. Note that the CAMS model includes both dry and wet deposition of SO_2. The noticeable monthly/seasonal variation of lifetime align well with our calculations based on the OH oxidation and SO_2 dry deposition. This indicates that our calculated SO_2 lifetime will not change significantly even if wet deposition and other chemical reactions are considered. Additionally, it is important to note that this model-intrinsic SO_2 lifetime of each month consistently exceeds 7 hours. The lifetime we derived here and shown in the paper also match the results ranging in 0.6-2.6 days of previous studies (Lee et al., 2011 and the papers therein).

In addition, we set a closed loop validation to validate the calculated SO_2 lifetime from CAMS model in Sec. 3.4 in paper. This validation shows the emissions derived from the SO_2 lifetimes are comparable with the model input emissions, indicating the lifetimes we derive are in line with the model. We also include the high uncertainty of these lifetimes in the estimated error of the final emissions.

We add section 3.2.4 started from line 231 in the revised manuscript:

The monthly mean SO_2 effective lifetime is calculated based on OH oxidation and the SO_2 dry deposition. We assume little influence on the lifetime from SO_2 wet deposition and other chemical reactions occurring in the cloud's droplets in terms of monthly mean lifetime, especially since we use only cloud-free observations. To show this, we derive a new monthly mean SO_2 lifetime ($\bar{\tau}$) from the CAMS model by considering all SO_2 consuming processes and all kinds of sink according to Eq. (6)

$$\bar{\tau} = \frac{C}{E}, \tag{6}$$

with C being the total SO_2 concentration and E the total SO_2 emissions. We sum both the concentrations and the emissions of the model for each month covering the entire India to derive a monthly mean averaged C and E for the whole India. Fig. 2 shows the monthly $\bar{\tau}$ in 2019-2020 and 2022-2023 based on the CAMS model. This model-intrinsic SO_2 lifetime of each month consistently exceeds 7 hours. The lowest lifetime is in summer, around 9.5 hours on average, while the longest lifetime is in winter, around 25.5 hours on average. The lifetime in spring and autumn is comparable, around 19 hours on average. Note that the CAMS model

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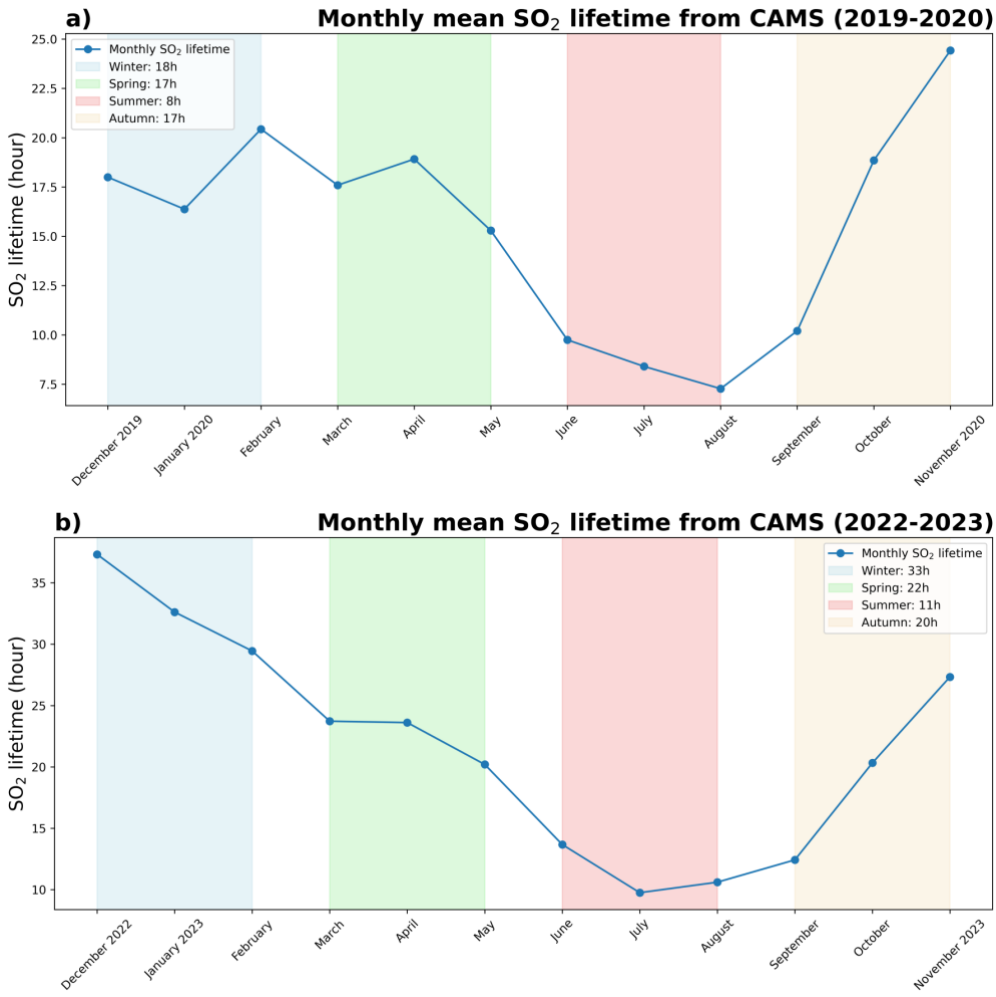

Figure 2. Monthly averaged SO_2 lifetime in India for (a) 2019-2020 and (b) 2022-2023. The lifetime is calculated by accounting for all SO_2 producing processes and all kinds of sink in the CAMS model.

The SO_2 emission estimated here is much less (even 50%) than other datasets, does this indicate that the SO_2 lifetime calculated in this study is too long?

Since our resulting emissions are half as low as those of some other studies, we provide additional evidence to support our results in the revised manuscript. In the bottom-up emission inventories, i.e. CAMS-GLOB-ANT and EDGAR, the estimation of Indian SO_2 anthropogenic emissions is in the range of 10-14 Tg/year. It is calculated that India emitted approximately 11 Tg/year according to the CAMS-GLOB-ANT v5.3 inventory in 2023. However, we found the CAMS model simulated SO_2 densities, which are driven by the CAMS-GLOB-ANT v5.3, are much higher by a factor of 2 than TROPOMI SO_2 measurements in recent years (see the comparison in 2023 in Fig. 9). Considering the good quality of the TROPOMI observations (Theys et al., 2021), we attribute the higher simulation results to the overestimation of the model input emissions.

We add the discussion started from line from 445 in the revised manuscript:

We calculate Indian SO_2 emissions to be 5.2 Tg $year^{-1}$ using the SO_2 local lifetime, and 12.0 Tg $year^{-1}$ using a fixed 6-hour lifetime. The country-total emission obtained with a local lifetime are about 50% lower than the reported emissions in the most used bottom-up inventories, i.e. CAMS-GLOB-ANT and EDGAR. The CAMS-GLOB-ANT v5.3 inventory estimates that India emitted 11 Tg $year^{-1}$ SO_2 in 2023. However, the CAMS model simulated SO_2 densities, driven by CAMS-GLOB-ANT v5.3, are much higher by a factor of 2 than the TROPOMI measurements (see the comparison for 2023 in Fig. 9). Considering the good data quality of the TROPOMI observations (Theys et al., 2021; De Smedt et al., 2021) and assuming the CAMS model has a good performance, we attribute the higher simulation results mainly to a positive bias in the emissions that were input to the model.

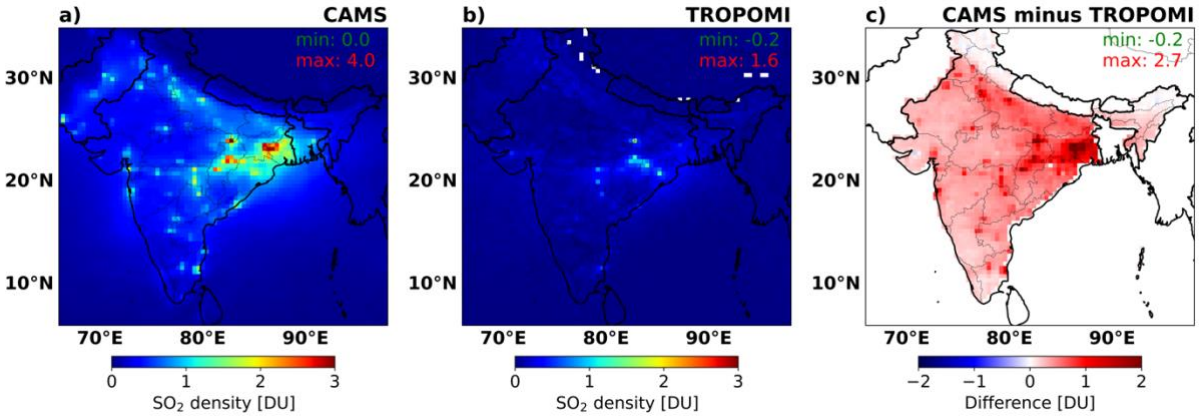

Figure 9. Indian SO_2 vertical column densities (VCDs) averaged in 2023 from (a) CAMS global composition forecast dataset, and (b) TROPOMI Level-2 COBRA dataset (at about the overpass time of 6 UTC). We integrate the TROPOMI observations to a resolution of 0.4° × 0.4°, the same as the CAMS datasets. (c) is the difference obtained by subtracting (b) from (a). The data of the same days are used for comparison The CAMS SO_2 density with total cloud coverage larger than 30% are excluded from the averaging.

And a minor comment, line 213: 'the SO_2 monthly dry deposition lifetime within the PBL height is calculated by dividing the PBL height by 0.4 cm s^{-1}', shouldn't it be '... dividing half the PBL height by 0.4 cm s^{-1} '?

Slinn et al. (1978) calculated the dry deposition lifetime within a layer by dividing the height of the layer by the dry deposition velocity. Similarly, we calculate the dry deposition lifetime within PBL by dividing the PBL height by the velocity of 0.4cm s^{-1}. We treat the PBL as a single layer here.

Reference

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