

We would like to thank reviewer 1 for his/her constructive comments and suggestions.

### General points:

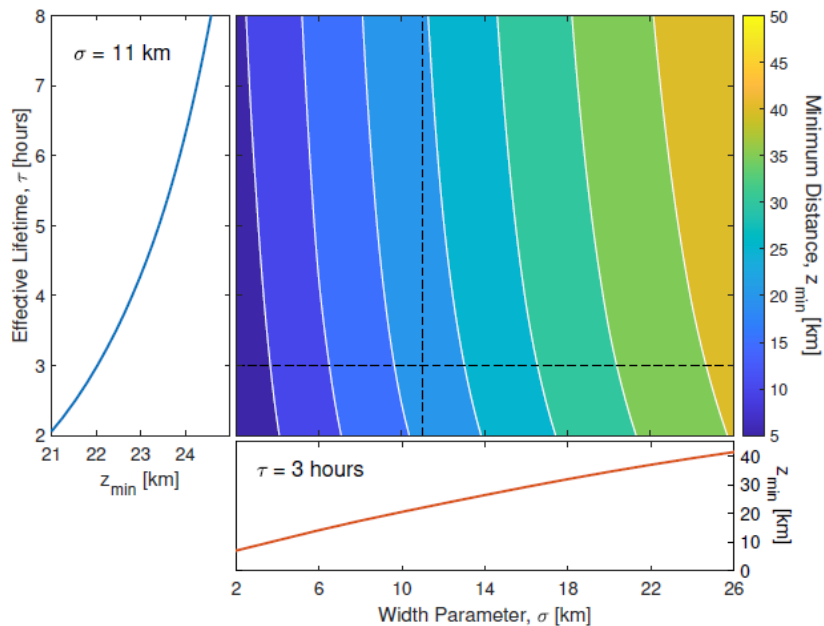
- 1. The manuscript needs to be thoroughly checked for the logic and for the English grammar. Many statements are unsupported or confusing. Many commas are misplaced, making sentences difficult to read and understand. Some sentences make no sense by themselves, but do make sense if joined to the next sentence. Many citations use parentheses incorrectly. Subscripts are sometimes missed.**

Response: The manuscript has been edited in detail by a native English speaker. Furthermore, we tried to make all requested changes. Instead of listing all the edits here, we point towards the author's tracked changes document in the next submission.

- 2. The authors assume an NO<sub>2</sub> lifetime of 4 hours when deriving emissions, but shouldn't this lifetime impact the footprint on the sources when comparing to the satellite data? With 5 m/s winds for example 4 hours of advection transports the NO<sub>x</sub> ca. 72 km, but the Gaussian smoothing used to produce Fig. 3 uses a  $\sigma$  of 1 grid cell, ca. 10km. Would more smoothing produce better SSIMs and more accurate inversion of the emissions?**

The footprints of a source is indeed linked to the lifetime of the emissions. But the lifetime is taken into account while creating the source-receptor relations, the  $Ax=B$  linear system, and thus the inversion. The inversion approach is the leading driver in producing accurate emission totals. The posteriori smoothing is only there to bridge the limitations of the method and instrument. The spatial limit to resolve 2 sources of similar size depends on the effective lifetime, the pixel size and meteorological factors such as typical diffusion etc. Of these the pixel size and lifetime are dominant at our  $\sim 5.5 \times 3.5 \text{ km}^2$  pixel limit. The pixel size combined with diffusion gives us a typical plume width of around 7 km (e.g.  $\sigma_{plume}^2 = \sigma_{plume}^2 + \sigma_{pixel}^2 + \sigma_{source\_size}^2$ ). This value varies depending on typical size of a source but most sources of NO<sub>2</sub> are limited in size (except for large mines etc, and one could argue a city can be one larger source). Based on McLinden et al., 2023 (to be submitted but showing an example figure here) a plume-width 7 km combined with a lifetime of 4 hours gives an effective resolvability limit of 15-20km, which for  $0.1 \times 0.1$  degrees source cells (e.g.  $\sim 10 \times 10 \text{ km}^2$ ) explains our choice for a sigma of 1 grid-cell. More smoothing can produce better results, but also reduces the observable details. SSIM should be seen more as a metric to judge

comparability, and not accuracy of the emissions, as the inventory emissions are not perfect either.



**Figure E2.** Minimum distance required to distinguish between two point sources as a function of plume width and effective lifetime. The line plots to the left and below show this quantity for  $\sigma = 11$  km and  $\tau = 3$  hours, respectively, and are cross-sections indicated by the white dashed line.

3. Related to this, the TROPOMI-derived emission uncertainty is stated to be 30–40% (p21, L428), but it isn't clear where these numbers come from. The text on p21 discusses many sources of uncertainty (including the lifetime issue), but how were these combined to make 30–40% and the error bars (whatever they are) in Fig. 8?

Response: The discussion on the uncertainties (lines 430-479) has been moved and extended to form its own section (2.2.2, from line 290 onward) which can be read in the updated version of the manuscript. Additionally a table (1) has been added to summarize the individual uncertainties/errors. Some further explanation has been given for the individual error terms, linking back to earlier sections in the manuscript where needed. The discussion section has also been shortened to account for the moved section.

4. p2, L39–49, and also p20, L403 onwards. I missed a discussion of the known problems of real-world emissions. For example, as cited in Oikonomakis et al. (2018), several studies showed a significant discrepancy (a factor of 2–4) in the NO<sub>x</sub> emissions from light-duty diesel vehicles between two driving cycles, and Anenberg et al. (2017) and found similar issues for heavy duty vehicles, indicating inadequacy of the testing procedures to capture real-world emissions. The diesel-gate

**scandal was also a good example of the limitations of emissions reporting (Jonson et al., 2017).**

Response: The reviewer nicely points out why there is a need for more validation and verification of the emission inventories using independent data streams. As the reviewer notes there can certainly be further “unknown unknowns” in the emission inventories. Independent verification, for example based on satellite observations, could more rapidly trace and reveal discrepancies. While the diesel-gate scandal is the obvious example, it is complicated to point out additional “unknown unknowns” without moving too much into speculation territory. We added a few sentences to the discussion on the importance of independent verification to trace and reveal potential “unknown unknowns”.

Added to introduction: Furthermore, accurate methods that allow for independent verification, can potentially be used to trace and reveal significant discrepancies in the current emission inventories. An example of a discrepancy in the past would be the diesel-gate scandal which was a good example of the limitations of emissions reporting (Jonson et al., 2017).

#### **Other points:**

- **p1, L12, Explain 100kt as percentage, so that the reader knows if this is a large or small number.**

Response: Changed 100kt to 75-100kt (<10%) at L12.

- **p1, L14, Add a year here so that the reader knows when this recovery was ‘projected’.**

Response: Changed line to “The recent projections for the inventory emissions of 2021 pointed to a recovery of the 2021 emissions towards pre-COVID19 levels this increase was not observed.”

- **p1, L17. It sounds odd that satellites help faster “projections”, since that term is usually reserved for future values. Re-phrase.**

Response: Changed sentence to “This again illustrates the value of having a consistent satellite based methodology for faster emission estimates to guide and check the conventional emission inventory reporting.”

- **p1, L18. Change meet to meets, or method to methods. Check such things throughout the manuscript.**

Response: As mentioned above, edited the document. Changed to “meets”.

- **p2, L2. Add also a more recent reference than Crutzen 1970.**

Response: Changed to Seinfeld and Pandis 2006.

- **(nit-picking I know): Say ‘largely’ primary. Some NO<sub>x</sub> is produced by lightning, and NO<sub>2</sub> is mainly produced from ozone reactions with primary NO.**

Response: Added “for the most part” L23 P2.

- **p2, L27. The units should be formatted correctly, not in italics. Also, be consistent. The unit on L28 has a space between g and m, whereas on L27 it doesn’t.**

Response: Formatted throughout document added a space at L27.

- **p2, L30. Give reference for the statement about acidification and eutrophication.**

Response: Added reference: Galloway et al., 2003.

- **p2, L36. There is no such thing as the ‘Geneva Convention ...’, in this context at least. The authors mean the Air Convention or CLRTAP equivalents, e.g. <https://unece.org/environment-policy/air>. ‘Nations’ is also not an appropriate reference; maybe ‘UN-ECE’ or similar.**

Response: Correct, realizing the drawbacks of the Overleaf editor. Changed accordingly.

- **p2, L44. The word projections confused me here and elsewhere. Usually the term is used for future scenarios, e.g. for 2030 or 2050. Here I think the authors just mean emission estimates.**

Response: Changed to “emission estimates”.

- **p3, L60. Give info on this ‘unprecedented horizontal resolution’, or refer to appropriate section for details.**

Response: changed to “with its unprecedented spatial resolution of 3.5 × 5.5 km<sup>2</sup>”

- **p3, L65. Refer to appropriate section for details of code and availability.**

Response: Removed "latest draft", links to code and availability are located in the code and data availability sections.

- **p3, L71. Tell the reader where this is 'described further'.**

Response: Removed line, added url to <https://space-emissions.net/>.

- **p3, L75 (also p7). What is Umweltbundesamt (UBA) for those not familiar with German institutions?**

Response: Added German Environment Agency.

- **p3, 1st paragraph. This section is somewhat repetitive of Sect. 1, and isn't really 'Methodology and Datasets'. Some is also repeated, or better placed, in Sect. 2.1.**

Response: Moved part of the section to the introduction, removed "The national inventory data is reported through the informative inventory report (IIR): For the case of Germany it is publicly available from its original source: " as it was repeated at a later point.

- **p4, L104. Where does this 300% number come from?**

Response: The 300% comes from:

[https://iir.umweltbundesamt.de/2023/general/uncertainty\\_evaluation/start](https://iir.umweltbundesamt.de/2023/general/uncertainty_evaluation/start) ;

*"Compared to other pollutants, NOx emission uncertainties are moderate. The national total has a 95% confidence interval of about -8.5% to +15.0% in 2021, which amounts to about 230kt of NOx. Interestingly, with NOx, the differences between the two approaches in uncertainty combination (EP and MC) are particularly visible. This is because of the highest contributing sector 3.D - Agricultural Soils, where emissions and uncertainties are high (> +300%) and, crucially, do not follow a normal distribution. Therefore, only the MC simulation, which takes the log-normal distribution of these emissions into account, correctly reflects this source, while the EP yields unrealistic high uncertainties at about 15.4% in both directions.*

- **p4, Sect. 2.1.1. Some of the text here is also more introductory material (e.g. L104 onwards) than technical description of the emission inventory.**

Response: Removed lines 104-108.

- **p4, L120. Change 'mol' to emissions (mol is a unit, not a quantity).**

Response: Changed to emissions.

- **.5, L124. NO<sub>x</sub> is a mixture of NO and NO<sub>2</sub>, so one needs to specify the assumed molecular weight associated with your 5 kt NO<sub>x</sub> per year figure, or state as e.g. kt(N) NO<sub>x</sub> per year.**

Response: Added ((NO<sub>2</sub>) to line 124.

- **p5, L130 onwards. Same issue with NO<sub>x</sub> units and emission amounts.**

Response: Added line to state same usage of kt (NO<sub>2</sub>) unit throughout the document when mentioning NO<sub>x</sub>.

- **p5, L151. What is ATBD?**

Response: Added "Algorithm Theoretical Basis Document".

- **p6, L166. Say 'well correlated with ...'; the sentence was difficult to read.**

Response: Rewritten as The TROPOMI NO<sub>2</sub> data correlate well when compared to ground-based MAX-DOAS and PANDORA instruments (Verhoelst et al., 2021) but tend to show an underestimation of the tropospheric column.

- **p6, L165–173. Various statistics are given concerning bias, but are the instruments being compared with (MAX-DOAS, PANDORA) free of bias themselves? Are some of the difference due to problems with these instruments?**

Response: The MAX-DOAS and PANDORA instruments are not completely free of bias themselves but typically have much lower uncertainties than the TROPOMI-NO<sub>2</sub> product as stated in Verhoelst et al., 2021.

- **p7, L205. Mangled 'from in the naive'?**

Response: Broken link, fixed accordingly L205, changed to "uses the TEMIS monthly L3 data product available at <https://www.temis.nl/>".

- **p10. The 'Column' term in Eqns. (2) and (7) looks very ugly. Use a symbol, as is done for all other terms. In any case, shouldn't this be VCD?**

Response: Changed to V, removed NO<sub>2</sub>/source indicator j.

- **p10, equations (3)–(6). Give in order of usage, thus  $\sigma_1$  before  $f(x, y)$ ,  $\lambda_1$  before  $g(y, s)$ .**

Response: Reordered

• **p10, L255. Where is  $\tau$  explained?**

Response: Its rewritten from equation 2, so stated in line 239. Made italic to make clearer.

• **p10, L258: 'Following Beirle et al. (2016) we assume a lifetime of about 4 hours( $\pm 25\%$ )'. I can't find the terms hour or lifetime in Beirle 2016, and that paper deals mainly with the stratosphere. Why didn't you use estimates of NO<sub>2</sub> lifetime from LOTOS-EUROS for Germany? Does the 25% estimate really capture the uncertainty here?**

Response: Note that a detailed answer to a similar question has been given in the response to Reviewer 2 (second question). To answer the specific question of this reviewer: the year stated for Beirle et al's manuscript was incorrect and should be 2019. A similar number is stated in Beirle et al's 2011 manuscript. Based on results in earlier studies (Goldberg et al., 2021, Fioletov et al., 2022, Beirle et al., 2011, Lange et al., 2022) on average the 25% estimate should hold. The results (Fig 6b/d) shown in the study by Lange et al., (2022) seem the most representative yet for our study, who give an average range of 3-5 for Paris/Northern Latitudes (49-56).

We did not use estimates of NO<sub>2</sub> lifetime from LOTOS-EUROS as there is currently no option within the model to directly write out lifetimes. We can look at earlier studies that used a tagging approach to label emissions from individual hours. An earlier study by Curier et al., 2014 did just that to study the source sector contribution of emissions from individual hours to the OMI NO<sub>2</sub> column at OMI overpass for several industrial regions in Europe. For the region somewhat representative of Germany (Benelux) the study states: "Approximately 50% of the modelled OMI signal results from NO<sub>x</sub> emissions in the 3 h prior to OMI overpass.". This statement holds for most the source sectors. Assuming a relatively constant source this translates to a lifetime of about 4 hours (at column level, and assuming basic mass balance).

To summarize we changed lines 258-259:

The effective lifetime of NO<sub>x</sub> depends on both the chemical decay rate and loss to surfaces (dry deposition). Within our domain of interest the chemical decay will be the dominant factor. Earlier studies using the EMG plume functions derived lifetimes between 2-5 hours based on the decay downwind of major sources worldwide (Beirle\_2011, deFoy\_2015, Goldberg\_2021, Lange\_2022, Fioletov\_2022}. Following those results we assume a mean lifetime of 4 hours  $\pm$  1 hour to account for local and seasonal variations.

- **p11, L287. I didn't find the factor 1.32 in Beirle et al. (2016) either.**

Response: The year stated for Beirle et al's manuscript was incorrect and should be 2019. Changed to 2019.

- **p11, L292-293: 'The gridded NFR data .... summed to the ... grid'. Does gridded data need to be gridded? CLRTAP inventories are usually gridded by NFR categories.**

Response: changed the sentence to "The GNFR data is used as a basis and summed and regridded for all the NFR classes, to match the 0.1°x0.1° grid used in this study."

- **p12, L307. Why a comma after 'way'? This is just one example of a common problem.**

Response: A native English speaker re-edited the document and made changes throughout the manuscript.

- **p12, L323. Again, somewhat sloppy. Fig. 4 doesn't say anything about previous sensors, and the text doesn't explain what the authors are thinking. If you make comparative statements, back them up.**

Response: Removed sentence.

- **p12, L330. Here I also wonder about the 4-hour footprint issue mentioned above.**

Response: Accounted for advection/diffusion in the source-receptor relations. See the more detailed explanation in the major comments.

- **p14, L345. Remove 'the before mentioned'.**

Response: Removed "the before mentioned"

- **p14, L349. How should non-Germans know where the A1 motorway is?**

Response: Added "(the line of emissions between the major emissions clusters at C and H)"

- **p15, Fig 5. Explain letters in top-right fig. Also explain whether positive values (red color) means that the satellite has more or less emission than the inventory.**



Response: "added, The letters in the figure indicate the following; H: Hamburg B: Berlin, C: Cologne, LU: Lusatia, LE: Leipzig, M: Munich, S: Stuttgart, F: Frankfurt.", also added "The red values indicate a higher value for the satellite derived emissions compared to the inventory emissions.", typically positive values, here red, indicates that x is larger than y in difference between x and y.

• **p16, Fig 6. What are the triangles? What are the various letters (NEU, WW, ...)? The latter are explained in the text, but the caption should be informative.**

Response: Added The red triangles indicate the larger Power-Plants, with the letter combinations indicating the names of the powerplants; NEU (Neurath), NIE (Niederaussem), WW (Weisweiler), LD (Lippendorf), JAN (Janschwalde), SP (Schwarze Pump), and BB (Boxberg).

• **p16, L372. Why are 65 kt NO<sub>x</sub> added only at this stage?**

Response: As discussed in the manuscript the 60+5kt NO<sub>2</sub> emissions are rough indications of the totals within the domain, but no spatially varying result could be calculated. Hence the values were only added at this stage.

• **p18, Fig. 8. Again, the caption explains too little. What are the error bars? Be explicit and say slight rise in 'reported' emissions.**

Response: added "Black error bars indicate the uncertainties on the inventory emissions while the red error bars show the uncertainty in the satellite derived emissions. Note the slight rise in the reported emissions of 2021,"

• **p18, L375. Start a new paragraph from 'Emissions sources'; so that the reader knows the subject has changed.**

Response: Added break

• **p18, L380. Fig. A3 doesn't support that the Agri emissions are spread out across the country, at least not if the text is about the >50% region.**

Response: Removed agricultural emissions.

• **p18, L382. Why are 'non-agricultural sources' mentioned here? Only 3 sources are addressed, so many sources are excluded.**

Response: changed to " Public Power, Industry, Road Transport and Shipping sources"

- **p20, L395. When starting the discussions, be explicit that the reported emissions are for Germany.**

Response: rephrased to “from the emissions reported for Germany”.

- **p20, L402. Why does Fig. 5 ‘hint’ at a small and widespread source? The values seem close to zero in most areas.**

Response: While the contribution is small the value (at 2.5 tonnes/km<sup>2</sup>) is above the estimated detection limit (1.4 tonnes km<sup>2</sup>) and due to its wide-spread occurrence adds up to quite a total.

- **p20, L403. Again start a new paragraph when the subject is changing.**

Response: Added breaks throughout the discussion.

- **p21 L432. ‘improved’ - from what?**

Response: rewritten section on uncertainties, removed.

- **p21, L436. ‘approach u’ typo**

Response: Removed u.

- **p21, L437. CAMS-Europe - reference?**

Response: Added Douros et al., 2023.

- **p21, L440. ‘detect’ should be ‘detection’**

Response: removed in rewritten section.

- **p21, L445. Mangled sentence.**

Response: removed in rewritten section.

- **p21, L445. How do you know that the 4h timescale is correct for Germany as a whole in 2019?**

Response: See earlier discussion on lifetime and responses to Reviewer 2.

- **p21, L448. Lifetimes are not only location dependent; they depend on**

**complex interactions between meteorology, chemistry and vegetation state (via deposition).**

Response: See earlier discussion on lifetime and responses to Reviewer 2.

• **p21, L453. What is meant by several %? That number seems low.**

Response: Removed in rewritten section, added % in renewed section 2.2.2.

• **p22, L472. What is '(8)'?**

Response: Added "Fig. "

• **p22, L484. The web-tool is mentoned, but I would hardly say it was 'presented'.**

Response: Removed text.

• **p23, L496. What is 'ads'? Provide a web address.**

Response: Added web address. <https://ads.atmosphere.copernicus.eu/#!/home>

• **p23, L498. No need to use words like 'truly' in a scientific statement.**

Response: Removed sentence.

• **p23, L502. It was discussed earlier that the resolution is not really 3.5x5.5km<sup>2</sup>, and the derived-emissions resolution are certainly not at that level.**

Response: Removed part of sentence.

• **p22-24. Again, one sinlge paragraph over more than a whole page! Break up the text into separate topics.**

Response: Added breaks throughout.

• **p23, L508. Reference the Green Deal.**

Response: Added website link.

• **p23, L513. The sentence starting 'While' ends abruptly, making no sense. Also, rephrase 'whole there here developed methodology'.**

Response: Changed to "While the estimated errors of individual years are larger than those variations, most error components will stay consistent between the years.", changed to "The here developed..."

- **p23, L518. 'tooling' should be 'tools'.**

Response: Adjusted to tool.

- **p23, L52. What is 'link website, mode fields'?**

Response: Removed

- **Fig A1. Be explicit: NOx emissions.**

Response: Added "NOx"

- **p28, Fig A4. Again (as with Fig.5), explain what positive values mean.**

Response: Added "The red values indicate a higher value for the satellite derived emissions compared to the inventory emissions."

- **p30, L562. Mangled NO 2.**

Response: Fixed, also checked adjusted other NO 2 references.