Report #1

Submitted on 22 Dec 2024
Anonymous referee #1

Anonymous during peer-review: Yes No
Anonymous in acknowledgements of published article: Yes No

Checklist for reviewers

(1) Scientific significance Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?	Outstanding **Excellent** Good Fair Low
(2) Scientific quality Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?	Outstanding **Excellent** Good Fair Low
(3) Presentation quality Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?	Outstanding Excellent **Good** Fair Low

For final publication, the manuscript should be

accepted as is

accepted subject to **technical corrections**

accepted subject to **minor revisions**

reconsidered after **major revisions**

rejected

Were a revised manuscript to be sent for another round of reviews:

I would be willing to review the revised manuscript.

I would not be willing to review the revised manuscript.

Suggestions for revision or reasons for rejection

(visible to the public if the article is accepted and published)

The authors have addressed the issues I raised. With the caveat that my expertise in remote sensing and inverse modelling is limited, I'm happy to recommend publication.

Response: We appreciate reviewer for his comments that enables us to improve the discussion on the atmospheric chemistry of NH3 in a more comprehensive way.

Report #2

Submitted on 06 Jan 2025
Anonymous referee #2

Anonymous during peer-review: Yes No
Anonymous in acknowledgements of published article: Yes No

Checklist for reviewers

(1) Scientific significance Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?	Outstanding Excellent **Good** Fair Low
(2) Scientific quality Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?	Outstanding Excellent Good **Fair** Low
(3) Presentation quality Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?	Outstanding Excellent Good **Fair** Low

For final publication, the manuscript should be

accepted as is
accepted subject to technical corrections
accepted subject to minor revisions
reconsidered after major revisions
rejected

Were a revised manuscript to be sent for another round of reviews:
I would be willing to review the revised manuscript.
I would not be willing to review the revised manuscript.

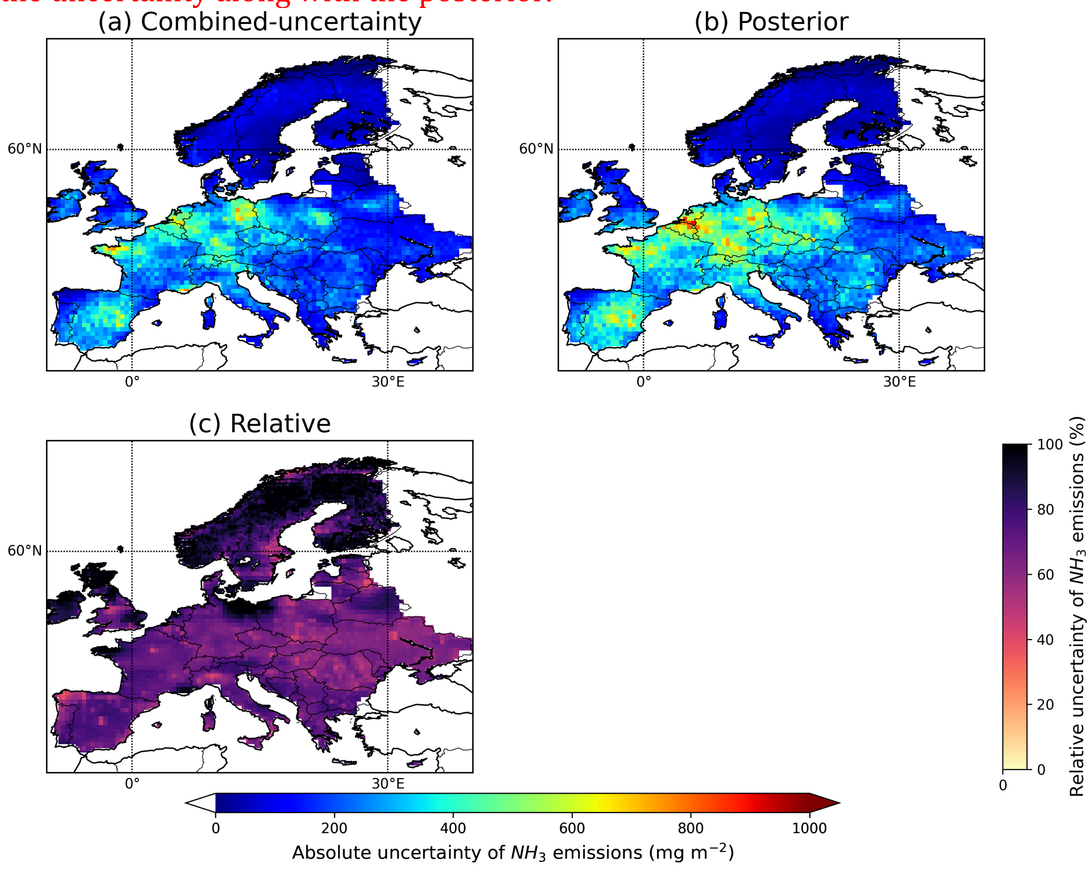
Suggestions for revision or reasons for rejection

(visible to the public if the article is accepted and published)

Evangeliou et al. present constraining ammonia emissions in Europe to evaluate why PM2.5 did not decrease as much as expected during COVID. Ammonia is an important precursor for controlling secondary inorganic aerosol, which in turn can control other aerosol processes. However, ammonia is not well characterized. The authors have addressed many of the concerns raised by both reviewers. However, there one main concerns that need to be addressed prior to publication:

The biggest concern is still associated with Fig. 3 and the associated text and uncertainty. If I understand what is being said in the caption of Fig. 3, Fig. 3d is the relative ratio between Fig. 2c and Fig. 3c. Estimating values between Fig. 2c and 3c, for the Belgium/Netherlands area, as an example, I calculate a relative uncertainty in the range of 30 - 70% (vs the <20% shown in Fig. 3d). More information in how the relative uncertainty was calculated is needed as currently the figures suggest the values should be much higher than what is currently being shown/stated.

Response: We appreciate for this comment. As the reviewer says, the relative uncertainty is calculated as the specific uncertainty divided by the posterior. To calculate uncertainty, we have performed numerous inversions using different priors and different surroundings, as described in the manuscript. So, instead of using the posterior calculated using the avgEENV as the prior (please see section 2.3) in the calculation of the relative uncertainty, we have mistakenly used one of the others (the posterior using EGG as the prior). We have corrected now Fig.3 and we also show below the uncertainty along with the posterior.



Report #3

Submitted on 09 Jan 2025
Anonymous referee #3

Anonymous during peer-review: Yes No
Anonymous in acknowledgements of published article: Yes No

Checklist for reviewers

(1) Scientific significance Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?	Outstanding Excellent **Good** Fair Low
(2) Scientific quality Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?	Outstanding Excellent **Good** Fair Low
(3) Presentation quality Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?	**Outstanding** Excellent Good Fair Low

For final publication, the manuscript should be

accepted as is
accepted subject to technical corrections
accepted subject to minor revisions
reconsidered after major revisions
rejected

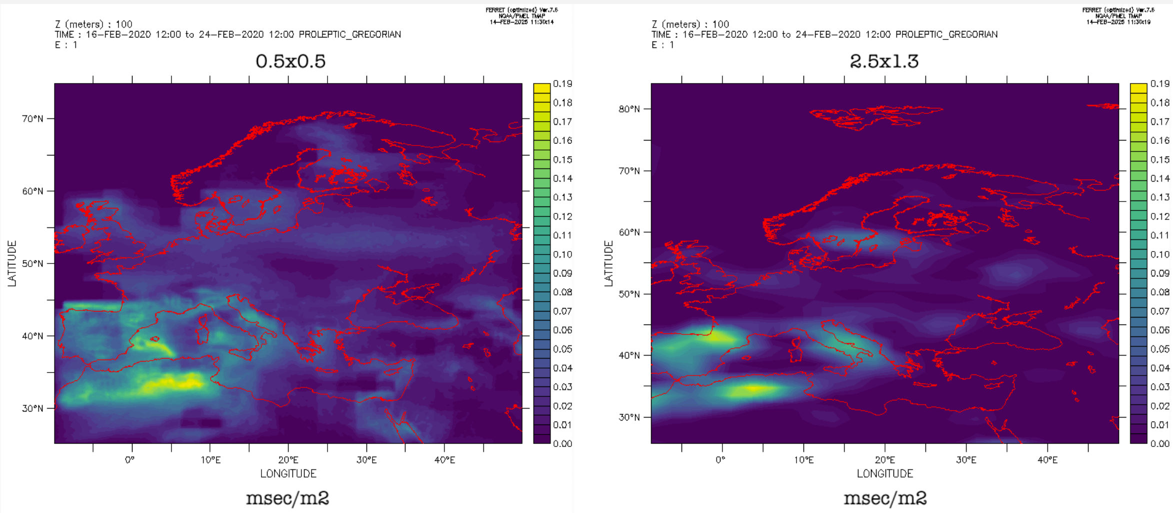
Were a revised manuscript to be sent for another round of reviews:
I would be willing to review the revised manuscript.
I would not be willing to review the revised manuscript.

Suggestions for revision or reasons for rejection

(visible to the public if the article is accepted and published)

Evangeliou et al. examined the impact of lockdown measures in Europe due to COVID-19 on the ammonia emissions and concentration using satellite observations and a global model combined to inverse modelling algorithm. The reviewer #1 et #2 have already addressed many concerns on result part nevertheless, I have some major comments on methodology employed that should be considered by the authors before publication.

1/ FLEXPART and CrIS are used with 0.5°x0.5° of horizontal resolution. Life time of ammonia is calculated with very coarser resolution 2.5°x1.3° by LMDz-OR-INCA. How this large difference of resolution can impact the results?
Response: This is a tricky question, and we do not think anyone could give an informative answer. The reason is because the LMDz-OR-INCA model is a Eulerian model and only exists in this resolution (2.5x1.3).
However, we have performed a test to answer this question:
Normally the 2.5x1.3 lifetime calculated with LMDz-OR-INCA is read in FLEXPART and based on this (and the methodology presented in Tichý et al. (2023)), we calculate the source-receptor matrices (SRMs or footprints) that are needed for inverse modelling. If FLEXPART resolution is different than 2.5x1.3 (of the lifetime), then the lifetime is regridded within the code.
Here, we test how SRMs are affected by the regridding of lifetime in FLEXPART. For this reason, we calculate SRMs in 0.5x0.5 resolution (where regridding is needed) and SRMs in the exact resolution of the lifetime (2.5x1.3, for which regridding is not performed as lifetime matches the resolution of the model). The results are shown below:



The SRMs averaged over Europe for random day in February 2020 do not show large differences.

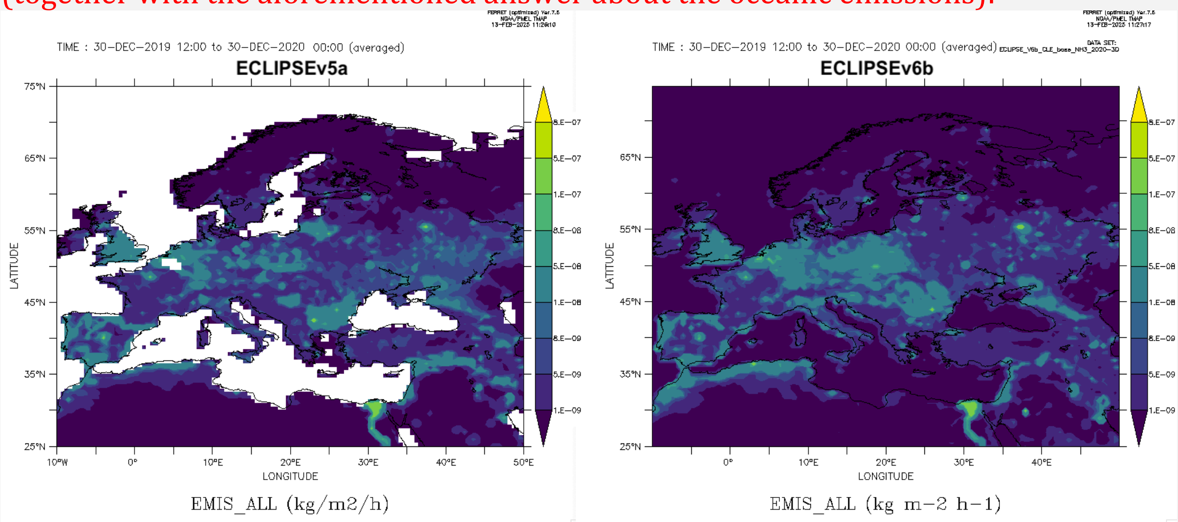
2/ It was very hard for me to understand clearly which emissions inventory was really used and why. I think this would require a dedicated section in the paper.
Response: We understand the concerns here; Our intension was to not confuse the readers with details from previous publications, but the reviewer is probably right. Therefore, we have tried to answer to the following questions in the text and therefore we point to page number and line number of the clarifications.

a) EGG is defined as ECLIPSEv5+GFED4+GEIA. Is it really ECLIPSE v5 or v5a? Which inventory does GEIA refer to? ECLIPSE are yearly emissions. Which monthly profile did you use?

Response: – We used ECLIPSEv5a, so we have changed this in P6-L160.
– The GEIA inventory is explained in P6-L162 and is also associated with a citing article (Bouwman et al., 1997). We used it to adopt the oceanic emissions of NH3 that were not present in ECLIPSEv5a. A link to the initiative can be found in https://igacproject.org/activities/GEIA.
– ECLIPSE is given annually of course. However, in the dataset, the developers give access to a file called "ECLIPSE_V6a_monthly_pattern.nc", where the users can use monthly weights (for specific sectors) to acquire monthly emissions (Please see details here: https://iiasa.ac.at/models-tools-data/global-emission-fields-of-air-pollutants-and-ghgs).

b) EC6G4 is defined as ECLIPSEv6+GFED4. Is it really ECLIPSE v6 or v6b. Here you don't use GEIA emissions. Why? What is the difference in terms of spatial distribution and total mass between ECLIPSEv6 and ECLIPSEv5?

Response: – We used ECLIPSEv6b and we have changed this in 2 places in the manuscript (P8-L227, P9-L246).
– As mentioned previously, the GEIA emissions were used together with ECLIPSEv5a and GFED to represent the oceanic emissions of NH3. In ECLIPSEv6b, oceanic emissions are included. Nevertheless, due to the very low oceanic emissions (10-fold or lower), as compared to agricultural and other anthropogenic, we removed oceanic emissions from our constraint and we only report terrestrial ones in the posterior.
– The spatial distribution of NH3 emissions in ECLIPSEv5a and v6b is shown below (together with the aforementioned answer about the oceanic emissions):



c) NE emissions refer to Evangeliou et al., 2021. In this paper, NE comes from calculations from IASI observations with lifetime calculated from LMDz-OR-INCA using EGG emissions. The NE emissions are calculated from 2008 to 2017. How do you extent this database for the period 2016-2020?
Response: The publication Evangeliou et al. (2021) refers of course to years 2008-2017. However, IASI NH3 from METOP-B is available for the years after. Since the leading author of the NE emissions paper is the same with the one of the present, it is evident that an extension of the calculation until 2020 is more than easy.

d) VD emissions are based on Van Damme et al. (2018) calculations (Evangeliou et al., 2021). In Evangeliou et al., 2 emission inventories called VD0.5 (life time of 12h) and VDgrlf (whith life time from a model) are used. Which one is used here?
At the end, the average of these 4 emission inventories are used. Is it realistic? Two of these inventories (EGG and EC6G4) are not independent (different version of ECLIPSE emissions) and NE emissions are calculated from the lifetime calculated with the first inventory (EGG).
Response: We have now clarified this point the first time that the emissions are mentioned in the manuscript (P8-L226).
Whether the use of the average of 4 emission inventories is realistic or not is shown very well in Figure 1 (far right panel). The average emissions appear to create the most realistic model concentrations that are in the best agreement with ground-based observations. This is a good argument to use them. Besides, we calculate and report the uncertainty in the posterior from use of different prior emissions (see section 3.2).

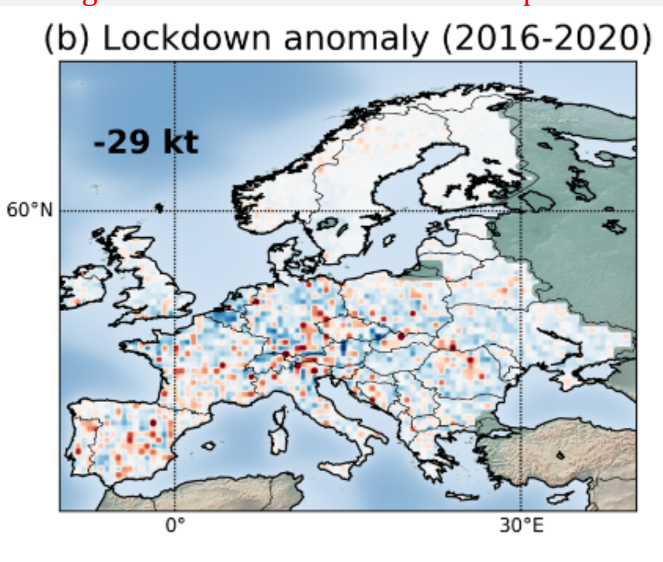
3/ To calculate prior emissions for year 2020, adjustment factors from Doumbia et al. (2021) are used. Use them with EGG and EC6G4 is probably not a problem because the same emission sectors are defined. It seems more complicated to me to use this for NE and VD emissions. It should be explained in the manuscript.
Response: This is a great comment that gives us the opportunity to correct the manuscript accordingly. The adjustment factors from Doumbia et al. are only applied to the bottom up emission inventories, namely EGG and EC6G4. The reason is that the top-down inventories are based on real measurements, where potential changes due to COVID-19 lockdowns are already included in the measurements. We now clarify this in P8-L233-235.

4/ Different publications have shown NH3 peaks in spring over northwestern European countries. This is confirmed by your figures 2 and 4 with a maximum of NH3 concentration observed by CrIs in April. In addition, the NH3 lifetime simulated by the model is not very variable (from 0.47 to 0.53 days). However, avgEENV and VD emissions show a continue increase of emission fluxes from January to July. How do you explain this?

Response: NH3 levels peak in spring and late summer in Europe. This is not necessarily a result of maximum emissions occurring in spring and late summer, although we know quite accurately that spring is the fertilization period and late summer when the temperature dependent volatilization occurs. An issue that is often overlooked and seems to have been the case in 2020 is the relationship of the NH3 levels with the changing chemistry of the atmosphere due to rapid decreases of NOx and SO2. This is what we have tried to link and examine in the present study. While NH3 emissions appear to not peak in spring, when they are averaged over Europe (Figure 2f), the country-specific seasonality appears to be correct. For instance, in Denmark emissions peak in late February, in France, Netherlands, Ireland, Austria, Czechia and Belgium in March, in Greece and Spain in April.

5/ I agree with Reviewer #2, several studies have shown specific weather conditions during the covid period (Deroubaix et al., 2021; Gaubert et al., 2021, van Heerwaarden et al., 2021…) impacting the concentrations of many species (NO2, O3 etc.). This should be investigated with anomalies map not with an average over Europe that hides and compensates for geographical patterns.

Response: Figure 5b (a print screen is shown below) shows emission anomaly relative to the 2020 lockdowns from the 2016-2020 period (15 March – 30 April). The latter shows that emissions during the 2020 lockdowns dropped by -29 kt with respect to the same period in 2016 – 2020, which shows the impact of the COVID-19 restrictions. Maximum decreases were seen in The Netherlands and Belgium, both countries comprising high emissions that also suffered heavily from the COVID-19 outbreak, as well as other countries (see description in L13-14 of the manuscript). This agrees well with the state of the epidemic in these countries in spring 2020.



6/ CrIS data were used with Quality and Cloud flags. Over the period, the number of available data can therefore greatly vary in time and space. The quality of posterior emissions should be assessed on the basis of the number of data sets available.
Response: In principle, using the quality flags that are specified in section 2.1 gives a very large number of observations per gridcell in the regions of the highest NH3 levels (central Europe, the Po valley and Holland and Belgium). The number of around 10,000 observations per day per vertical level (see P5-L122) is way larger than any measurement network of ground-based observation can give, which are traditionally used in inverse modelling of BC, CO, CF4, C2F6, CH4 or N2O (see references below).

- M. Jia, F. Jiang, N. Evangeliou, S. Eckhardt, X. Huang, A. Ding, and A. Stohl, Rapid decline of carbon monoxide emissions in the Fenwei Plain in China during the three-year Action Plan on defending the blue sky, J. Environmental Management, 337, 117735, doi.org/10.1016/j.jenvman.2023.117735, 2023.
- Evangeliou, N., Platt, S. M., Eckhardt, S., Lund Myhre, C., Laj, P., Alados-Arboledas, L., Backman, J., Brem, B. T., Fiebig, M., Flentje, H., Marinoni, A., Pandolfi, M., Yus-Dìez, J., Prats, N., Putaud, J. P., Sellegri, K., Sorribas, M., Eleftheriadis, K., Vratolis, S., Wiedensohler, A., & Stohl, A. (2021) "Changes in black carbon emissions over Europe due to COVID-19 lockdowns", Atmospheric Chemistry & Physics, 21, 2675-2692, https://doi.org/10.5194/acp-21-2675-2021.
- J. Kim, R. L. Thompson, H. Park, S. Bogle, J. Muhle, M-K. Park, Y. Kim, C. Harth, P. Salameh, R. Schmidt, D. Ottinger, S. Park, and R. Weiss, Emissions of tetrafluoromethane (CF4) and hexafluoroethane (C2F6) from East Asia: 2008 to 2019, J. Geophys. Res. Atmospheres, 126, e2021JD034888, doi:10.1029/2021JD034888, 2021.
- Evangeliou, N., Thompson, R. L., Eckhardt, S., & Stohl, A. (2018), "Top-down estimates of black carbon emissions at high latitudes using an atmospheric transport model and a Bayesian inversion framework", Atmospheric Chemistry & Physics, 18, 15307-15327, https://doi.org/10.5194/acp-18-15307-2018.
R. L. Thompson, M. Sasakawa, M. Machida, T. Aalto, D. Worthy, J. V. Lavric, C. Lund Myhre, & A. Stohl, Methane fluxes in the high northern latitudes for 2005-2013 estimated using a Bayesian atmospheric inversion, Atmos. Chem. Phys., 17, 3553-3572, doi:10.5194/acp-17-3553-2017, 2017.

7/ The resolution of the data used to make the Country mask seems very too coarse to use it on grid of 0.5° of resolution. By example, Belgium is represented by 4 cell grids.
Response: The resulting posterior emissions were calculated onto a 0.5 degree resolution grid, due to the use of 0.5 degree resolution meteorological fields from ECMWF ERA5 (see section 2.2). Therefore, when trying to mask specific regions, and calculate country-specific fluxes, it is inevitable that we will use country masks of the same 0.5 degree resolution. We are unaware of any other method to do this calculation.

References:

Adrien Deroubaix, Guy Brasseur, Benjamin Gaubert, Inga Labuhn, Laurent Menut

Guillaume Siour, Paolo Tuccella: Response of surface ozone concentration to emission reduction and meteorology during the COVID-19 lockdown in Europe, Meteorol Appl., 2021.

Gaubert, B., Bouarar, I., Doumbia, T., Liu, Y., Stavrakou, T., Deroubaix, A., Darras, S., Elguindi, N., Granier, C., Lacey, F., Müller, J.-F., Shi, X., Tilmes, S., Wang, T., and Brasseur, G. P.: Global changes in secondary atmospheric pollutants during the 2020 COVID-19 pandemic, J. Geophys. Res.-Atmos., 126, e2020JD034213, https://doi.org/10.1029/2020JD034213, 2021.

van Heerwaarden, C.C., Mol, W.B., Veerman, M.A. et al. Record high solar irradiance in Western Europe during first COVID-19 lockdown largely due to unusual weather. Commun Earth Environ 2, 37 (2021). https://doi.org/10.1038/s43247-021-00110-0