Global and regional emissions of 1,2-dichloroethane derived from AGAGE and NOAA observations

Correspondence to: Joseph R. Pitt (joseph.pitt@bristol.ac.uk) and Dominique Rust (dominique.rust@bristol.ac.uk)

Comment: The paper "Global and regional emissions of 1,2-dichloroethane derived from AGAGE and NOAA observations" by Pitt et al., (2026) reports data from continuous measurements of 1,2-dichloroethane (DCE), made by the authors, within two important global monitoring networks, specifically AGAGE and NOAA, which they will make it publicly available to the community through the networks' websites. The paper primarily reports the sampling, analysis, and calibration techniques for the reported data. Then, they use these data to estimate global total emissions for years 2017-2023 using a global inverse modeling method. Authors also use regional inverse modeling methods to estimate regional emission maps for NW Europe and California.

First, I would like to extend my appreciation, on behalf of all users, to the measurement teams for their hard work and dedication in making these measurements. Atmospheric science community needs and relies on these long term datasets to better understand the Earth system and how it evolves.

Second, this study makes a valuable contribution to better understanding the DCE abundance and emissions, and it is worthy of publication in ACP. Nevertheless, the paper needs more discussion on some parts and potential restructuring of some sections and figures. Below, I list my comments, questions, and suggested edits that need to be addressed. "L" refers to "line" number in the manuscript.

Reply: We thank the referee for their detailed review and address their comments (in black) below (in blue).

Title

This paper primarily reports the measurements of DCE and the emission estimation is of a secondary importance as the authors correctly state in line 87-88 that "*Here we present, for the first time, ongoing surface-based measurements of DCE from the AGAGE (AGAGE, 2025; Prinn et al., 2018) and NOAA (NOAA, 2025) global measurement networks*." However, the title gives the impression that gridded emission files will be the main product of the paper, which is not the case as per my understanding. I would recommend using a more accurate title.

Reply: We have changed the title to better reflect the focus on observations: *"Observations of 1,2-dichloroethane from the AGAGE and NOAA networks and derived global and regional emissions".*

Abstract

L7-8: Please use mean ± std, instead of using brackets (e.g., [268, 638]). Using the bracket suggests a range, but my understanding is that these are the standard deviations following the results in L381.

Reply: We have followed this suggestion for the global results and now use mean ± std throughout. For the regional results the uncertainty is asymmetric so characterising it using a single value could be misleading. There does not appear to be a specification for the presentation of asymmetric uncertainties in the ACP guidelines, so we are happy to defer to the editor and/or typesetter if they have alternative suggestions for how these regional results should be presented.

Section 1 Introduction:

L33: I would suggest also mentioning 'DCA' as one of the abbreviated forms of 1,2-Dichloroethane. While both "DCA" and "DCE" have been used in the literature for this compound, DCE has been used for dichlroeth**E**ne too, when doing a 'Web of Science" search for "dichloroethene DCE".

Reply: Good point – we've added this to the list.

L80: Please check the references to make sure they are used correctly. For example, it cites Pan et al. (2024) for ground based measurements. But, this mission was focused on UTLS using airborne measurements.

Reply: In that sentence we had combined all citations for the studies including aircraft and ground-based measurements at the end of the sentence. However, we have now separated them, putting all respective citations after the word "aircraft", and all respective citations after the half-sentence concerning the ground-based studies.

L89: Please reword the sentence to be accurate "With measurements from archived air samples, we extend the record back to 1995", as my understanding is that this step is done for only a few sites.

Reply: Edited to *"With measurements of archived air samples from three sites, we extend the record back to 1995"*

Section 2.1: Measurement sites

I couldn't find any information on the "AGAGE&NOAA" and "KOPRI" sites in the text (maybe lines 239-242) while they are shown in Figure 1. Please add what they are, and edit the results section accordingly.

Reply: We have edited the figure caption to explicitly state *"Dark purple squares: sites in both the AGAGE and NOAA networks".*

We have also added the following to the text: *"These flasks were collected by the Korea Polar Research Institute (KOPRI) and analysed by the Swiss Federal Laboratories for Materials Science and Technology (Empa)."*

L132: For clarity, please replace 'std' with another word, e.g., 'stdm' or another word. 'std' could also refer to standard deviation. Also, consider changing 'air' to be consistent with the standard measurement.

Reply: We have added *"_sample"* in both cases.

Section 2.2: Sampling and analysis

It is a pretty long section. Please separate it to sub-sections, maybe separate to AGAGE and NOAA measurements.

Reply: We have split this into AGAGE and NOAA sub-sections as suggested

Section 2-3: Calibration

Same as the previous comment. Please consider separating it two calibration methods for AGAGE and NOAA.

Reply: We have split this into AGAGE and NOAA subsections with a third short subsection on the network comparison.

Section 2-4-3: Regional modelling

I think presenting the differences between these models in a table format would be very helpful for the readers to better understand them. Please add a table to either the main text or the appendix.

Reply: We have added the following table to Appendix F

Table F1. Details of the differences in model configuration for the four inverse modelling approaches.

	RHIME	InTEM	ELRIS	FLEXINVERT
Bayesian inversion type	Numerical (hierarchical using Markov chain Monte Carlo)	Analytical	Analytical	Numerical (iterative M1QN3 solver)
Dispersion model	NAME	NAME	NAME	FLEXPART
Dispersion model transport duration	30 days	30 days	30 days	10 days
Time step (observation averaging period)	4 hours	4 hours	4 hours	3 hours
Prior background	MHD clean sector (Europe) 12-box model (California)	Manning et al. (2021), adjusted using the annual average RPB:MHD ratio	REBS method based on MHD observations	REBS method based on individual sites

We also realised that ELRIS used a different correlation length for the prior emissions compared to the reference Vollmer et al. (2025). We added the following: *"Similarly ELRIS used a spatial correlation length of 250 km for the prior emissions, as opposed to 500 km used by Vollmer et al. (2025), reflecting the expectation of more point-like sources and greater uncertainty on the prior spatial distribution for DCE."*

Section 3.: Results and discussion

L 318: Since authors are showing the measurements, I would recommend removing the sentence about modeling results from this section, and move it to the modeling results section.

Reply: Although section 3.1 mixes pure measurements (e.g. Figs. 2 and 3) with results from the 12-box model, we think it makes narrative sense to group the discussion of mole fractions together in this way, such that section 3.2 focusses solely on emissions. We have changed the heading for section 2.1 to *"Atmospheric mole fractions"* to better reflect the fact that it includes model results.

Figures2,3: Please use the stations' abbreviated names instead of full name in these figures, so readers can easily find the location in Figure 1.

Reply: We have changed the station names to use their abbreviations in the plot legends instead of their full names. For clarity, we list the full names in the respective captions.

Similar to the comment above, please use consistent naming in the whole text; i.e., use the abbreviated names always (and use the station name in parenthesis if needed).

Reply: We have followed this suggestion and changed to the site abbreviations everywhere within the main text. In figure captions we have kept it as "long name (abbreviation)" for consistency with Fig. 1. We have added the following to Sect 2.1:

"The sites and their associated three-letter abbreviations are shown on a map in Fig. 1 and listed in Appendix B. Sites are referred to throughout by their three-letter abbreviations for ease of reference against Fig. 1."

Figures 2,3: Also, the order for separating stations in each panel is not clear. I would recommend separating either by latitude or longitude ranges. It will help readers better understand the zonal distribution also. Or, maybe use a logarithmic scale and one panel in each figure might provide more insights on the differences; this way, extreme values would be also present which are important.

Reply: We changed the order of the records in the sub-panels of the plots. The records are now ordered after the latitudes of the sites. We now also changed the y-axes scales to show the extreme values. Additionally changing to better distinguishable colours in each sub-plot, the records are still nicely visible, and we did not choose a logarithmic scale, as we believe this will introduce some confusion about the interpretation of the relative magnitudes of the pollution events between the records.

L336-345: I believe this section deserves more discussion; maybe an additional paragraph on more specifics. For example, which sites found high concentrations, what the range of those concentrations were, etc.

Reply: We have added the following paragraph:

"NOAA flask samples are typically collected under background conditions, although several samples at Mauna Loa were found to be elevated above baseline due to the influence of East Asian outflow. In contrast the in situ AGAGE data show pronounced pollution events (deviations from the baseline level) at several northern hemispheric sites. In particular, mole fractions more than twice the baseline value were frequently observed at Tacolneston, with extreme events exceeding 100 ppt. These events reflect the proximity of this site to known producers and users of DCE (see Sect. 3.3). Monte Cimone is the

next most polluted site, followed by Mace Head and Taunus. No other AGAGE site observed mole fractions exceeding 40 ppt."

L340-342: What do the ranges show? Are they min-max? please specify. Also, please quantify the variability.

Reply: We have replaced our previous vague statement with a min, max and standard deviation: *"In the northern hemisphere, observed baseline monthly-mean mole fractions range between a minimum of 4 ppt and a maximum of 27 ppt (with a standard deviation of 4 ppt across both the AGAGE network and NOAA network) roughly 5–25 ppt, which is consistent with boundary layer mole fractions previously reported from aircraft campaigns"*

L346-L352: This paragraph quantifies other Cl-VSLS compounds, but misses to specify the corresponding measured values for DCE.

Reply: We have added the 2020 DCE values here:

"Although the global production volume of DCE is by far the largest among the Cl-VSLS regularly measured and publicly reported by the two networks, the global mean mole fractions (estimated directly from the measurements or from the model posterior) of DCE are third highest. The 12-box model global average mole fractions for DCE in 2020 (6.7 ppt (AGAGE) and 7.7 ppt (NOAA)) sit between those of DCM (38.3 ppt (AGAGE) and 45.5 ppt (NOAA)) (Laube and Tegtmeier et al., 2022) and PCE (1.01 ppt (AGAGE) and 1.12 ppt (NOAA)) (Laube and Tegtmeier et al., 2022), and are approximately of the same magnitude as CFM (8.7 ppt (AGAGE)) (Laube and Tegtmeier et al., 2022)."

L353-364: This paragraph doesn't show any results. Please move this paragraph to the methods sections.

Reply: We have removed this paragraph as the sensitivity of the network is discussed in the context of the regional emission estimates.

L365-369: For the increasing trend, please quantify it. On the other hand, even if the trend is less clear after 2018, Figure 4 does show a change. Are there any speculations or hypothesis for that change?

Reply: We have used the 12-box model to estimate global emissions from 2002 onwards based on the air archive measurements, enabling a more quantitative comparison between the air archive measurements and the Hossaini emissions, which is presented in Appendix F. The sparsity of the air archive dataset leads to a high uncertainty on the emissions growth rate derived using the 12-box model, such that our results are consistent with Hossaini sc05 emissions both in terms of magnitude and trend. See the response to reviewer 1 for more detail.

If we take the central estimate of the 12-box model at face value, we have shifted from an earlier period with steadily increasing DCE emissions, through plateau in emissions, to a period with decreasing emissions from 2018 to 2022 (albeit with an increase from 2022 to 2023). Given the large uncertainty on the trend for the pre-2017 period it is hard for us to say anything beyond that our results are consistent with Hossaini et al., who derive an increase in emissions due to an increase in DCE production/consumption (resulting from increased demand for PVC). In the pre-2017 period we have added the following discussion to Sect. 3.2:

"The decrease in emissions between 2018 and 2020 observed in our results is larger than the corresponding decrease in the Hossaini et al. (2024) scenarios. Improved abatement measures could play a role in this discrepancy, as they would reduce the average global emission factors from DCE production over time. Similarly, a reduction in emissions during feedstock use or transport of DCE could contribute (especially if these sources constitute a larger share of global emissions than assumed by Hossaini et al. (2024)), as could a reduction in solvent use of DCE (a source that is not included in the Hossaini et al. (2024) scenarios). Extending both the bottom-up inventory estimates and the top-down 12-box model estimates to future years would enable a better assessment of whether systematic changes in these DCE emission sources are indeed occurring."

When presenting the measurements, authors state there is not a consistent trend (line 319), and again in line 368. They also state that the estimated emissions do not show a trend (L381). However, Figure 4 shows a reduction in the last few years. At the same time, prior emissions were similar for years 2020-2023, but the posterior emissions are showing a decreasing trend in these years. They all suggest to me that there is a decreasing trend in the last few years. Please provide more clarifying information on these points.

Reply: After reviewing this again we agree that the decrease in estimated emissions from 2018 to 2022 is a significant trend that deserves more discussion. We have added this discussion to Sect. 3.2 (see response to the previous comment) and we have edited the start of Sect 3.2 to:

"Estimated global annual emissions are shown in Fig. 5. Over all years covered by this study (2017–2023) mean emissions are 453 ± 185 Gg yr−1 (AGAGE) and 525 ± 209 Gg yr−1 (NOAA), with a decreasing trend observed between 2018 and 2022."

At L319 we have changed the text to:

"There is not a monotonic trend in the annual average global atmospheric abundance over this period. We observe growth between 2017 and 2018/2019, followed by a decline until 2022, returning to growth from 2022 to 2023. This may reflect changes in emissions (see Sect. 3.2), although any changes in the DCE sink over this time period could also contribute."

Section 3.2: Global emissions

L405-413: Based on Figure 5, "sc3" is also within the uncertainty range. Why didn't authors include it in the discussion?

Reply: Sc03 is just within the uncertainty range of our AGAGE-derived estimate but not for our NOAA-derived estimate. We have clarified this by editing the sentence to: *"Our derived global emissions from both networks fit well to the emissions magnitudes of scenarios 4 (sc04) to 6 (sc06) described in their study".*

Similarly, I find 50% variability pretty large in this case. It virtually removed all the differences made for scenarios in Hossaini et al. (2024). In other words, it seems the choice of prior emission scenario would not affect the results. Is that a correct assumption? If no, how sensitive are the results to the selected priori emission? Please clarify in the text.

Reply: The large uncertainty range stems from the assumption that the 1-sigma error in DCE lifetime is 50%. This may well be a conservative estimate, but it was chosen to reflect the fact that the lifetime within each semi-hemisphere is highly variable, making it challenging to model such a short-lived species using this coarse resolution approach. We have added the following text to Sect. 3.2:

"The large uncertainties on these results stem from the assumption of a 50 % uncertainty in the derived total effective lifetime of DCE. This represents a systematic uncertainty, which impacts the absolute emission magnitude and not the emission growth rate, as discussed further in Appendix E."

It is worth noting that only the Hossaini scenario sc05 is used as a prior in the 12-box model runs presented here, but we did do some sensitivity testing at an earlier stage with a prior of 278 Gg yr^{-1} for all years and observed that the posterior results are not sensitive to the selected prior emissions.

Section 3.3: Regional emissions

L465-466 I guess another implication of this analysis is that emissions in Asia are much higher than what Hossaini et al., (2024) estimated. If authors agree, maybe they could comment on that in the text.

Reply: We originally included a statement to this effect, but in fact when you subtract NW Europe and California from the global sc05 emissions, and subtract our estimates of NW Europe and California emissions from our global 12-box model estimates (either network), the two values agree within uncertainty. In other words, our results are not inconsistent with sc05 being too high in these regions, but correct everywhere else.

Code and data availability

The links seem to be expired in the time this review was done. Please make sure all the data becomes publicly available.

Reply: Apologies for this – we have created new links and we will ensure the data becomes publicly available on acceptance of the manuscript.