

On the atmospheric budget of ethylene dichloride and its impact on stratospheric chlorine and ozone (2002–2020)

We thank both reviewers for their positive reviews and helpful comments. Reviewer comments are repeated in *italics* below and our responses to each are given in **blue**.

Responses to Anonymous Reviewer #1

The paper by Hossaini et al al presents a detailed study of 1,2, Dichloroethane, also often referred to as ethylene dichloride. It uses a chemical transport model, atmospheric observations and estimates of production and emission rates to derive an atmospheric budget. The paper is well written and the assumptions and the methods are well justified. I have a few issues, which are more of technical nature concerning the data used which I would like the authors to clarify. One further issue is that I do not like the use of the name ethylenedichloride, which is not a systematic name and also not a specific name. I suggest to use 1,2-dichloroethane instead, which is the correct name and also clearer as it is clear that it is the unsymmetrically substituted compound. Most importantly, I would like the authors to discuss little bit more about the data quality (sample stability and comparison of calibration scales). Apart from that I only have a few minor issues below. I recommend the paper to be accepted after minor revisions.

We thank the Reviewer for this comment around naming. Although 'ethylene dichloride' (EDC) is the common trade name, we agree that 1,2-dichloroethane ('DCE' for shorthand) is the preferred IUPAC name. The latter is also how the molecule is referred to in WMO/UNEP Scientific Assessment of Ozone Depletion reports. Therefore, we have amended the manuscript throughout accordingly (including the title).

I. 203: is ethene mainly of anthropogenic origin? or are there significant natural sources?

Ethene has significant natural and anthropogenic sources. However, for the purpose of this work, we consider only the anthropogenic emission distribution (as a proxy for the DCE emission distribution).

section 2.4.: please include some information on the calibration scales used for the measurements and the comparability of the different observations from three different groups. Are they all on the same scale? Also, has the stability of 1,2-dichloroethane in the samples been analysed?

As a universally adopted international DCE calibration scale is not available at the time of writing, there are likely some differences between groups. The NOAA DCE measurements from ATom are based on the "NOAA-2021" scale. The UCI group use a scale provided by the University of Miami. Details of the UEA scale are given in Oram et al. (2017): DCE "was calibrated at UEA using the established static dilution technique recently described (Laube et al., 2012)". These details have been added to the revised manuscript in the relevant places of Section 2.4.

A detailed examination of differences between the measurement groups is beyond the scope of the paper and will be led by the measurement community in forthcoming work. However, note that scales among the labs considered in this study have historically not differed by more than 10-30% for gases like DCE. To more directly address the reviewer's query, we have conducted a preliminary comparison of background atmospheric DCE mole fractions obtained at two remote sites (Barrow and Samoa Observatories) where both the NOAA and UCI groups sample. This informal intercomparison reveals an average offset of up to ~30% at these two sites. It should be emphasised that this comparison is limited in scope and a more formal and extensive examination

will be required that is beyond the scope of this work. In the revised manuscript we have added the following text at the bottom of Section 2.4:

“Compared to other CI-VSLS, scientific interest in DCE from an ozone depletion perspective is relatively new. As such, an international standard calibration scale has not yet been universally adopted across measuring groups. Historically, the scales among the labs considered in this study have not differed by more than 10-30% for gases similar to DCE. However, in the absence of any formal assessment of calibration scale differences, an informal intercomparison for DCE was performed for this work. Background atmospheric DCE mole fractions from two remote sites (Barrow and Samoa Observatories), where both the NOAA and UCI groups sample, were compared (2017 – 2023). This intercomparison revealed an average offset of up to ~30% (UCI relatively high / NOAA relatively low), i.e. at the upper end of the above range. While this comparison is limited in scope and will require further effort to refine (beyond the scope of this paper), this uncertainty is highlighted in the ensuing discussion”.

In addition to the above text, we will acknowledge more explicitly that model-measurement differences from mission to mission (e.g. Figure 3, Figure S1) could reflect differences in calibration scales used by the labs supplying measurements and to some degree may confound the assessment of model performance and emissions. We do already make a point along these lines (see line 335 of original manuscript) but will strengthen/expand it by pointing to the above findings. It is important to emphasise that calibration scale differences don't affect our main conclusions around the existence of substantial global DCE emissions.

Regarding sample stability, the NOAA ATom sampling was conducted via pressurization into glass flasks, and there has been no indication of systematic growth or destruction of DCE in glass flasks over time within the measurement precision. Similarly, the UCI group has run extensive tests on the stability of compounds in their canisters in the time between sampling and analysis, and EDC is stable within the canisters. UEA samples are collected in silco-treated cylinders (stainless steel with inner surface coated with fused silica, Restek) and no issues with sample loss have been noticed.

These points are also now made in Section 2.4 of the revised manuscript.

I. 270: does this mean that only the data below 3 km of HIPPO and ATOM were used? Why have the free tropospheric data not been used?

The model-measurement comparisons in Table 4 and Figure 3 (a,b) focus on the boundary layer as we are interested in comparing in the region of the atmosphere where the influence of surface DCE emissions will be clearest. However, note that we do also compare model-measurement vertical profiles from HIPPO and ATom in Figure S1.

I. 371, Figure 4: I could not find the blue shaded region to represent the model scenario ranges.

The shading is given but we agree that it is difficult to see. This is due to the high frequency of measurements. We will make this clearer in the revised manuscript.