

We would like to thank the reviewers for their very helpful comments and we have addressed these as follows (reviewer's comment in bold):

Reviewer 1 (Sinnhuber)

As one general comment I feel that the presentation of CH₂Cl₂ emission estimates and the correlation with CH₂ClCH₂Cl to infer new CH₂ClCH₂Cl emission estimates (lines 337 - 343) deserves (and requires) more detail, given its importance. Part of the information on estimating the CH₂Cl₂ emissions given in the supplementary material should be included in the main text and a bit more explanation on the "simple correlation" should be included.

We have moved the discussion about the emission estimates from the supplement into the manuscript as suggested. We have also made a slight modification to our analysis and rather than giving the extreme range of potential CH₂Cl₂ emissions as before (based on the 40:60 and 50:50 production ratios) we have opted to give an estimate based on the more likely ratio of 45:55. This leads us to production and emission figures of 715 kt and 455 kt respectively, with an approximate uncertainty of ± 10%.

We have also expanded the section on the correlation between the two compounds to include the following text

"There is a strong linear correlation between the observed CH₂Cl₂ and CH₂ClCH₂Cl data at both Bachok (R² = 0.9799) and Cape Fuguei (R² = 0.9189). Combining the datasets yields a slope of 0.4456 ± 0.0194 (R² = 0.9228). Using the emissions for CH₂Cl₂ derived above (455 kt) and making the assumptions that (1) all emissions originate in China and (2) there are no significant relative losses of the two compounds since emission (lifetimes are 144 days for CH₂Cl₂ and 65 days for CH₂ClCH₂Cl) we can estimate Chinese emissions of CH₂ClCH₂Cl to be of the order of 203 ± 9 kt yr⁻¹. If accurate, the scale of these emissions is a major surprise as CH₂ClCH₂Cl is highly toxic (suggesting that local emissions would be minimised) and believed to be used almost exclusively in non-emissive applications."

Abstract, I.31: "higher than expected": what is this expectation based upon? Based on previously reported measurements? On line 360 and following it is discussed that many of the previous measurements have been made over a decade ago and in different regions ("... not the 2 key regions ..."). I believe it would be good to make a bit clearer from the start if the enhancements seen in this study are likely because of recent increases in emissions, regional differences, or both.

By "higher than expected" we do indeed mean higher than previously reported data. This was stated earlier in the sentence and refers to both our surface and aircraft measurements. We compare our data with the most recent review (WMO, 2015) as we describe in the text (Section 3, line 236-238) and in Table 1. The enhancements we observe are likely to result from a combination of increasing emissions and the location of the measurements, although based on our measurements alone, which are over a relatively short period of time, we have no evidence that emissions are increasing. We do know from previous work that atmospheric levels of CH₂Cl₂ have increased, which implies increasing emissions of this compound. The long term trend of CH₂ClCH₂Cl is unknown.

I.36: define "Cl-VSLs" when first used. Moreover better use this consistently throughout (e.g. Table 1 uses VSLs-Cl, which I suppose means the same)

Done

I.45: you may want to cite also the recent study by Hossaini et al., The increasing threat to stratospheric ozone from dichloromethane, Nature communications, 2017, that was published after submission of the present manuscript.

As our paper was published in ACPD before the Hossaini et al paper it does not seem appropriate to include it in our reference list at this stage.

I.104: I suggest to break the sentence in two: "... in the TTL. Surface measurements ..."

Done

I. 126: I don't understand the meaning of "globally" here.

We have removed the word "globally".

I.129: "shorter lifetimes" could be misleading here, as it may imply lifetimes shorter than the 10 days for air masses to travel from East Asia to the TTL, which is probably not what is meant?

Text changed to "*despite their relatively short atmospheric lifetimes*"

I.150: "the CARIBIC aircraft": better include a sentence or two on the CARIBIC project, describing that these are measurements from in-service aircrafts, ideally including a reference paper (in addition to the http link).

Done

I.328: "CH₂ClCH₂Cl is exclusively anthropogenic in origin ...": WMO (2014) lists also biomass burning as a source of CH₂ClCH₂Cl. Can you include references on additional sources?

We have added the following text: "*Simpson et al. (2011) observed a small enhancement in CH₂ClCH₂Cl in Canadian boreal forest fire plumes (background average, June-July 2008, 9.9 ± 0.3 ppt, plume average 10.6 ± 0.3 ppt) and estimated a global boreal fire source of 0.23 ± 0.19 kilotonnes (kt) yr⁻¹.*" Of the references given in WMO 2014 (Chapter 1, page 1.38), this is the only one that reported CH₂ClCH₂Cl.

We have also changed the word "*exclusively*" to "*predominantly*" when referring to anthropogenic sources of CH₂ClCH₂Cl.

I.334: "Production has increased rapidly ...": Can you give a reference for this increase in production?

Reference added (same as in the next sentence)

I. 362: Does the superscript "1" have any meaning? Footnote?

The superscript was actually a missing reference, which has now been added (Carpenter and Reimann et al. 2015).

I. 367: "2"-> "two"

Done

Table 1: Why not use the IATA code "FRA" for Frankfurt (rather than "FFT", which is the IATA code for Frankfort, Kentucky)?

Done

Table 1: Why is the sum of VSLS-CL excluding CH₂ClCH₂Cl not given for the other data for comparison?

The sum of Cl-VSLS excluding CH₂ClCH₂Cl is not a widely used number so it was not included in the Table apart from where necessary. There is no equivalent number reported in WMO. However, we have added the information for the CARIBIC flights between Frankfurt and Bangkok for comparison as suggested.

Reviewer 2

In general, all the information on the samples: time, locations for all the data, number of flights for CARIBIC data, number of sampling at the ground-based stations should be added in this section. For instance, the 7 IAGOS-CARIBIC flights time should be mentioned. Days and months of samples should be specified in this section as well. It will help the reader to get the general feature of the sampling.

The altitude of CARIBIC needs to be shown. Have you filtered IAGOS-CARIBIC data to analyze data between 10 and 12 km only? The statistics of the sampling in this layer is needed.

We have added more detail at the beginning of the methods section:

“A total of 21 samples were collected at Hengchun between 7 March and 5 April 2013 with a further 22 samples taken at Cape Fuguei between 11 March and 4 April 2014. 28 samples were collected at Bachok between 20 January and 5 February 2014, during the period of the NE winter monsoon. The approximate location of each surface site is shown in Figure 1. The CARIBIC aircraft samples were collected during seven return flights between (i) Frankfurt (Germany) and Bangkok (Thailand), and (ii) Bangkok and Kuala Lumpur (Malaysia) during the periods December 2012 - March 2013 (4 flights) and November 2013 - January 2014 (3 flights). All CARIBIC flights in this region between December 2012 and January 2014 have been included in this analysis. With the exception of 3 samples that were taken at altitudes between 8.5 and 9.8 km, the CARIBIC samples were all (n=179) collected at altitudes between 10 and 12.3 km.”

The full CARIBIC flight dates have also been added to Figure 5.

In the text it is mentioned that 10-12km over East Asia is the lower boundary of the TTL. It would be very helpful to show a map of TTL or a figure of TTL and aircraft altitudes together with respect of the flight tracks (latitude). It would be also useful to directly refer to Box 1-3, Figure 1 of Carpenter and Reimann et al. (2015) that shows the altitude range of the TTL.

We have added a reference to the Figure from Carpenter and Reimann et al. (2015). We do not think it is necessary to reproduce a similar Figure here.

About the results shown in Figure 3, a sentence explaining that three days have been chosen out of the seven days of the cold surge event would be helpful. The term “cold surge” should be mentioned.

New text added: *“Three examples during this cold surge event are shown in Fig. 3 (b-d).”*

In general, “see in supplement” is largely used in the manuscript but I would suggest to refer to figure number and section names of the supplement materials to help the reader.

Done

Results from Carpenter and Reimann et al., 2015 are cited as reference for CI-VSLS last information. For results at the lower TTL, it would be useful to recall the type of observations used in the assessment report: aircraft campaigns and balloons.

We have added the following sentence to Table 1: *“Data from the TTL was derived from various aircraft and balloon campaigns.”*

Table: Units need to be added in Table 1 and its caption

Done

In general, the way to write CI-VSLS should be consistent along the captions and the text (sometimes chlorinated VSLS or VSLS-CI).

Done

Figure 1: “(red crosses)” for surface sites on the map need to be added in the caption to guide the reader.

Done

Figure 2: We don’t see the blue circle on the map.

We apologise that the incorrect Figure was included in the original submission. The blue circles have now been added.

Figures 2,3,5: Helpful to have a recall of ground-based stations location.

Blue circles have been added to Figures 2 and 3 showing the location of the surface sampling sites.

Figure 2 and 3: I would rather use letter to name the panels in the caption and I would rather use numbers to link plots in upper panel with maps of the bottom panels.

We use letters to label the arrows which refer to the individual NAME plots underneath. Mixing letters and numbers would, in our opinion, be more confusing.

Figure 3: Use arrows as for Figure 2 to help the reader to find the days that are chosen for air masses origin (map below). In the caption it is mentioned “true background levels”, how these levels are estimated? Figure 3 a) is not specified, “a)” should be added on the figure.

Arrows have been added to Figure 3 as requested. The baseline levels are taken from WMO 2015, based on tropical mean background levels. The actual expected background is difficult to define for VSLS as they would be expected to vary substantially across the globe (i.e. with latitude and with distance from source). We have added a reference to WMO in the Figure caption.

The missing (a) has been added to Figure 3.

Figure 4 and S2: What does CO anomaly mean? What is the reference value?

By the term “CO anomaly” we mean the fraction of CO observed at Bachok from industrial emissions from regions north of 20N. This is explained in Section 3 (lines 285-288) and in the supplement. We have added a sentence in the caption for Figure 4 to remind readers.

Line 126: “Both in the western Pacific region and globally”. It is not clear what globally means.

We have removed the word “globally”.

Line 147: “Various time” needs to be specified (see general comments).

More sampling information has been added (see earlier response to reviewer 2))

Line 150: Change “CARIBIC aircraft” to “IAGOS-CARIBIC aircraft” as CARIBIC is part of the IAGOS program.

Done

Line 230: Change “shows the 2014 data ...” to “shows data from CAPE Fuguei in the end of March, beginning of April 2014”.

Text has been changed to “*shows the March/April 2014 data...*”. In addition the sampling dates have been defined more clearly in the text (see earlier comment of reviewer 2). The dates are also evident in the Figures.

Line 237: Change “March/April 2013” to “mid of March/beginning of April 2013”.

To be consistent with the text in the previous comment we have not made this change but note that the sampling dates have been defined more clearly in the text (see above). The dates are also evident on the Figures.

Line 246: “January/February”: the entire months are not shown so “end of January/beginning of February” would be more appropriate. “During this phase of the monsoon”: A recall about the Asian Monsoon phases and references would be useful, maybe recall that it is the East Asian winter monsoon circulation as mentioned in the introduction.

Text has been changed to “*late January/ early February 2014*”. Sampling dates have been clarified in methods section.

We have added the words “*East Asian*” when referring to the monsoon and reminded readers that this was described earlier.

Line 252: change “often” to “most of the case studies we are analyzing here” or “for N days out of Ntotal days of observations” or give the information in Line 277:

We would prefer to keep the word “often”. This was deliberately vague as we have not done any specific analysis to determine how many times the air may have picked up emissions from Taiwan. The observation was also based on the NAME animations which are referred to in line 285.

Change “(see supplement for further details)” to “(Fig. S2 in the supplement material)”.

Done

Line 287: Change “in all CARIBIC flights” to “in the seven CARIBIC flights” and remove “(7)”.

Done

Line 334: Need a reference.

Reference added

Line 362: Need a reference.

Reference added

Line 383: Examples of other chemical pollutants would be useful.

The pollution we were referring to are chemicals that are routinely found in industrialised countries. These include CO, O₃, CH₄, volatile organic compounds (VOCs) including hydrocarbons, oxygenated hydrocarbons and certain halocarbons. These measurements are likely to feature in a future publications.

Line 384: Remove “etc”.

Done