Response to reviewer 1

Reviewer comments in blue and our response in black. All page, figure and table numbers refer to the Discussion manuscript. As the reviewer comments were listed per page and then numerically we refer to other reviewer comments using the following numbering scheme: #Page 20723/point 5 for the 5th point made about page 20723.

We thank the reviewer for their detailed review of our manuscript.

General comments:
Have all been addressed through our responses to the specific comments.

Page 20722:
1) L2-5: These sentences need references and are likely better placed in the Introduction.

We have reduced some of the detail pertaining to previous trends in CH_2Cl_2, as the focus of this paper is on the recent increase and have updated the opening sentences to read: "Atmospheric concentrations of dichloromethane, CH_2Cl_2, a regulated toxic air pollutant and contributor to stratospheric ozone depletion, were reported to have peaked around 1990 and to be declining in the early part of the 21st century. Recent observations suggest this trend has reversed and CH_2Cl_2 is once again increasing in the atmosphere." Following the guidelines set by ACP the abstract should not include references "unless urgently required" (see: http://www.atmospheric-chemistry-and-physics.net/submission/manuscript_preparation.html). The statements made are general, e.g. "an increasing atmospheric burden throughout the 1900s" to remove a pressing need for references.

2) L6: CH_2Cl_2 is a fairly minor contributor to overall stratospheric ozone depletion. Elsewhere in the text please provide specifics on the importance of CH_2Cl_2 relative to the other ozone depleting substances.
We have added 'minor' to the abstract where we mention CH_2Cl_2 as a contributor to stratospheric ozone depletion. We have added the % contribution of CH_2Cl_2 to total chlorine from organic compounds at 15 km based on Laube et al. (2008).

3) L8: The Simmonds et al. paper from 2006 reported CH_2Cl_2, C_2HCl_3 and C_2Cl_4. Did CARIBIC also measure other short-lived chlorinated compounds? Just curious why the focus here is limited to CH_2Cl_2.
CARIBIC measures a range of other chlorocarbons but we decided to focus on CH_2Cl_2 due to the recent increase in atmospheric abundance. To repeat the detailed regional analyses of the CH_2Cl_2 time series for other gases would have vastly increased the length of the manuscript and reduced clarity of our overall message. Analysis of further chlorocarbons may be forthcoming, but is beyond the scope of this response.

4) L13: Global monitoring networks monitor CH_2Cl_2 at the Earth's surface. Is the CARIBIC time series being called 'unique' because it is collected at high altitude?
Yes, as far as we are aware CARIBIC is the only long-term (spanning > 15 years) airborne platform reporting halocarbon measurements.

5) L16: The impact of seasonality on the long-term increase estimates needs to be presented.
We have added a new table (Table 3 in the updated manuscript) which provides the seasonality. The table is in the same style as the one shown as part of our response to #Page20731/point 2. We also address this point throughout the rest of our response.

6) L20: Likewise the possible impact of interannual variability on the long-term increase estimates needs to be discussed.
Addressed as part of our response to other comments.

Page 20723:

1) L2: Provide a reference for the 90% industrial origin. Simmonds et al. (2006) suggested that ~70% of CH_2Cl_2 emissions were anthropogenic.

This reference is Montzka et al. (2011b) page 1.41, and refers to the total possible anthropogenic emissions. We wrote '**up to** 90%' to draw attention to the fact this was the largest estimate and that there are differences in this estimation, e.g. as discussed in Simmonds et al. (2006). One difference may arise from the fact that whilst both Montzka et al. (2011b) and Simmonds et al. (2006) use the term 'anthropogenic' Montzka et al. specify that this term includes industrial emissions and biomass burning. We have updated this sentence to read: "Dichloromethane, CH_2Cl_2, is a short-lived chlorocarbon of mainly (up to 90 % **(Montzka et al., 2011b)**) **anthropogenic** origin." to include a reference and remain faithful to the terminology used in said reference.

2) L5: Simmonds et al. used 10% to force their model to better fit Mace Head and Cape Grim observations, and they also tested 30% from natural sources. Please revise the text to reflect the uncertainty in natural sources.

We have updated this sentence to read: "The contribution from natural sources (mainly biomass burning and an oceanic source) is uncertain. Simmonds et al. (2006) obtained a good model fit to their observations using a 10% combined oceanic and biomass burning source, although they showed that a stronger terrestrial source could support natural emissions of up to 30%. However, recent field measurements of biomass burning plumes have indicated that this source is likely to be smaller than previously estimated (Simpson et al., 2011)."

3) L14: Specify that this is a ratio for the lower troposphere.

Done.

4) L16-17: Please provide a reference. This sentence can probably be combined with L22-24.

This section has been restructured as part of our response to #Page20723/point 5.

5) L16: The opening sentence of this paragraph is about trends; expand the discussion of trends for the SH, for example by folding in the trends reported at Cape Grim by Simmonds et al.

We have rearranged this section of the Introduction. Previously it was structured:

Introduction to CH_2Cl_2 (paragraph 1) > Introduction to the previous CH_2Cl_2 measurements and the post 1990s decline (paragraph 2) > Atmospheric impacts of CH_2Cl_2 and indications that it is now increasing again in the atmosphere (paragraph 3).

For clarity we have restructured as follows:
Introduction to CH_2Cl_2 (paragraph 1) > Atmospheric impacts of CH_2Cl_2 (paragraph 2) > NH measurements of CH_2Cl_2 followed by SH measurements, which has been expanded as suggested above up to c.2004 (paragraph 3) > the current observed increase in CH_2Cl_2 in both the NH and SH and that this is the rational for this study (paragraph 4).

As part of this restructuring we have addressed both point #4 and point #5 by combining the sentences on L16-17 and L22-24 (they share the same reference) and including a discussion of the SH trends.

Page 20724:

1) L2-3: Provide some of this discussion here. For example cite the long-term downward trend at Mace Head from 1998-2004 reported by Simmonds et al.

Addressed as part of our response to comment #Page 20723/point 5.

2) L4-8: This discussion needs to be more quantitative. The contribution of short-lived chlorocarbons to stratospheric ozone depletion is minor, even considering the declining concentrations in long-lived halocarbons. Quantify the percent contribution of CH_2Cl_2 to ozone depletion relative to the other compounds.
Addressed as part of our response to #Page 20722/point 2.

3) L8-10: Please provide a reference.
This sentence has been removed as part of our response to #Page20722/point 2.

4) L10: Provide a reference for the predominantly natural origin of $CHCl_3$.
This sentence has been removed as part of our response to #Page20722/point 2.

5) L12: (1) Be careful about citing a growth rate (8%) based on one year of change : : : a one year growth rate isn't too meaningful without understanding year-to-year fluctuations in concentration, especially for a short-lived compound. This also contrasts with the NH declines presented in the previous paragraph. (2)Try to synthesize the trends in CH_2Cl_2. For example the 1990 peak, decline till the mid-2000s, and apparent increase in recent years could be presented together in the same paragraph.
(1) A growth rate of 8% is given implicitly in the 2010 WMO report (we did not infer it from their data) and we provided the year range (2007-2008) in our manuscript to inform readers that they had based this increase only on a one year period. We also follow this with the additional Montzka et al. (2011a) reference which showed the increase over a longer time period. The fact that the WMO2010 reported increase was based on only a short time period was part of the motivation for this study.
(2) We have addressed this as part of our response to #Page 20723/point 5.

6) L15: (1) The statement that $CHCl_3$ has 7% natural origin contradicts line 10, which said that $CHCl_3$ is primarily of natural original. (2) Also the Worton paper discusses a time-varying anthropogenic contribution to $CHCl_3$, peaking in 1990. (3) In general the introduction needs tightening; often the same idea is presented several sentences apart.
(1) This should read 70% and has been updated.
(2) Worton et al. (2006) do report a time-varying anthropogenic contribution to $CHCl_3$. However, in our manuscript we wrote: "Montzka et al. (2011b) reported increasing CH_2Cl_2 concentrations in recent years, with an increase of around 8% between 2007 and 2008, based on updated AGAGE data from Simmonds et al. (2006). There was no corresponding increase in $CHCl_3$, 70% of which is believed to be of natural origin (Worton et al., 2006)". So we are referring to a defined time period during which the ~30% anthropogenic contribution applies.
(3) We have addressed this as part of our response to #Page 20723/point 5.

7) L18-19: Please define EU and provide a reference to the regulations. Also according to the IARC CH_2Cl_2 is only a possible carcinogen; please clarify the text.
Done.

8) L20: General comment: The importance of CH_2Cl_2 needs to be put into perspective. CH_2Cl_2 is only a minor contributor to ozone depletion, a possible carcinogen, and its ambient concentration falls far below its permissible short-term exposure limit (12.5 ppm in 8 hours).
We have improved our explanation of the importance of CH_2Cl_2 with respect to ozone depletion and as a carcinogen as part of our response to your comments #Page 20722/point 2 and #Page 20724/point 7. Due to the restructuring of the introduction (see our response to #Page 20723/point 5) the sentences refer to here no longer exist in this form.

Page 20725:

1) L4: (1) Please provide basic information about this phase, including the total number of whole air samples that were collected. (2) Also reference Figure 1 here.

(1) The total number of air samples collected during either CARIBIC phase is not relevant to this manuscript as some flight routes are not discussed (as mentioned in the first paragraph of Section 3, we selected only routes which were visited over a range of years to allow us to determine temporal changes). Throughout the text, and in particular in Tables 1 and 2, we provide details of the sample numbers for relevant flights, routes, time periods etc. (also in the newly created Table 3).

(2) Figure 1 is specific to the flight routes selected for use in this study, whereas line 4 is providing an overview of CARIBIC1. Page 20725 line 1 directs readers to our website if they require further information, a figure of all flight routes can be found there.

2) L13: (1) There are 28 samples/flight. Please state the typical interval between samples. (2) Also change the semi-colon to a comma.

(1) The interval between samples varies by each flight route. Fig. 1 provides the distribution of samples along each flight track.

(2) Done.

Page 20726:

1) L9: Change 'to allow for' to 'to allow correction for' or something similar.

Done.

2) L22: Change the comma to a period.

Done.

Page 20727:

(1) 1) L12: If the precision of the V.G. Autospec is better, why was it replaced by the Entech system?

2) L14-22: This paragraph is confusing. The V.G. Autospec was replaced by the Entech system, but now there is a Waters Autospec that is the direct replacement of the V.G. Autospec. Are there now two instruments, the Entech and the Waters Autospec? If the Waters Autospec system has higher precision, why is the Entech system being used? (2) Also while comparison data have been presented (e.g. +/- 1 ppt agreement) please state the detection limit, precision and accuracy for each of the three instruments so their capabilities can be directly compared. (I see precision is included later on P20728; present it here or refer the reader to below.)

(1) The Entech-MSD instrument was bought as a dedicated fully-automated workhorse instrument capable of a rapid turnaround of samples. The Autospec instrument is a labour-intensive manual system not suited to the multiple sample analysis that is required by CARIBIC. The Autospec instrument is also widely used for other projects. We have made several small changes to this paragraph to try and clarify the use of these instruments changes, mainly altering page 20726 line 11 to show that the V.G. Autospec was replaced as the main CARIBIC halocarbon instrument (not replaced physically).

(2) Around 20727 line 3 we direct readers to the precision etc. listed in the penultimate paragraph of the methods section. The limit of detection for all 3 analytical systems was 0.1 ppt or better. The CH_2Cl_2 data are reported on the latest NOAA-ESRL calibration scale (2003). NOAA do not provide an absolute accuracy on their calibrated gas standards but, in a recent international comparison exercise (IHALACE), the mean of the CH_2Cl_2 calibration scales from the three independent calibration laboratories was found to have a standard deviation of ± 9% (Hall et al. 2014). We have added both these facts to our method.

3) L29: Much of the analytical detail on P20726 (column details, temperature programs, etc.) could also be moved to the Supplementary Material.
As our manuscript presents a time series we felt it was important to detail the steps taken to ensure consistency over time – e.g. the duplicate analysis on two systems (Entech & Autospec) and the standard intercomparisons – and to keep this information in the main manuscript. As this means a large proportion of the methodology had to be in the main manuscript we kept all details here to prevent readers having to flick backward and forward between two documents.

Page 20728:
1) L1-10: This paragraph could be tightened.
We have restructured parts of the manuscript and feel this section is OK as it is.

2) L8: 'compare well' and 'no apparent offset or change' is too vague.
We have updated this sentence to read: "Cape Grim samples analysed by both groups compare well (65% agree within the respective 1 sigma standard deviations), with no apparent offset or change in the relationship between both groups' results over time (NOAA/UEA ratio of 1.02±0.06)."

3) L16-18: The stated precisions have too many significant figures.
Significant figures have been reduced.

4) L19: As well as precision, detection limit and accuracy need to be stated.
Addressed as part of our response to #Page 20727/point 2.

5) L21-29: This paragraph could be tightened, with less qualitative description and some examples of the seasonal ozone thresholds that were used.
We have added in the equation used to determine the O_3 tropopause, two extra references and moved the more quantitative description of this method that was in Section 3.4 to this section of the manuscript.

Page 20729:
1) L3: Without some indication of the O_3 thresholds that were used the reader can't assess whether the 6-40% rejection is based on conservative estimates of stratospheric air.
See our response to #Page 20728/point 5.

2) L5: (1) Please state the total number of non-stratospheric samples that were kept and how the number varied from year to year. (2) Does Figure 1 show all the samples that were collected or just the non-stratospheric samples?
(1) We have added the total number of samples (trop. and strat.) to Table 1 for all regions.
(2) We have updated Fig. 1 to show which samples were labelled as predominantly tropospheric or stratospheric.

3) L6: Tell the reader the purpose of the CO measurements. Why only South Africa and India?
CO is used where it assists us with interpreting the CH_2Cl_2 results. Where CO is used in the discussion of results its purpose is described, including relevant references. Section 2 is for the description of any appropriate methodologies.

4) L6: The verb tense is changing from past to present; past is more appropriate.
We have corrected this.

5) L6-8: (1) The availability of CO measurements was given on P20725. (2) L18-19. Why does P20725 reference Brenninkmeijer et al. (2007) for CARIBIC2 while here it's Scharffe et al. (2012)?
(1) Page 20725 only mentions that CO measurements are made by CARIBIC, as part of our introduction to the CARIBIC platform. Analytical CO details are provided on page 20729 alongside the analytical details of all other ancillary measurements.
(2) The section you refer to on page 20725 is the introductory paragraph of the method section and is introducing the CARIBIC platform. Benninkmeijer et al. (2007) is an overview paper describing the CARIBIC2 system as a whole. Therefore it is a suitable reference for the section: "The fully-automated CARIBIC2 system contains a range of other sampling equipment, including, but not limited to, equipment for the in-situ or post-flight analysis of ozone (O_3), carbon monoxide (CO), aerosols and water vapour. Further information can be found in Brenninkmeijer et al. (2007)." On page 20729 we are describing just the CO system. Scharffe et al. (2012) is a specific paper for the CO system and so is referenced here.

6) L10: There is no need to abbreviate 'integrated' as 'int.'
In the x-axis of Fig. 5 'int.' is used and so we abbreviated it upon first use here. However, as there is no other use of this abbreviation we have moved this information to the Fig. 5 caption.

7) L12: Change the comma to a period or semi-colon.

8) L20: 'a substantial portion' is likely overstated; in reality it is probably a very small percentage of the global free troposphere, especially because the measurements are limited to the upper free troposphere.
We have changed 'portion' to 'area'.

9) L21-22: (1) So are only these five regions considered in this paper? (2) Are the data in gray in Figure 1 not used? (3) Overall please state what percentage of the data were used and how many samples this corresponds to. (4) This is a 14-year data set and it seems that the data coverage could in fact be quite sparse both spatially and temporally, so not robust enough to pull out long-term trends.
We believe this paragraph clearly states the reasons for selection and use of only these 5 regions. Due to the fact that CARIBIC does not provide 100% global coverage we do not make claims regarding global trends and instead focus on regions where CARIBIC flew over multiple years, spanning a time period long enough to see changes in CH_2Cl_2 as well as selecting regions which covered different areas of the globe.

(1) We have clarified this by modifying the opening section of this paragraph to read: "Between 1998 – 2012 CARIBIC flights covered a substantial area of the global free troposphere (Fig. 1). However, for the purpose of this investigation several regions were selected and only the data from these flights will be discussed. These regions and the rationale behind their selection are described here."
(2) The caption for Fig. 1 states that "Flight routes used in this study are highlighted", the fact that only these routes are used has been clarified, see (1).
(3) As we do not use a subset of the whole CARIBIC dataset (our regions) to derive global changes (we only discuss regional changes) we are not sure how providing these numbers will provide anything of benefit to this manuscript. Fig. 1 provides a visual overview of the subset of data used compared to the whole dataset. Sample numbers for each region, which show how many samples are discussions/conclusions are based upon, are given, in detail, in Tables 1 and 2.
(4) We are clear throughout our work what sample size we base our results upon, providing 'n=' in the text or directing readers to Tables 1 and 2. Table 1, in particular, provides details not only on the total sample number for each region but also how this is divided into the different time periods or seasons (Indian monsoon flights). See also our response to other comments (mainly #Page 20722/point 5) and our newly created Table 3.

10) L22: Is this the box shown in Figure 1? If so please state this in the Figure legend.
The inset figure legend for Fig. 1 labels the box as 'European region' - we have expanded this legend to make it clearer.

Page 20730:

1) L3: The legend in Figure 1 is confusing. What does the colour gray indicate? Aren't the regional samples (Africa, etc.) also CH_2Cl_2 samples? Some indication of the time frame for the different routes also needs to be given.
We have improved this legend. Time frames are provided in Table 1, we have now mentioned this in the Fig. 1 legend.

2) L4: What does 'this average' refer to?
We have changed this sentence to read: "Throughout the manuscript mean values prefaced by ± refer to the 1 sigma standard error associated with that mean value." Wherever a mean value is used in the text its origins are provided.

3) L6: (1) The Figure 2 legend is confusing. The text says 'NOAA Mace Head data' but Figure 2a shows a gray dot for 'NOAA' and a black diamond for 'Mace Head' (it took me a few minutes to work out that half the legend in Figure 2a is actually for Figure 2c). Please fix. (2) Also I recommend presenting Figure 2c with Figure 1 and discussing it earlier in the text, say on P20725 L4.
(1) We have made general improvements to the Fig. 2 legend and description.
(2) We present Fig. 2c as part of Fig. 2, not Fig. 1, as it is directly relevant to the discussion in of Fig. 2 in Section 3.1, for example the differences between NOAA Mace Head and CARIBIC data due to the wide geographical spread of CARIBIC samples within the European region.

4) L6-7: Instead of 'can be seen' use 'is shown'.
We prefer "can be seen" to activate the reader.

5) L8: The font size or figure size needs to be increased in Figure 2.
All figures were submitted in a size-independent format, figure sizes are at the discretion of ACPD. Font sizes on all figures can/will be adjusted for final publication as necessary.

6) L9: Try to use more direct language. Instead of 'is mainly as a result of the fact that' use 'is mainly because' or something similar.
Updated.

7) L10: Could the lower variability in the Mace Head data also be because these data have been edited for outliers?
The data are not edited for outliers. Pairs of flasks are filled for each measurement and data are only removed if the agreement between these flasks is greater than 0.66 ppt (a situation that occurs in around 2% of samples), suggesting a problem with sampling or analysis. No flask pair results were deleted on the basis of the flask pair average being anomalous. However, sampling is done in a manner to characterise only air that is arriving at Mace Head from the clean air sector, specifically when wind direction is between 180° to 320° and the wind speed is > 4m/s. We have clarified this fact in our manuscript.

8) L11-15: Even though Mace Head is a clean air site and CARIBIC can be impacted by pollution, the CARIBIC levels are still generally lower than the Mace Head values, most likely reflecting the vertical gradient of CH_2Cl_2 in the atmosphere. This should be briefly pointed out and discussed here.
This is discussed on page 20730 lines 10-13.

9) L16-17: Suggest rewording this because seasonality seems apparent in later years.

Our current wording "we do not see a strong seasonal pattern" – even in later years the seasonality is not as strong as observed in the less-sporadic NOAA Mace Head data.

10) L21: (1) While the Mace Head data are fairly evenly distributed so that annual averaging is reasonable, please describe how the CARIBIC annual averaging was done. Was any weighting done, for example averaging 12 monthly averages, or was it a straight average of all the data points in a given year, in which case the averaging could be biased if the data points are clustered. What tests were done to make sure the annual averaging calculation is robust? (2) Also CARIBIC data time series need to be shown for each region, not just Europe. The reader needs to be able to see the gaps in the data relative to the seasonality, and whether the long-term averaging and trend analysis is reasonable.

(1) Firstly – we do not call it an 'annual average' on line 21 we write "the mean of all tropospheric samples taken within each year" due the fact we realise that the CARIBIC samples are not necessarily spread evenly throughout each year. I think this sentence also conveys how we calculated the average (averaging all values collected within 1 calendar year). We attempted to use this terminology throughout the manuscript, e.g. when page 20732 line 4 when describing the same averages for the Africa data. However, we recognise that we slipped occasionally, e.g. in the Fig. 2 caption where we did call it an "annual mean" – we have now checked all discussion of these averages and made sure to carefully describe it more carefully.

Secondly - we made several attempts to address the possibility of seasonality; Table 1 included a list of months sampled and on page 20730 line 22 onward it is mentioned.

Thirdly - due to the possibility that different months may be sampled in different years we do not discuss year-by-year increases and group years where possible.

Fourthly – as another method of avoiding the impact of seasonal bias we calculate the increase between the combined 1st four years of the dataset and the combined last four years of the dataset to increase the sample size and reduce the impact of seasonal bias.

(2) see our response to #Page 20722/point 5, #Page 20731/point 1 and our new Table 3.

11) L25-28: This discussion is very qualitative. What does 'very similar' mean? Does CH_2Cl_2 have a latitudinal gradient between Barrow and Mace Head?

This section is qualitative as it is a supporting statement designed to provide reassurance that we are dealing with the issues described in your previous comment. The gradient is minimal; the annual mean ratio (Barrow/Mace Head) from 2003-2013 is 1.1(±1.6) %. However, as we are discussing the similarities in trends which are independent of any latitudinal gradient.

Page 20731:

1) L2-5: This is also qualitative. What does 'relatively stable'mean? It seems very difficult to tell whether the concentrations are in fact relatively stable when a good portion of the data, including two sets of winter data, is missing. Do the data actually increase beginning in 2004, or does the increase begin more around 2006? What do linear fits to different subsets of the data show?

We have improved and restructured this section. We have rearranged this section to discuss the Mace Head data first, which provides the better data coverage. We provide the results of linear fits in Fig. 2 and discuss them in the text to show an increase from 2006. We improve our discussion of the limitations due to the limited data coverage in the early years of the CARIBIC database, including the steps taken to validate the increases seen here. We have added values to support qualitative statements. Also see our response to #Page 20731/point 2.

2) L4-8: I am concerned that trends are being deduced from data sets that are incomplete for each time period. The 1998-2001 CARIBIC data set for Europe (n = 16) is extremely sparse and almost entirely missing wintertime data, when concentrations will be the highest, whereas the last four years (2009-2012) have relatively more wintertime data. Therefore the latter data set can be expected to have higher concentrations on the basis of seasonality alone. The pre-2002 data do not seem nearly robust enough to be used for long term trend determination. At very least you would need to compare data from similar months, but even this is questionable because unlike Mace Head, which is a fixed location, the location of the flight paths from CARIBIC may be different, introducing another difficulty in the comparison. A better starting point would be taking the Mace Head 2003-2004 data as a baseline and comparing it with Mace Head 2011-2012 data. What increase does this show?

Firstly – the seasonality of sampling is shown below. For the CARIBIC data the early data is predominantly summer based compared to a winter/spring mix for the later period. However, the early period covered my Mace Head is much more evenly spread throughout the year and, as we describe on page 20731 line 8 the increase is very similar at both regions. This may be due to the fact that the seasonal cycle is weaker between 10-12 km, as we reported on page 20730 lines 15-18.

This figure shows the months when sampling occurred for both periods. If a number is included the month was sampled during 2 or more years (number referring to the number of years).

Mace Head	J	F	M	A	M	J	J	A	S	O	N	D
1998-2001											x2	
2009-2012	x4	x4	x3	x4	x4	x4	x4	x4	x4	x3	x4	x4

CARIBIC	J	F	M	A	M	J	J	A	S	O	N	D
1998-2001					x2	x3	x2	x3				
2009-2012	X2		x3	x4	X4	X3	X2	X5	X4	X4	x2	

Secondly – we have not discussed increases per amount of time (e.g. 10 ppt yr^{-1}) and we agree that our data do not have the coverage to calculate a trend of this manner.

However, we do recognise the reviewers concerns and have addressed this as follows. Firstly – we have rearranged this section to discuss the NOAA data first (see #Page 20731/point 1) which has the higher data coverage, making it clear that this is the best indicator of trends and that the CARIBIC data fit within this trend. Secondly – we have increased the earliest period to 1998-2002 (previously 1998-2001) which increases monthly coverage as shown in the diagram below (changes shown in orange). This provides some extra coverage, in particular to the Mace Head data (remembering that the seasonal cycle is stronger here). Thirdly – we retain our comparison between 1998-2002 and 2009-2012 as this covers the largest time period. However we add the suggested comparison for Mace Head 2003-2004 and 2011-2012 data. This increase was from 33.6 ppt (±0.7, n=48) to 47.0 ppt (±1.0, n=45), an increase of 13.5 (±1.2) ppt. This is still a substantial increase and supports our original findings. We have added an additional mention of the possibility of seasonal bias alongside this additional comparison period to demonstrate that the increase is independent of seasonal bias.

Mace Head	J	F	M	A	M	J	J	A	S	O	N	D
1998-2002	x2	x2		x2						x3	x3	x1
2009-2012	x4	x4	x3	x4	x4	x4	x4	x4	x4	x3	x4	x4

CARIBIC	J	F	M	A	M	J	J	A	S	O	N	D
1998-2002					x2	x3	x2	x3				
2009-2012	x2		x3	x4	x4	x3	x2	x5	x4	x4	x2	

3) L11-13: It is almost certainly because of the altitude difference so I recommend discussing vertical gradients of CH_2Cl_2 earlier in the text (see also comment for P20730 L11-14).
We have moved the discussion of the standard comparison (see #Page 20731/point 4) to Section 2 to reinforce the fact that the difference is as a result of the altitude difference. We now mention the altitude difference first, following it with a brief reminder of the intercomparison. We have added a reference here to the vertical profiles in Sections 3.4 and 3.5. Altitudinal profiles are most pertinent in the tropical region where the most efficient transport pathway to the stratosphere occurs (page 20742) and so have left a discussion of the vertical profile to the later sections. One reason that vertical profiles do not form a more prominent aspect of this manuscript is that CARIBIC mainly flies within a relatively narrow band of 10-12 km (for the European region we are discussing here the mean altitude was 9.8 km with a standard deviation of 1.8 km).

4) L15: Why is the Cape Grim data presented if it's not going to be discussed?
The (brief) mention of Cape Grim data is to provide additional support to our comparison of CARIBIC and Mace Head data. The Cape Grim samples provided the best comparison of NOAA Mace Head and CARIBIC samples, as we explain in the Supplement. To avoid further confusion we have cut the mention of Cape Grim data from this section (see response to #Page 20731/point 3) – it has already been discussed in Section 2 which directs the reader to the Supplement for further information.

5) L17: Following from the above comments, I am concerned that the trends in Table 2 are questionable and will be misinterpreted; I recommend using more robust portions of the data set from long-term trend analysis, if the sample size and data coverage is adequate.
See our response given #Page 20731/points 1 and 2. All changes have been updated in Table 2. For the European region we have added the increase for the NOAA Mace Head data between 2003-2004 and 2011-2012 (as above) which supports our increasing trend between 1998-2002 and 2009-2012. Other regions are discussed in later responses, but briefly: Africa – The increase >30 °N is based on only 3 samples during the early period. No flights were made to Africa between 2000 and 2009 so we cannot select other time periods. In the text we have kept a qualitative discussion of the increase >30 °N and clearly state that we see an increase but based on a small sample size. We have removed this increase from Table 2, where people may read it without the associated caveats. India – we have added in additional results from 2011-2012 (n=30). All the other datasets have suitable sample sizes and adequate seasonal coverage (further details in Table 1 or throughout the manuscript). Throughout the manuscript we have reduced the use of s.f, improved the discussion of our caveats and generally improves our description (qualitative and quantitative) of the CH_2Cl_2 increase.

6) L17-18: (1) The Simpson et al. altitudes were lower than CARIBIC (0.8-4.7 km instead of 10-12 km).
(2) Also the Simpson et al. measurements were only performed in the summer months so a better comparison would be with the CARIBIC summer data from 2008, though the effect of altitude still needs to be discussed. (3) Please also provide an uncertainty on the average of 35.8.
(1) We have clarified this detail.
(2) We have replaced out 2008 average for summer-only measurements. We include not only the mean summer value for 2007, 2008 and 2009 but, as these were small sample sizes (n=6, 4 and 6 respectively) the combined value for these three summers. We mention the effect of altitude.
(3) This was in Table 2 – we have moved it to the main body of the text.

7) L22: The legend for Figure 3a has many more colors than the three that are shown.
To provide consistency across all figures we used the same colour scale for all figures to allow for a quick comparison between figures. We have added a brief explanatory note to this effect.

8) L24-25: The data are too sparse to infer any kind of long-term trend.
See our response to #Page 20731/point 5.

9) L25: Were the flights in 2000 and 2009-2011 made in the same season? Also provide uncertainties on the averages.
This section has been altered/removed - see our response to #Page 20731/point 5.

Page 20732:
1) L1-3: This conclusion is difficult to support without a more robust data set and full modelling analysis to understand all the sources and sinks of CH_2Cl_2.
We do not provide a conclusion. We clearly state this result is 'indicative'. This paper is not the last word on the issue, but a step forwards.

2) L5: From Table 2 there are 3 samples in the north of 30 Africa data set. It is not possible to present a meaningful trend analysis with such small data sets. Please remove this discussion.
See our response to #Page 20731/point 5.

3) L8: Why does Table 2 present north of 30 data for Africa for 2000 but the text cites 1998-2001? How many samples are in the 1998-2001 data set? From Figure 3a it still only looks like a handful of data points and long-term trend analysis is not credible. Most of the discussion in this paragraph should be removed.
This section has been altered/removed - see our response to #Page 20731/point 5.

4) L16: Change 'trace gases' to 'trace gas emissions'.
Done.

Page 20733:
1) L6: Put 2011 before 2012.
Done.

2) L12-14: This is a bit confusing because flights before, during and after the ASM would cover all the 2008 data. Please clarify what time frame you mean.
We have simplified this sentence.

3) L21: The elevated samples resulting from Southeast Asia influence is a good illustration of the difficulty in using small numbers of flights to assess long-term trends with time.
We have addressed your points relating to small sample sizes, e.g. #Page 20731/point 5.

Page 20734:
1) L1-10: The discussion is weakened when too much weight is placed on snapshot measurements. Is this a shift in latitude with time or just variability? More analysis and discussion is needed before such conclusions can be drawn.
We have added to this paragraph to make it clearer. It now follows this pattern: Describes what we seen in the early India flights (a N-S latitudinal gradient) > describes that this isn't apparent in the later flights and that this is in contrast to other regions where the latitudinal gradient remains stronger than it does over India > concludes that this indicate increasing emissions from India. It then goes on to highlight the one flight in 2011 with exceedingly high CH_2Cl_2 to demonstrate that we have looked into these results further.

2) L12: How large was the 'small sample size' in 2011-2012? Why weren't the 2000 Africa data north of 30 degrees (n = 3) omitted for the same reason?
We have addressed the small sample size for Africa. We have included the increase including the 2011-2012 India values as part of Table 2 and have expanded the discussion to match. See also our response to #Page 20731/point 5.

3) L13: There are too many significant figures. It is not possible to measure a change in CH_2Cl_2 to within 0.1 pptv over 10 years, especially with a relatively sparse data set. The impact of variability needs to be realistically addressed.
We have reduced the s.f. throughout the paper, improved our discussion of variability (see previous responses) and updated Table 2.

Page 20735:
1) L1: Change 'us' to 'use'.
Done.

2) L2: Change 'CH_2Cl' to 'CH_2Cl_2'.
Done.

3) L6: Is it realistic to estimate emissions based on just 35 samples?
The emission estimate method is based on previously published literature using similar or smaller number of samples (Baker et al., 2012; Scheeren et al., 2002). A smaller sample size does increase the potential influence of variability upon our assumption that the ratio seen in our sample set is representative of the actual ratio. To account for this we perform the bootstrapping procedure detailed within the manuscript which shows that despite the variability in the measurements significant changes in emission estimates can still be observed.

4) L10: Fix the subscript. Also show Equation (1) immediately after it is cited.
Done.

5) L14: The justification for not using an excess concentration needs to be briefly presented here (not just referring the reader to two other papers).
This section has been restructured as the full discussion of why we used this method – the benefits and assumptions are all provided in the subsequent sections.

6) L19: CO is a combustion tracer and CH_2Cl_2 is primarily industrial. Why is CO's biofuel source being highlighted here? How large is India's biofuel source of CO compared to its other CO sources such as fossil fuel and biomass burning?
CO is a general combustion tracer but, in India the combustion of biofuel is "the single largest contributor to CO emissions in South Asia" (Dickerson et al., 2002 p. 16-4 – we cite this paper on line 19). Table 2 in Dickerson et al. (2002) gives CO emissions from India in 1999 as being dominated by the burning of biofuels which released between 16.7 and 42 Tg CO yr^{-1} (the range coming from a range of different studies) and when combined with the burning of agricultural land and forests released a total of between 31.5-85 Tg CO yr^{-1}. For comparison, the estimates they had assimilated placed petroleum/gasoline combustion as a source of 9.4 Tg CO yr^{-1}. Even with the uncertainties associated with these estimate, the contribution of combustion of biogenic material is dominant. We have added to this sentence to highlight that CO is a general combustion tracer and that in India the largest portion of this source will be the combustion of biogenic material.

7) L20: This argument isn't convincing. Even if CO is the best option it still doesn't guarantee that it's a good option. Also remove the comma after 'Other'.
We have restructured this section (combined with our response to #Page 20735/point 6) to try and clarify why we have used CO for these estimates. We believe CO to be a good option based on the fact that both compounds are co-located within the monsoon anticyclone (and, as we demonstrate elsewhere, these air masses are isolated), both show strong signatures from the Indian subcontinent and we build on successful analyses comparing these two compounds in a wealth of other studies. We also provide evidence later in this section that this relationship stands up to scrutiny with respect to the potential influence of transport times/routes (page 20736, in particular line 17 onward) and is unique to the Indian monsoon samples (i.e. the correlation is linked to the emissions and not dominated by sampling air of different ages leading to a range of different concentrations).

8) L27: Do not hyphenate 'air masses'. In Table 3 add a space between 'airmass'.
Done.

9) L26: What was the correlation between CO and CH_2Cl_2 in the 35 samples? Is there evidence that they were co-located? (I see that it is presented later : : : please provide it here.)
We have moved the introduction to Fig. 5 to the start of this section.

Page 20736:
1) L1-2: Were all the referenced comparisons between CO and CH_2Cl_2 based on surface studies or were any high altitude studies like yours?
Millet et al. (2009), Palmer et al. (2003) and Scheeren et al. (2002) (discussed earlier in the paragraph) were all aircraft campaigns but used boundary layer measurements. Gentner et al. (2010) and the Millet et al. (2009) Mexico values were ground-based. We have added this information into Table 3 to make this clearer. Our ratios are age corrected with respect to CO which improves our ability to compare our ratios with those taken at a lower altitude. The meteorology of the summer monsoon (page 20733) also supports our attempts to compare these emission ratios.

2) L15: Add error bars to all slopes given in Table 3 and if possible to the Baker et al. slope (the significant figures will probably need to be adjusted). In the 'other study' part of Table 3 adjust the significant figures (e.g. 'Urban, California' should be 3.1 not 3.10 if the error is 0.3).
These errors were originally omitted as they were smaller than the errors arising from the bootstrapping procedure and we wished to present the largest source of error for the clearest understanding of our emission estimates. However, we will include them in the updated manuscript. We had added errors to Table 3 and for the Baker et al. slope. Significant figures in Table 3 have also been updated.

3) L24: Some of the arguments on P20735-36 seem weak. Better correlation in the India samples than the Africa samples still doesn't exclude some impact of transport or different lifetimes on the CH_2Cl_2/CO ratio.
We never state that our attempts to define the potential sources of error in our emission ratio calculations can 'exclude' certain variables. Instead we combine multiple approaches. Individually these may not justify our entire approach, but together they provide a strong basis on which we built our emission estimates.

One example, the following **five** methods were all used to provide support to our assumption that our measured emission ratio (at 10-12 km) was suitably representative of the source emission ratio:

1. Age-correcting data, based on published methods, to account for losses of CO.
2. Looking at the potential impact of transport times using the Indian vs. Africa correlations (the e.g. given above).

3. Looking at the potential impact of transport times using a comparison with the Baker et al. (2011) sample selection method (page 20737, line 6).
4. Working with and referencing detailed CARIBIC monsoon studies as well as independent monsoon studies that show isolation within the monsoon anticyclone (page 20737. line 3).
5. Our use of the bootstrapping technique.

Page 20737:

1) L2-6: Many of the arguments in this paper refer the reader to other papers; the assumptions need to be quantified here. How much is 'minimal'?

We have added a description of how Baker et al. (2011) judged mixing to be minimal. We have added a reference to the Randel & Park (2006) study which provides quantitative analysis of the isolation within the ASM.

2) L10-15: A general comment is to try to rearrange the flow of the paper. Many times a result is presented but not supported until later. Try to bring common ideas together. This will also help condense the text and avoid repetition.

Restructuring has been part of many of our responses above. At points where it is best to mention an idea but refer to it fully later we have signposted the relevant section, figure or table.

3) L19: State the lifetime of CO. Why does CO with a 2-month lifetime experience changes in 4 days but not CH_2Cl_2 with a 5 month lifetime?

We have given the lifetime (mid latitude summer) as 2 weeks (Scharffe et al., 2012 referencing Warneck, 1988) which would suggest changes have occurred. Scheeren et al. (2002) gave a CO lifetime in the tropical marine boundary layer of 19 days.

4) L23: (1) The emissions occur at the surface and are measured at 10-12 km. Is 298 K appropriate? (2) By 'average concentration of OH' do you mean throughout the troposphere?

(1) We have used 298 K to standardise with Scheeren et al. (2002) and Baker et al. (2011), the two papers we discuss and compare our results to most often.
(2) We have explained this in further detail.

Page 20738:

1) L5: State the magnitude of the two errors so the reader can see how much smaller the first is compared to the second.

The errors associated with the measurements of CH_2Cl_2 and CO were given in Section 2. We have added a reminder of this page 20738.

2) L26: What is the basis for assuming CO emissions are constant year-round?

We have restructured and clarified this section.

Page 20739:

1) L3: 'hopefully' – not sure what is meant by this.

Replaced with 'likely'

2) L6: It's difficult to follow all the assumptions in this paper. Some seem unsubstantiated and it's difficult to know which assumptions are negligible and which are assumed to be negligible but it's not really known.

Addressed as part of responses to other comments.

3) L8: As noted above this correlation could be presented and discussed earlier.

See our response to: #Page 20735/point 9

4) L15: Remove the semi-colon.

Done.

5) L18-20: 'the rest of the ratios show a consistent increase over time due to increased emissions of CH_2Cl_2'. This conclusion seems to come out of the blue. What evidence is there that there is a quantitative increase and that it's related to increased CH_2Cl_2 emissions rather than variability? Are you just talking about the urban ratios or also the biomass burning ratio of Rudolph et al.? Even if it were possible to draw such a conclusion, the 2006 ratio from Mexico City for example is lower than the 2005 ratio from Los Angeles.

As this section was restructured and moved earlier in the manuscript the part described above has been removed.

6) L20:Which ratio is 'this ratio'?

We have clarified this point.

7) L22: What about seasonality in CO emissions?

We addressed the seasonality of CO emission on page 30738 line 16 onward where we combined EDGAR and GFED CO data.

8) L28:I'm not convinced that all the uncertainties have been addressed well enough to draw this conclusion.

Hence our use of 'roughly' and a reminder that there are caveats.

Page 20740:

1) L8: What is the uncertainty on these estimates?

We described the uncertainties associated with the CO monthly estimates and our treatment of them on page 20739 line 2.

2) L10: Or emissions of CO could decrease with time because of catalytic converter technology. What is the situation in India?

This section was merely designed to provide additional support to the EDGAR measurements (again, reducing our uncertainties). To go into this in too much detail would detract from our overall message.

3) L22: Change '200' to '2000'.

Done.

4) L23: It isn't clear to me that uncertainty has been handled well enough to provide confidence in these results.

We hope our answers to the above points have now clarified how we have addressed the uncertainties and also our general improvement of uncertainties with respect to increases over time, etc.

5) L26: Does 'North and central Atlantic' correspond to 'C. America' in Figure 1 (green)? If so this should be clarified.

Done.

Page 20741:

1) L4-9: The text is vague. Quantify 'very little variation', 'increases over time', 'less pronounced' and 'slight decrease'. Are the differences statistically significant? As before, was annual averaging based on a bulk average of all samples, which will bias the average towards heavier sampling times, or was some kind of seasonal weighting performed?

We have restructured this section; providing additional quantitation where appropriate and stating where our sample size prevents us from doing more than providing an indication of the situation. This section does not include annual averages.

2) L10-11: The long-term trend in Figure 6c does not look statistically significant.

We made no claim that the increase was significant but we have now clarified this point.
3) L13-14: As with other regions a better uncertainty analysis needs to be provided. Is it realistic that a long-term trend can be assessed to within 0.7 ppt from this data set?
Altered as part of our response to #Page 20741/point 2.

4) L17: What time of year did Scheeren et al. make their measurements? Is it an annual average like the CARIBIC data or over a shorter time period (in which case the comparison is not meaningful without a discussion of seasonality)? Also 'Similarly low' does not seem like a correct interpretation of 16 versus 23.
(1) March 1998. However, these measurements were in the tropics so seasonality is not as important as at mid latitudes. We used 'similarly low' to compare both 16 and 23 ppt with the high levels seen over, e.g. India (following the comparison on line 7). We have restructured and clarified this section.

5) L18: Why is the Scheeren discussion paper cited rather than a final published version?
The paper did not make it past the discussion paper. We checked the reviewer comments and this paper did not make it out of discussion due to their further interpretation of the data not the data themselves, leading us to believe the data are valid and OK to cite in our manuscript.

6) L20-21: Similar to elsewhere in the paper, before the data can be used to look at changes in CH_2Cl_2 with time there needs to be a full analysis of other impacting factors such as variability, seasonality, sample size, etc.
See our responses to other sections and the updated manuscript with the new Table 3 and improved discussions.

Page 20742:
1) L2-3: Where can the reader see that the samples were evenly distributed across many months? What about elsewhere in this paper where samples were not evenly distributed over time, but long-term trend analysis was still performed?
See our new Table 3.

2) L3-4: The statement that 'it is unlikely that seasonal bias plays a role' needs to be substantiated : : : in other words time series need to be shown for all regions, not just Europe.
See our previous responses.

3) L6-8: This type of argument applies everywhere in the paper. A few low or high samples will affect each average and the sample size is too small for long-term trend analysis, especially when seasonality hasn't been factored in and the averaging seems to have been done in bulk.
See our previous responses

4) L22: Figure 8 is very hard to read.
See our response to #Page 20730/point 5.

5) L22: (1) Not sure it's a 'clear increase' : : : the error bars are so large that none of the differences look significant. The maximum increases for 2007-2008, decreases for 2009-2010, and increases for 2011-2012, so even if the differences were statistically significant this could just be interannual variability rather than a long-term increase. (2) Also it would help to have all five graphs on the same scale for CH_2Cl_2.
The sentence referred to here states: "A clear increase in the **magnitude of high CH_2Cl_2 "pollution" events** in recent years can be seen. We are not referring to an increase in average values. The error bars are actually the range, as described on line 24 and again in the Fig. 8 caption. The maximum values have clearly increased from panel a to panel d and the lower values in 2009-2010 are discussed in more detail in Section 3.4. We have removed "in recent years" from the above sentence to clarify that we mean an increased over the entire time period.
(2) Done.

6) L27-28: Again avoid 'clear increase'. If you omit 2011-2012 instead of 2009-2010 then it would be a decrease.
We have removed the words 'clear increase' and replaced them with a discussion of the combined 2009-2012 dataset (including the low 2009-2010 values) which still show a statistically significant increase for the region discussed.

Page 20743:
1) L2: (1) In what season did Schauffler et al. make their measurements? (2) How does seasonality affect CH_2Cl_2 measurements at 15.3-17.2 km, and how might this affect the comparison?
(1) Schauffler measurements were made in 1991-1992, this information was/is provided in Table 2.
(2) We showed that seasonality was reduced at altitude in Section 3.1. We are also discussion tropical measurements were seasonality in tropospheric CH_2Cl_2 is likely to be of less importance than at mid latitudes.

2) L2-8: It's unlikely that the Schauffler average is 'lower' than the early CARIBIC value because the range shown in Figure 8a is so large. What is 1-sigma error on the CARIBIC value of 18.1 (13.0-25.0; n = 20) for 1998-2002? Is 18.1 significantly different from 14.9 +/- 1.1? Variability and small sample size are two other considerations that also need to be discussed.
The σ is 3.0, this is just discussing a comparison within the stated altitude band, so the range is not that large (especially at the lower end, as discussed, the variation tends to be in the high concentration events). Sample size for both is provided in the text. We do not make claims as to the significance of a difference between the two datasets, just that the Schauffler measurements do appear to be lower, something that is discussed in the Schauffler paper and which we reference on line 6.

3) L19-20: This statement is speculative. When did the rise of industrial activity begin in Asia? The CARIBIC data set spans 1998-2012 and the very high values of Barletta et al. were already observed in 2001, so it's unclear why a flight in 2012 is highlighted and linked to a rise in industrial activity.
This is not a central section of the paper; we are merely trying to explain a few measurements by providing ideas (supporting your earlier comments that conclusions should not be drawn from small sample sizes) that could prompt future work. We do not 'highlight' a flight in 2012 – this is the **only** flight to South East Asia made by CARIBIC between 1998-2012. So emissions may have risen earlier but we do not have the data to discuss this.

4) L22: Add an error bar to the value of 226 ppt.
This was in Table 2 but has also been added to the main body of text.

Page 20744:
1) L4: Avoid using 'about' with a number as precise as 95.9%. Are the NH emissions of HFC-32 really known to this level of precision?
Updated.

2) L8-9: Is it a rate of increase or just an increase? You could also just use 'growth'.
Changed to growth.

3) L16: Use a semicolon instead of a comma.
Done.

4) L16-17: Why is India in particular highlighted here? What are the world's first and second largest consumers of CH_2Cl_2? Are they experiencing rapid growth?
India is highlighted here because we made measurements over India, conducted emission estimates for India and saw a large increase that we wish to discuss/explain (see lines 18-20). We explain on page 20745 that India is still not the largest contributor to CH_2Cl_2 emissions, but its emissions are something we need to discuss based on our focus on this area in Section 3.3.

5) L21-22: Change 'the O'Doherty et al. (2014) study' to 'O'Doherty et al. (2014)'.
Done.

6) L22: The inter-hemispheric gradient of which compound? All HFCs or just HFC-32? Suggest condensing and clarifying the text in L21-25.
Done.

Page 20745:
1) L3-6: It is difficult to know whether large point source emissions translate into a major emission source at a national level. Does the Majumdar and Srivastava study discuss this?
We are clear in this paragraph that what we providing suggestions with respect to CH_2Cl_2 sources. Majumdar and Srivastava (2012) use their measurements top calculate an 'estimation of CO_2 equivalent of VOC emission from landfill' so they believe it is worth considering the importance of these emissions on a national scale.

2) L9-11: While this could be the case, the arguments leading to this conclusion seem speculative. Also the following sentences in this paragraph seem to contradict the importance of India on a global scale : : : if it's 4% then it's not an important source. Try to draw out a clear conclusion in this paragraph.
We have rephrased/restructured this paragraph.

3) L14: What year does the 515 Gg/yr apply to? Is it closer to 2008 or 2005?
1999-2003, so closer to 2005. We have clarified this point as part of rephrasing/restricting this paragraph.

4) L20: The paper and its conclusions need to be reworked within the limitations of what the CARIBIC data set can provide.
This has been done to reflect all the changes we have in response to earlier comments.

Page 20746:
1) L2: 'must be linked' is too strong without direct evidence.
Changed to 'most likely'.

2) L11: The minor contribution to ozone depletion should be mentioned in the introduction.
Done for #Page 20722/point 2.

Figure captions:
The captions would be easier to read if they had full sentences. For example try 'Error bars are given for combined values based on multiple measurements' instead of 'Error bars given for combined value based on multiple measurements'.
All figure captions have been updated.

Response to reviewer 2

General comments:

1. (1) "Introduction" needs to be more organized to focus on the issues authors want to address in this manuscript; physico-chemical properties of CH2Cl2, its environmental significance, overviews of long-term trend and NH-SH gradient based on the previous records, and recent changes inferred from ground measurements in the trend would be stated before introducing what additions could be made to recent findings by flight observations like CARIBIC campaign. Then authors specify questions that they deal with based on their data sets. (2) It would also be helpful if brief description about the following sections or guide on the rest of the manuscript were given at the end of the "Introduction".

(1) The introduction was restructured as part of our response to reviewer one - see #Page 20723/points 1-5.
(2) This has been added.

2. It will be easier to read if the "Methods" section is divided into following subsections: 2.1 Whole Air Sampling, 2.2 Halocarbon Measurement Systems (2.2.1 Instrumentation 2.2.2 Measurement Precision 2.2.3 Calibration), 2.3 Ozone and Carbon Monoxide, 2.4 Back Trajectories.

We have added some sub headings to the methods section.

3. In "Results and Discussion", data from four regions of Europe, South Africa, India, and the North and Central Atlantic were selected for each subsection. Explanatory titles of the subsections can be given instead of the regional names to describe different issues of each regional data set.
Done e.g. Section 3.1 changed from '3.1 Europe' to '3.1 Long-term time series of CH_2Cl_2 measured over Europe'.

4. One major point I would like to see discussed more quantitatively in this manuscript is statistical significance of the temporal increase in the European time series and of latitudinal gradients in the flights to South Africa. Since the data intervals were not even with time and latitude, temporal and spatial binning should be examined very carefully. In addition, comparisons with the previous measurements should be made considering not only complexity of CARIBIC flight tracks, but also seasonality, horizontal and vertical gradients of CH_2Cl_2 and its year-to-year variations in atmospheric trend.
We have addressed this as part of our response to reviewer 1. See, in particular, the following responses where we clarified and improved our discussion of average values, how we account for temporal and spatial variation and how we calculate increases based on the dataset we have available.

- #Page 20730/points 4 and 10
- #Page 20731/points 1, 2 and 5

See also:

- #Page 20730/points 7 & 8 and #Page 20731/points 3– improved discussion of the comparison between CARIBIC and Mace Head data.
- #Page 20730/point 9 – improved discussion of seasonality

5. Another issue is regarding more quantitative justification of air masses from the Indian subcontinent to ensure convective isolation from the southeast Asian region during the Asian Monsoon and thus the annual emission estimates from the Indian region.
We have addressed this as part of our response to reviewer 1. See our responses covering pages 20735-20740, in particular, the following responses:

- #Page 20735/point 3 – a discussion of sample sizes and the bootstrapping procedure we applied to our dataset
- #Page 20735/point 5, #Page 20736/points 2 & 3 and #Page 20737/points 1 & 3 – further justification for the emission estimate method used and further analysis of potential errors
- #Page 20735/points 6, 7 & 9 and #Page 20736/point 1 and #Page 20739/point 7– further justification for our choice of CO as a combustion tracer

6. One of main conclusions in this manuscript was that recent increase in the atmospheric concentration of CH_2Cl_2, which was discussed from CARIBIC flights over the European and Indian regions, "must be linked to" increasing use of CH_2Cl_2 as a feedstock for the HFC-32 production. However, this conclusion should be stated with further investigations, since a recent study by Li et al. (EST, 2014) demonstrated that in China, the world's largest consumer of CH_2Cl_2, it had been used mainly as solvent in chemical manufacturing industry, and interestingly not shared the same emission source with HFC-32.
We have updated this sentence, as part of our response to reviewer 1, to read: "The increase is most likely a result of increasing industrial use of CH_2Cl_2…". The Li et al. (2014) paper identified air masses influenced by solvent emissions by their high CH_2Cl_2 concentration and did not investigate solvent use in China directly. Wemainly concentrate on increasing emissions from India, as this is the region for which we have data, and provide a range of possible emission sources for this region based on existing literature. We mention the potential importance of China, and the importance of further studies in regions we cannot provide data for, at several points throughout our manuscript.

Specific comments:
1. P20723, LN 2-3: References are needed.
Done – see our response to reviewer one #Page 20723/point 1.

2. P20724, LN 9-10: Significance of contribution of VSLS at 10-12 km to the stratospheric chlorine amounts depends on the latitude, and thus this sentence seems to be valid only in the TTL.
This sentence has been removed as part of our response to reviewer 1 #Page20722/point 2.

3. P20724, LN 10: (1) "Please provide references for the sentence saying "$CHCl_3$ is of predominantly natural origin". (2) "Also consider other study, done by Aucott et al. (JGR,1999), arguing that at least 50% of $CHCl_3$ emitted in the NH were originated from anthropogenic sources.
(1) This sentence has been removed as part of our response to reviewer 1 #Page20722/point 2.
(2) We have included a more recent reference (Worton et al., 2006) which provides an estimate of the % of CHCl3 from industrial origin.

4. P20728, LN 25-27: References are needed.
This section has been restructured – see reviewer 1 comment #Page20728/point 5. References are included.

5. P20728, LN 28-29: To exclude the stratospherically-influenced air masses more accurately, it would be better to examine O3, a stratospheric tracer in combination with CO data as a tropospheric tracer.
We use the method in Zahn & Brenninkmeijer (2003) for calculating an O_3 tropopause. Their paper uses O_3 and CO. It is beyond the scope of our manuscript to discuss this, but we do cite this paper for readers who wish to learn more.

6. P20730, LN 11: Back trajectories of the air masses in discussion can be provided to illustrate their geographical sources.
On page 20729 line 13 we provide the web address where back trajectory plots can be viewed by interested parties.

7. P20730, LN 28: Isn't there any discernible latitudinal gradient from Barrow to Mace Head?
See our response to reviewer 1 #Page20730/point 11.

8. P20731, LN 19: Uncertainty of the averages over Canada and Greenland should be given for comparison with the mean in 2008 of the CARIBIC European data.
Done as part of our response to reviewer 1 #Page20731/point 16.

9. P20731, LN 25: Please add the reason why 30N was selected as a latitudinal boundary. It seems a bit arbitrary. For the consistency with the rest of the manuscript defining the tropical band between 25S to 25N, 25N would be a better boundary.
We have added justification for this selection. In this section we are not discussion tropical samples specifically (as we are later when we use the 25 °S – 25 °N boundary) but comparing samples taken over Africa to those taken over Europe (within the European box defined in Fig. 1 and Section 2). 30 °N provides a northerly delineation between samples taken over Africa and those taken within our European region.

10. P20732, LN 1-3: References are needed. Otherwise, the argument that observed increase in latitudinal gradient implies increase in NH industrial activities associated with CH_2Cl seems less convincing.
Our introduction discusses this, see page 20723.

11. P20732, LN 20-24: Please state the sampling seasons to show the data from the CARIBIC flight tracks to South Africa covered possible biomass burning periods.
Data seasonality, including for the African flight dataset, was covered extensively as part of our response to reviewer 1, see our responses above and our new Table 3.

12. P20733, LN 11: Y-axis scales of Figures 4(a)–(e) should be all identical for comparison.
Our discussion of Fig. 4 involves two factors (1) the increase of CH_2Cl_2 over time and (2) the different latitudinal distribution seen during the monsoon and non-monsoon months for both 1998-2001 and 2008. Whilst the discussion of (1), that CH_2Cl_2 has increased over time, would benefit somewhat from the Y-axis scales being identical the discussion of latitudinal **patterns**, which are an important part of our subsequent emission estimates, is benefited by the use of individual scales. As the increase over time (1) is also shown in Table 2 we decided to focus on (2) in Fig. 4 and so the different Y-axis scales are important aids in the understanding and discussion of this figure.

13. P20734, LN 20-24: Please add the references for these statements. Otherwise, more quantitative analysis should be made for further discussion.
Done.

14. P20735, LN 1: 'data us' should be replaced with 'use'
Done.

15. P20735, LN 4-6: how can we justify that 35 samples < 40N were originated only from the Indian subcontinent, not from the Southeastern Asia? Is it possible to investigate the back trajectories for the 35 samples?
The references we cited in this section provide further justification of this method, including an analysis of back trajectories. We have clarified this point.

16. P20736, LN 15-17: Please compare a CH_2Cl_2-CO ratio for the air masses influenced predominantly by the Southeast Asian region to rule out transport effect.
As mentioned in our response directly above (point 15) samples were selected based on back trajectory analysis and are only originating from the Indian region within the monsoon anticyclone. The samples we mention on page 20733 lines 17-21, that originate from southeast Asia were sampled during the non-monsoon months so would not make a fair comparison of transport times. They are also few in number and so not suitable for calculating an accurate ratio. We provide evidence throughout Section 3.3 that we took care to identify that transport and dilution were small (via several methods) and have also improved our discussion of this section in response to reviewer 1.

17. P20737, LN 15-20: These sentences are not properly stated. Age-correction for emission ratio is needed due to different loss rates between CH_2Cl_2 and CO (i.e., difference in chemical lifetime between CH_2Cl_2 and CO), which can cause apparent change in their emission ratio after transit for a certain time interval.
These lines state exactly what you have stated above. We include that we age correct due to different loss rates due to the different chemical lifetime and that we age correct our emission ratios to account for this over a 4 day window.

18. P20738, LN 1-5: Please cite the references.
We are uncertain about what references we could add here as we are discussing statistical errors. The paragraph read:
"Also associated with the emission ratio are errors arising during the calculation of the $\Delta CH_2Cl_2/\Delta CO$ slope. These errors arise from two sources: (1) uncertainties in the analytical measurements of both CH_2Cl_2 and CO and (2) uncertainties associated with using a slope calculated from a discrete set of samples to calculate a regional emission estimate. The errors associated with (1) are small compared to those associated with (2) and so we use (2) to set bounds on our emission estimates."

If the reviewer refers to the errors associated with (1), the analytical measurements, these were provided in Section 2.2, we have added a reference to Section 2.2 to this paragraph.

19. P20740, LN 12: Please specify the three previous studies.
We do in the following sentences, this is merely introducing them.

20. P20741, LN 9: 'that' should be 'than'.
Agreed, however this sentence has already changed as part of our response to reviewer 1.

21. P20741, LN 23-26: References should be given.
We provide two references on line 23 and these cover the information discussed in lines 23-26. This section has now been moved and assimilated into the methods section and no longer exists in this form.

22. P20742, LN 22: X-axis of Figures 8(a)-(e) should be identical.
Done as part of our response to reviewer 1 #Page20742/point 5.

23. P20743, LN 19-21: References should be given.
We provide a reference to Barletta et al. (2006). The sentence before this is introducing this reference.

References
All references are given in the Discussions manuscript unless cited below.

Hall et al. (2014) Results from the International Halocarbons in Air Comparison Experiment (IHALACE), Atmos. Meas. Tech., 7, 469–490, 2014, doi:10.5194/amt-7-469-2014.

Laube et al. (2008) Contribution of very short-lived organic substances to stratospheric chlorine and bromine in the tropics – a case study. Atmospheric Chemistry and Physics, 8(3) 8491-8518, doi:10.5194/acpd-8-8491-2008.

Randel & Park (2006) Deep convective influence on the Asian summer monsoon anticyclone and associated tracer variability observed with Atmospheric Infrared Sounder (AIRS). Journal of Geophysical Research, 111(D12), D12314, doi:10.1029/2005JD006490.

Warneck, P. (1988) Chemistry of the Natural Atmosphere. Academic Press, New York.