We would firstly like to thank the reviewer for their helpful comments. We have taken the reviewers comments into consideration and revised the manuscript accordingly. All the changes have been highlighted in the revised manuscript and are summerised as follows.

Reviewer's comments for the paper (acp-2017-244), entitled: "Atmospheric bromoform at Cape Point, South Africa, a first time series on the African continent" by Kuyper et al., submitted to ACP.
Recommendation: Major revision

General comments

> *This paper presents some in-situ atmospheric measurements (~130 samples) of bromoform ($CHBr_3$) from Cape Point, South Africa over a 2 month period in 2011. These are the first reported measurement of this important halocarbon from the southern African continent (although some of the data has already been discussed in Kuyper et al. 2012) so do provide a useful, if limited, new dataset. The concentrations observed are, on occasion, at the higher end of those previously reported. I have a number of serious reservations about the measurements, the interpretation of the data and the conclusions which prevent me from recommending the paper for acceptance in its present form. I fully understand that sometimes it is difficult to draw firm conclusions from a limited set of measurements, but in this case I believe the authors are rather over-interpreting their data.*

Response: We believe that the comments and recommendations from the reviewer will greatly improve the quality and substance of this paper. The reviewer raises some concerns over the validity of the data and the conclusions. It is hoped that through these discussions / responses we can alleviate the reviewer's concerns. As the reviewer highlights it is difficult to draw firm conclusions from an inceptive dataset and limited dataset, which were on occasion at the higher end of those previously reported. In trying to bring our data to life, we have perhaps strayed into overinterpreting the data and we have tried to take on board the reviewers helpful suggestions to remedy this. As a result of the reviewer's comments, the entire calibration method was extensively re-examined and subsequently, an error identified. This has had a material impact on the data and this is reflected in the revised manuscript. Specifically, the error resulted in a systematic overestimation of the observed bromoform mixing ratios by a factor of 2. Finally, the results and discussion have thus been simplified and updated as per the reviewer's suggestions.

1. *The instrumental methods are not described particularly well. The authors refer to a previous publication which does have a lot more detail, but there are some outstanding questions related to the identification of bromoform (CHBr3) and the exclusion of possible co-eluting species. The electron capture detector (ECD) is not particularly specific so is subject to potential interferences in different types of air mass, particularly when trapping such large volumes of air or when sampling in more polluted environments. Have potential co-elutions been thoroughly tested for and ruled out?*

Response: The reviewer is correct that the ECD is not a an absolute detection method and this is a limitation of the technique. The ECD (in combination with an analyte -specific chosen chromatographic column) is however, highly specific to polar compounds and well suited for halocarbon measurements, as has been extensively demonstrated in the literature extending back to the early 1990s (e.g. Schall and Heumann, 1993). Moreover, analyte identification by using relative retention time is a reliable and well documented method found throughout the published literature (e.g. O'Brien et al., 2009; Poole, 2003; Wevill, 2005).
The literature is also unanimous and unambiguous in showing that, in unpolluted environmental air samples, bromoform can be separated with no co-elution problems (column dependant - see below). This is made clear in the EPA 8260B method, which discusses the separation of numerous halocarbons. There is some evidence that separating bromoform from

water and in highly polluted gaseous mixtures can be problematic (EPA 504.1). The co-elution problem described above is entirely column dependant; in the study presented here a DB-624 column was specifically selected for its published ability to separate bromoform without co-elution (Mattson et al., 2012; Happell & Wallace,1997; Itoh et al., 1997; Andrews et al., 2016). Furthermore, Cape Point predominantly receives “clean” (i.e. with little/no detectable pollutants) marine air from the Southern Ocean and is only occasionally intruded by anthropogenically modified air from Cape Town (as is discussed in the paper). Given this the chances of co-elution would appear to be negligible for the majority of the data presented, which was recorded in pollution free air. Where the air being measured shows significant anthropogenic influence the possibility of co-elution may increase (although given the discussion above would still be expected to be low) and the discussion has been amended to reflect this potential source of uncertainty in those result which are affected.

Please see below an overlay figure of a calibration standard and an air sample taken from Cape Point. If there were co-elution occurring we would expect that the peak shape would be non-Gaussian, typically displaying tailing of the co-eluting peak. As one can see from the figure the peak shape of the calibration and the air sample for bromoform are almost identical. This gives us great confidence that the system was able to suitably separate and detect bromoform in air samples.

Remedial action: The methods section has been revised in the updated manuscript to be more explicit and unambiguous on all points raised. This includes the separation of samples as well as the calibration of GC-ECD system.

Following the reviewer’s comments on co-elution, as a possible mechanism to explain the observed elevated mixing ratios, an exhaustive examination of our calibration method was performed. It was found that the trapped calibrant volume had not been correctly calculated. The permeation tube emitted pure bromoform at 373 ng min^{-1} and this was diluted in nitrogen at 100 ml min^{-1}. The calibrant gas was loaded onto the trap for 30 s per sample loop. Our calculations had assumed a full minute of calibrant was loaded on the trap. Effectively a mass of 0.1865 ng of bromoform was loaded on the trap instead of the 0.373 ng previously calculated. As described in the text the number of moles of bromoform loaded in a calibration sample was calculated. This was then converted through the air number density and the number of molecules trapped to an atmospheric mixing ratio.

The net effect of the correction of the error effectively resulted in a halving of the reported bromoform mixing ratios. We apologise for this error. The data has been revised as per the new calibration parameters and presented below.

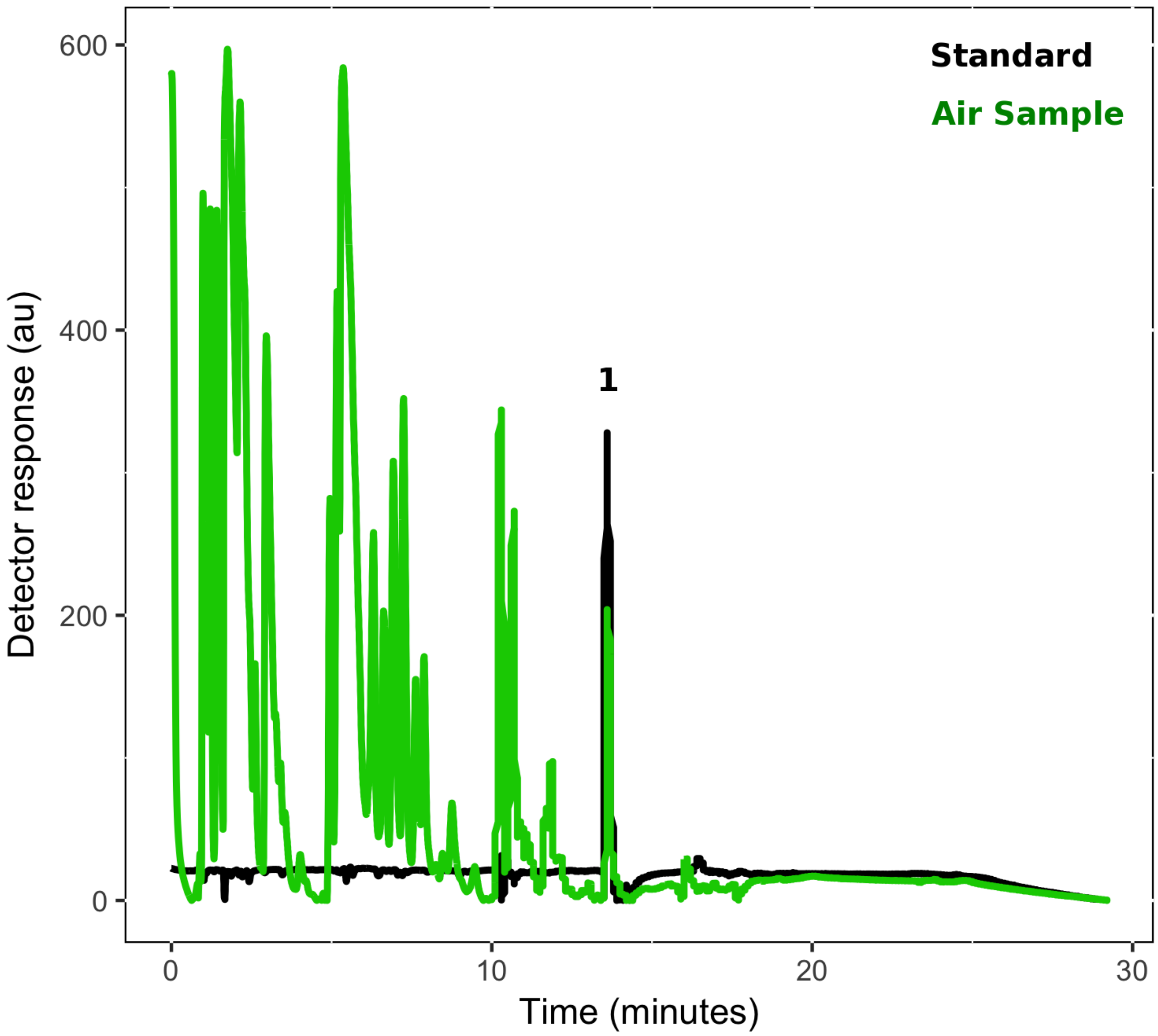

Figure: Overlay of air sample and calibration standard. Bromoform peak indicated by the number 1. The calibration standard reflects the injection of 2 loops of bromoform standard (23.6 ppt) from the permeation oven trapped on the adsorbent trap.

As this is not intended to be a methods paper and given the method was previously published elsewhere, we do not feel that publishing this graph in ACP adds enough to the work here to justify its inclusion. However, should the reviewer/editors deem it worthy of inclusion we would be happy to edit it and include for publication.

2. *Much of the discussion is highly speculative and potentially wrong. For example the conclusions on (a) anthropogenic sources of $CHBr_3$, (b) ozone stimulation of $CHBr_3$ release from seaweeds, and (c) a $CHBr_3$ source from the upwelling region, are all highly dubious. The authors present very little firm evidence to support these theories and, to some extent, they overlook a more likely, or simpler, explanation for the high levels of $CHBr_3$ at the site. They state that Cape Point and the surrounding coast-lines for many kilometres to the north and west (and south?) support large, extensive seaweed beds. If this is true then surely the most likely origin of the high $CHBr_3$ they observed is simply local (and regional) seaweed? The flux to the atmosphere would then be highly dependent on local tidal patterns, with large concentrations to be expected when the kelp becomes exposed to the air. Although this is mentioned briefly in one case study, which suggests that this process may be occurring further to the north, can the authors confirm that the same phenomena is not occurring closer to the*

site on a daily basis? Here a detailed description of the local /regional seaweed populations would be very helpful, ideally identified on a map of the area (Figure 1?). The authors need to convince the reader that the high levels of CHBr3 are not simply from very local sources.

There needs to be a wider discussion of the tidal phenomenon as it is very likely to be the reason for the high concentrations and some, if not all, of the spatial/temporal variation. Similar, and more extensive, studies have been carried out over different seaweed areas such as Mace Head and NW France so references to these should also be provided.

Response: The reviewer raises valid concerns over the discussion and the various attribution of bromoform sources. We would like to preface our response by addressing one important misconception. The three statements A-C the reviewer refers to as *conclusions* were not intended to be interpreted as conclusions. The purpose of A-C is to serve as *test hypotheses* used to frame the discussion of our results. These were chosen as a result of (i) a careful review of the literature, to identify the sources of bromoform in past studies, and then (ii) considering this review in the context of our specific knowledge of the local environment / climate and geography, to arrive at the stated test hypothesis. We are more than aware that this first tentative data recorded at the Cape Point site does not, and could not, falsify any of the test hypothesis in and of itself. We intended to use the hypothesis as a tool to frame the discussions as to which of these potential sources of bromoform this initial data is most consistent with, with a view to guiding future research. We apologise for any ambiguity here and will aim to make this clearer.

The reviewer suggests that the local kelp may explain the measurements observed at Cape Point and that we may have omitted this possibility from our discussion. This is in fact included in the manuscript, as a it effectively a restatement of hypothesis B above. We will aim to make this clearer in the text. To reiterate what we noted above, the result do not however clearly falsify any of A-C.

The reviewer highlights that there are examples in the literature where an increase in measurement signal is associated with low tides, specifically at Mace Head (Carpenter and Liss 2000). This literature argues that this is from the fact that at low tide the kelp is exposed (entirely or partially) to the atmosphere. While the tidal range in the vertical at Cape Point is similar to that of Mace Head (1 – 2 m), the horizontal extent of its range is vastly different. The horizontal change in tide at Cape Point is ~10 m, while at Mace Head the horizontal extent can be many 10s to 100 m (D. Shallcross, pers. comm.). Therefore, at Cape Point the kelp remain submerged throughout the tidal cycle. Thus, as the local kelp remain submerged we do not expect a large tidal signal at the Cape Point site. Work done for Kuyper (2014) showed that the bulk tide and bromoform measurements did not correlate.

In terms of the distribution and species composition of seaweed, the work of Anderson et al. (2007) divided the area in shown in the location map (Figure 1) into 19 areas and used a variety of remote sensing technique to assess the extent of kelp beds in each of those areas. The result show that kelp beds are present in all 19 areas ranging from a minimum of 11 ha coverage in Table Bay to a maximum of just under 1000 ha towards Port Nolloth. The species composition was predominantly Ecklonia maxima Papenfuss south of Yzerfontein but transition to predominantly Laminaria pallid Greville north of Yzerfontein.

Furthermore, whereas we are not excluding the local sources by any means there are arguments to support a (additional) source further afield. The bromoform signal as a function of wind speed and direction as displayed in the polar plot (Fig. 8.) suggests two sources of bromoform, one to the north and a second to the west. Although interpretation of these figures can be problematic, the figure indicates that the dominant sources are at higher wind speed thresholds and consequently from farther afield. Concurrent measurements of ^{222}Rn and CO at Cape Point were used in an attempt to isolate sources. Some of the cases the air mass can be traced to areas influenced by anthropogenic activity, and other times not. If the wind is clean (no or very low ^{222}Rn) and a high wind speed is present then we can assume that the bromoform is not local.

Remedial action: The discussion as a whole, along with the results, have been revised to clarify our position and further explain the points raised by the reviewer. The time series plots have been revised and includes a plot of the tide and meteorological measurements as requested. See time series figures below and the sub-plot figures. From the subplot figures it appears that the tidal height may be a necessary, but not sufficient factor to explain the elevated bromoform mixing ratios. This is explored in the revised manuscript.

3. *Results section: The order seems wrong. Why not start the results section by discussing the CHBr3 time series before going into the chemical climatologies? I would further suggest that you show the radon and wind direction data on the same plot as Figure 7 as this would make it much easier to spot patterns, etc. Similarly, when discussing the 3 individual episodes it would be helpful to see the same Figure expanded for the periods of interest.*

   I found some of the diagrams rather difficult to interpret. In particular the various polar plots (Figs 5, 6, 8). These types of diagram can sometimes be a little over-complicated. A better explanation, if not a full rethink, is required. For example, in the case of Figure 5, what do the individual circles represent? Are they individual samples or averages in a particular sector? Why is the red circle to the NE not represented somewhere in Figure 6. I would expect to see a red circle, albeit closer to the origin, in Figure 6. Perhaps this is because the colour scaling in the 2 Figures is different?

   Why not show Figure 8 before Figs 5 and 6, perhaps after discussing the time series (see earlier comment).

   However, before using these polar plots the authors need to explain why they would be expecting to see correlations of CHBr3 with tracers like CO and ozone. Furthermore, the argument that ozone increasing from, say, 25 to 35 ppb represents a significant enhancement of ozone is contentious. Can you really label this as "enhanced ozone" and would you really expect such a small enhancement to have any significant effect on CHBr3 release from seaweeds? Please provide a reference to support this. Surely any ozone effect will be much smaller than the local tidal effect (which has not been discussed)?

   There is no discussion of any diurnal pattern in the data. Were there measurements at night? Can boundary layer height or temperature have an impact on the observed concentrations?

Response: The reviewer expressed concern over the order and in particular some of the figures found within the results section. After consideration we concur that some of the polar plots and their interpretation proved to be problematic. Whereas we thought these figures offered valuable information, we accept the reviwers criticism that interpretation of the figures is difficult and therefore it would be better to try and tell the story without them. Thus, these the majority of these figures have been removed. As noted we now have the tracer data plotting in times series alongside bromoform as the reviewer requested.
In terms of why CO is expected to be related in some way to bromoform, it is here being used as a potential proxy for anthropogenic emissions. Bromoform has some known anthropogenic sources such as water chlorination and nuclear power generation (Quack and Wallace, 2003). While, the anthropogenic sources are on the whole small in comparison to natural sources they can, however, be significant on a local or regional scale (Quack and Suess, 1999). Since CO is a known tracer of anthropogenic influence, measurement of elevated CO in an air mass

strongly suggests some type of anthropogenic mixing. A correlation of elevated CO and bromoform was expected, if the bromoform was anthropogenic in origin. The absence of a correlation being observed helps demonstrate that the bromoform measured here was not predominantly anthropogenic in origin.

In terms of the reasons for measuring ozone, the presence of ozone has been shown to elicit a bromoform release in species of Laminaria kelp (Palmer et al., 2005). Palmer et al. (2005) show that ozone can elicit a significant and rapid response from kelp when ozone is introduced. Although the kelp are covered for most of the tidal range, the tops of the kelp fronds at Cape Point are exposed. So the ozone measurements were intended to help investigate whether exposure of the fronds to sunlight and ozone would notably impact the bromoform observed. Work was done in Kuyper (2014) in which these multivariate comparisons were performed. The net result was that there were too few data to conclusively explore this. The analysis was thus simplified to examine these in isolation.

We thank the reviewer for bringing up the diurnal cycle as this was something of an omission and appears to provide some interesting insight.

Remedial action: The section has been rearranged and reordered to make the flow more logical as per the reviwers comments. Furthermore, most of the polar plots have been removed as they appear to be a source of confusion. Discussion about ‘enhanced ozone’ has been removed or revised. The reasons for investigating a correlation between bromoform and other tracers have been made clear. A discussion of the diurnal cycle and any possible impacts from temperature and marine boundary layer height have been included in the updated manuscript.

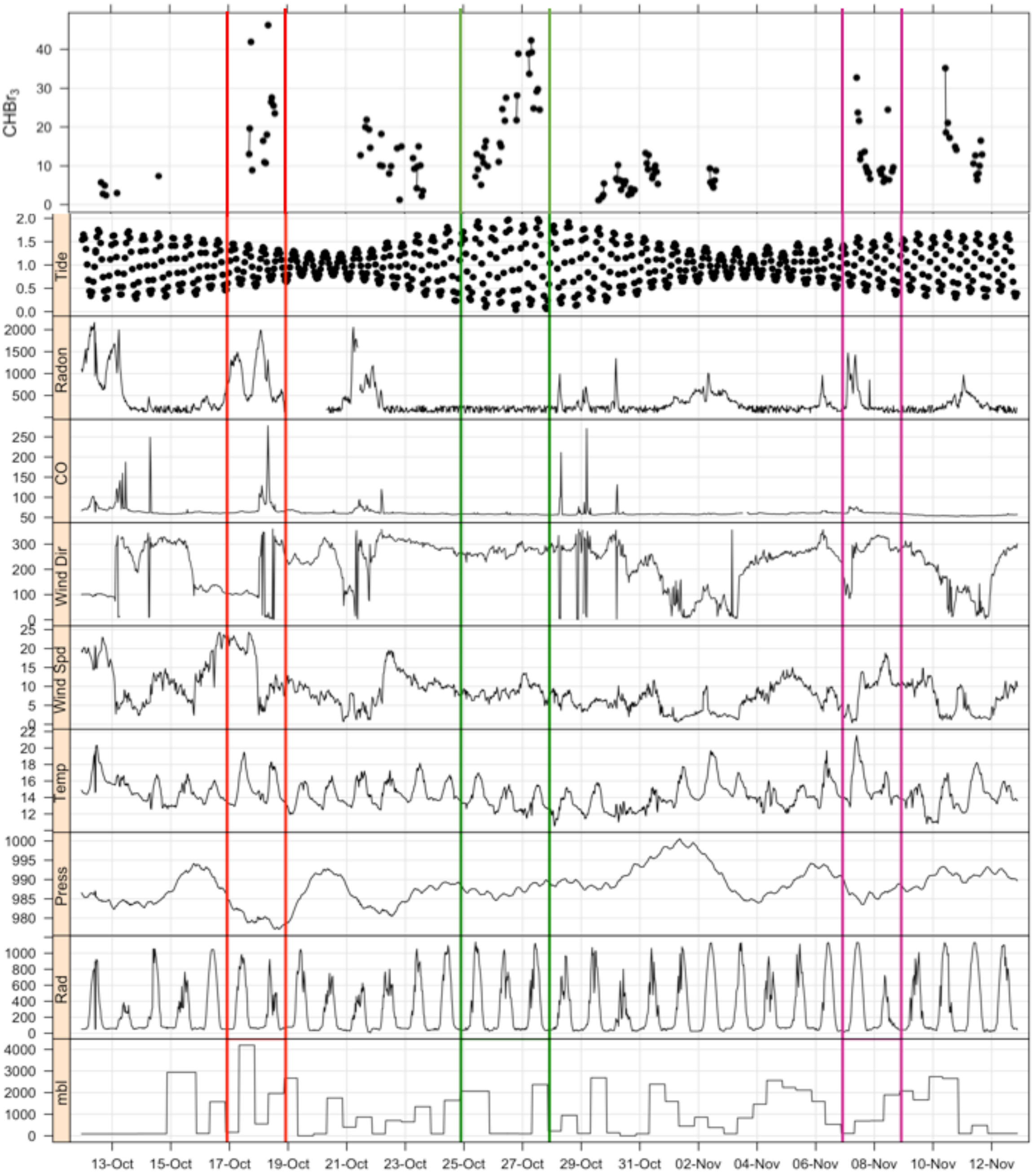

Figure: Time series plot of measurements at Cape Point during October / November 2011. Events are highlighted by the coloured lines: E1 = red, E2 = green, E3 = violet.

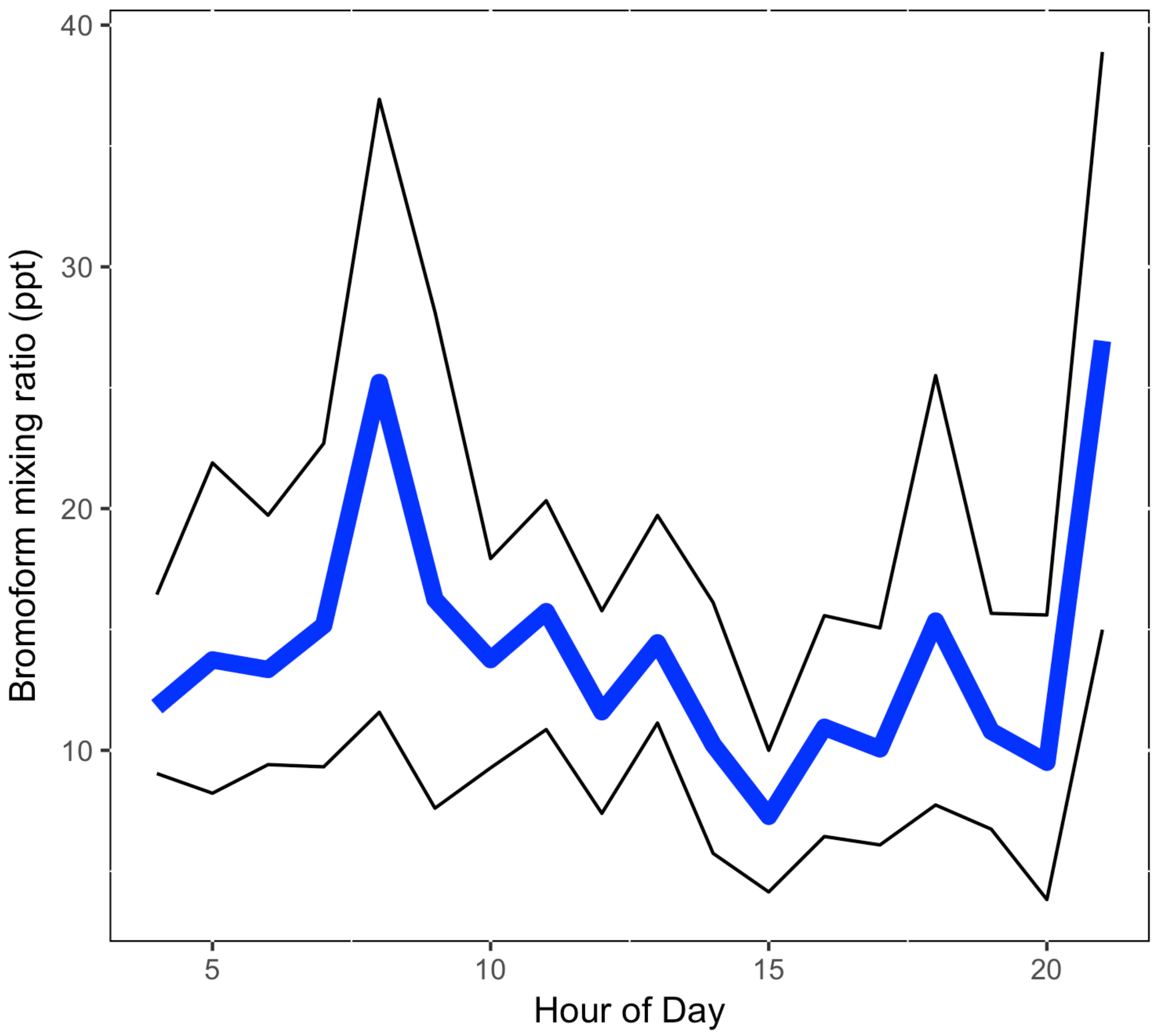

Figure: Mean diurnal cycle, calculated from all measurements binned by hour. The black lines above and below signify the 95 % confidence interval.

4. *The references to previous measurements (e.g. in Table 1) are not up-to-date. There have been a number of new studies in recent years that should be included (including a possible reference to the HALOCAT database).*

Response: The reviewer kindly noted that the Table of observations included in the paper is not up to date. We thank the reviewer for highlighting this oversight on our part.

Remedial action: The table has been updated to include some recent measurements and reference to the HALOCAT database. The updated measurements from Cape Point have been included in this table.

Table: Selected comparison measurements of bromoform in air samples above coastal, upwelling, open ocean and lower marine boundary layer regions.

			$CHBr_3$ (ppt)				
Location	Date	Latitude	Min	Max	Mean	Reference	Region

New Hampshire TF	Jun-Aug 2002-4	43.1 ºN	0.2	37.9	5.3-6.3	Zhou et al. 2008	Coastal
New Hampshire AI	Jun-Aug 2004	42.9 ºN	0.9	47.4	14.3	Zhou et al. 2008	Coastal
Hateruma Island, Island	Dec 2007 - Nov 2008	24 ºN	~0.5	7	0.91-1.28	Yokuchi et al 2017	Coastal
Mauritanian upwelling	Mar - Apr 2005	16-21 ºN	0.1	0.6	0.2	Quack et al. 2007	Upwelling
Cape Verde	May-Jun 2007	16.8 ºN	2.0	43.7	4.3-13.5	O'Brien et al. 2009	Coastal
R/V Sonne	July 2014	2-16 ºN	0.79	5.07	2.08	Fuhlbrugger et al. 2016a	Open ocean
R/A Falcon	July 2014	2-16 ºN	0.99	3.78	1.90	Fulbrugger et al. 2016a	MABL WASP
Atlantic Ocean	Oct - Nov 2002	10 ºN	0.5	27.2	-	Quack et al. 2004	Open ocean
SHIVA	Nov-Dec 2011	0-8 ºN	1.23	3.35	1.81	Sala et al. 2014	MABL WASP
Borneo	Apr-Jul 2008	4.70 ºN	2-5	~60	-	Pyle et al. 2011	Coastal
Strait of Malacca	Jun-Jul 2013	2-6 ºN	1.85	5.25	3.69	Mohd Nadzir et al. 2016	Coastal
Sulu-Sulawesi	Jun-Jul 2013	2-6 ºN	1.07	2.61	1.60	Mohd Nadzir et al. 2016	Coastal
Christmas Island	Jan 2003	1.98 ºN	1.1	31.4	5.6-23.8	Yokuchi et al. 2005	Coastal
San Cristobol Island	Feb - Mar 2002, 2003	0.92 ºS	4.2	43.6	14.2	Yokuchi et al. 2005	Coastal
Peruvian upwelling	Dec 2012	5-16 ºS	1.5	5.9	2.9	Fuhlbrugger et al. 2016b	Upwelling
Indian ocean	Jul-Aug 2014	2-30 ºS	0.68	2.97	1.2	Fiehn et al. 2017	Open ocean
Cape Point	**Oct - Nov 2011**	**34 ºS**	**1.10**	**46.2**	**13.2**	**This study (revised)**	**Coastal**
Cape Grim	2003	40.7 ºS	1.3	6.4	2.9	Yokuchi et al. 2005	Coastal

Coastal South America	Dec 2007 - Jan 2008	55 °S	1.8	11	7.4	Mattsson et al. 2013	Coastal
Antarctic coast	Dec 2007 - Jan 2008	65 °S	2.1	4.9	3.2	Mattsson et al. 2013	Coastal
Antarctic Ocean	Dec 2007 - Jan 2008	65-67 °S	1.9	3.9	2.3	Mattsson et al. 2013	Open ocean

Specific comments

> *P1, L2: why is the location "unique"?*

Response: Cape Point is unique in receiving clean air from the Southern Ocean and occasional influences from urban-anthropogenic sources.

Remedial action: The word unique has been removed.

> *P1, L13-14: the "sweet odour similar to chloroform" is irrelevant*

Remedial action: This has been removed.

> *P1, L14: what are these anthropogenic sources of bromoform? Please list with references. What fraction of global emissions are likely to be anthropogenic?*

Response: The likely anthropogenic sources in Cape Town are the nuclear power plant north of Cape Town and the numerous water treatment plants throughout the city that relies heavily on chlorination processes. From Quack and Wallace (2003) and Carpenter and Liss (2000), the estimated global budget of bromoform is total and divided into 1.6 (0.4–2.7) Gmol Br yr^{-1} for kelp, ~2 Gmol Br yr^{-1} phytoplankton and 0.346 Gmol Br yr^{-1} anthropogenic (Quack and Wallace, 2003).

Remedial action: These have been added to the text which now reads:

Bromoform, apart from the few anthropogenic sources including water chlorination, nuclear power and rice paddies, is naturally produced by kelp and phytoplankton in the upper layers of the ocean (Quack and Wallace, 2003). It is estimated that globally between 2.2×10^{11} – 2.5×10^{12} g $CHBr3$ yr^{-1}i is produced of which only 3.0×10^{10} g $CHBr3$ yr^{-1} is anthropogenic, the rest being from natural sources, including 1.3×10^{11} g $CHBr3$ yr^{-1} is from brown algae and 1.7×10^{11} g $CHBr3$ yr^{-1} from phytoplankton (Quack and Wallace, 2003, Carpenter and Liss, 2000).

> *P1, L16: "Outgassing to the atmosphere" sounds better than "Atmospheric outgassing" P2,*

Remedial action: This has been changed as requested.

> *L20: replace "within this region" with "in the tropics" P2, L22: same as above*

Remedial action: These have been replaced as requested.

> *P2, L22: What is meant by "discrete shipboard measurements"?*

Response: In this we intended to imply measurements that occur only when there are cruises which happen to pass through the region/ area of interest. They are not fixed point measurements and have not been made over long periods of time. I.e. they are highly irregular.

Remedial action: The sentence has been revised to read:

"Existing data in this region tend to be from transient ship cruises, which only provide a discrete snapshot at the point in space/time that the cruise transects the area of interest."

> *P2, L23-24: "No time series . . .. Like Cape Point.". The authors need to be careful with this sentence. Do they mean there are no time series in Africa, the tropics or globally? The latter 2 would both be wrong. Cape Point of course is also not in the tropics.*

Response: We thank the reviewer for highlighting this potential ambiguity.

Remedial action: The sentence has been revised to be more accurate. It now reads:

"No time series of measurements at a fixed point currently exists for a coastal site in southern Africa. Furthermore, the Cape Point monitoring station fills a critical Southern Hemisphere latitudinal gap between Cape Matatula, American Samoa (14 ºS) and Cape Grim, Tasmania (41 ºS; Brunke and Halliday, 1983)."

> *P2, L27-28: please explain why these gases might play a significant role in climate change.*

Response: Ozone in the upper troposphere is a potent greenhouse gas and bromine initiated destruction of ozone occurs in this region.

Remedial action: An explanation of the role of ozone in the upper troposphere and it greenhouse potential in this region have been added to in the text.

> *P2, L31: delete "to", i.e. "a unique location from which to measure. . .."*

Remedial action: This has been deleted.

> *P2, L33: what is meant by an "intermediate air sample"?*

Response: An intermediate air sample is a marine air sample that has become modified with continental air masses (as defined in Brunke et al., 2004.). This is indicated by the radon concentration; marine air typically has a radon concentration below 350 mBq m^{-3}, intermediate (or mixed) air samples between 1500 – 2000 mBq m^{-3} and continental air at above 2000 mBq m^{-3}. mixed air contains marine, continental and urban influences (Brunke et al., 2004).

Remedial action: The text has been updated to: mixed air as defined in Brunke et al. 2004.

" … ranging from marine sources (baseline), to continental and mixed air (baseline, continental and urban influences; Brunke et al 2004)."

> *P3, L1: Why might the subtropical location of CP make the region be an important source bromoform? Do you mean that if the region were a strong source of bromoform then this would be significant globally? I assume this is because it is relatively close to the tropics where convection could potentially transport it to the stratosphere? Please explain this sentence more clearly.*

Response: A bit of both. That this could be a large contribution to the global budget of bromoform. Although in all likelihood the contribution from the sub-tropics is going to be small, the significance of its proximity to the tropics and the deep convection cannot be overlooked.

Remedial action: The paragraph has been revised to redress this ambiguity. It now reads:

"The subtropical location of Cape Point may make this region a particularly significant source of bromoform to the atmosphere, specifically when considering potential impact on global ozone budgets; the region lies in close proximity to the tropics where deep convection is able to rapidly transport the outgassed bromoform into the UT/LS. It is here that bromine initiated catalytic ozone destruction occurs. Moreover, data recorded here is of particular value as the size of the contribution from Cape Point region is to date largely untested."

> *P3, L5: "biologically active" or similar is possibly better than "highly productive". Is the Southern Ocean active everywhere or just in certain regions? I assume the authors are referring to phytoplankton rather than macroalgae?*

Response: The Southern Ocean is well recognised as a high productivity low nutrient ecosystem and to a first approximation exhibits homogeneously high phytoplankton growth. There are of course fine scale various in time and space, but for the purposes of this work we suggest that the approximation of homogeneous high biological activity is sufficient. In this pelagic environment we are of course referring to microalgal productivity and not kelp.

Remedial action: The terms have been replaced as suggested by the reviewer. The text has been amended to remove any ambiguity.

> *P3, L11: The location of the local and regional kelp beds is highly relevant to the arguments used later in the paper. Is it possible to indicate on Figure 1 where the main kelp beds are?*

Remedial action: The discussion has been modified to expand on location and type of kelp found in area surrounding Cape Point and to the north up the coast. Please also see earlier comment.

> *P4, L5: define the term "GC-ECD"*

Remedial action: This has been done.

> *P4 L7: insert "the", i.e. "as per the method"*

Remedial action: This has been done.

> *P4, L12: What is meant by a "quasi-discrete sampling pattern"? "Quasi-continuous would be more appropriate, although you could simply say that 131 samples were collected during the period xxx to yyy.*

Response: Later the reviewer asks for the description of 'quasi-discrete' to be changed to 'quasi-continuous'.

Remedial action: We have followed the reviewers' suggestions here and later.

> *P4, L18: Please explain what the relevance of the sentence about gas viscosity is.*

Response: As the temperature of a gas increases so does its viscosity and so the flow rate decreases. In modern GC systems, an electronic pressure controller regulates the pressure at the head of the column to ensure a constant flow. The simplicity of the GC-ECD used here system meant that no adjustment could be made to account for changes in gas viscosity. This can have an effect on the separation of samples. Since this was consistent through all our samples this does not pose a problem, but the authors initially thought this fact should be noted for completion sake.

Remedial action: The sentence has been removed as it appears to be causing more confusion than clarity of understanding. A statement about the gas pressure being run at constant pressure has been added to the text as follows:

“Sampling

... A 30 ml min^{-1} nitrogen flow was added directly to the ECD in the form of make-up gas. Helium (Grade 5.0, Air Liquide) at a constant flow rate of 5 ml min^{-1} was maintained through the system. The oven was maintained at 35 ºC for 5 min following the injection of a sample. Thereafter, the temperature was increased to 60, 90, 150, and 200 ºC every 5 min. The temperature in the oven was increased at 65 ºC min^{-1} and held isothermally once the new temperature was reached.”

> *P4, L20: a ramp rate of 65 degrees per minute is very fast. Why was this necessary as it surely doesn’t help with peak separation?*

Response: The simple answer is that the GC model was not able to ramp at any other rate. A temperature could be programmed into the controller, resulting in a current being applied to the element and heating the oven. It was determined experimentally that this heating occurred at 65 ºC per minute. Once each ‘ramp’ or increase was complete, the oven was maintained at the new temperature for a period of 5 minutes, allowing for separation at that temperature to occur.

Remedial action: The paragraph has been rewritten to remove mention to a temperature ramp. The word ramp is a technical term that is not appropriate in this context. Please see the updated text above.

> *P4, section 2.3: there are a few details missing in this section which should be included. Was the air stream dried before pre-concentration? How did you measure the volume of air trapped? Did the system trap CO_2 and, if so, how did this affect the chromatography? An example chromatogram would also be helpful as well as some discussion on possible co-elutions (see earlier comment).*

Remedial action: The text in the methods section has been revised and expanded. As per the text sample volume was calculated based on sampling rate and time. A flow rate of 100 ml min^{-1} through the trap was measured on a digital flow meter. Since the trap adsorbents and desiccant remained constant so did the gas flow rate. This was checked on a weekly basis. A time of 15 minutes was used to collect the sample resulting in a 1.5 l sample volume.

The trap was flushed with helium (grade 5.0) before and after trapping and adsorbents were picked specifically such that they did not retain CO_2. The relevant paragraphs now read as follows:

“Sampling …

Air samples were pre-concentrated in a custom built thermal desorption unit (TDU, Kuyper et al., 2012). Adsorbents (Carbopac X and Carboxen 1016, 9 mg each) held in a glass tube were cooled to -20 ºC during the trapping phase. **To exclude air from the adsorbent trap a flow of helium (100 ml min^{-1}, Grade 5.0) was maintained both before and after sampling. Before being passed to the trap, samples were dried using magnesium perchlorate held**

in glass moisture trap (as per Moore and Groszko, 1999). Air was passed through the **adsorbent** trap at 100 ml min^{-1} for 15 min, resulting in a 1.5 l sample size. **The sampling flow rate was checked weekly by means of a digital flow meter.** The cooling of the system was achieved by a recirculating chiller filled with glycol. **An oil free piston pump was used to draw air through a 60 m Decabon sampling line and the adsorbent trap.** This was routed through a T-piece with the excess gas vented to the atmosphere. A mass flow controller was used to regulate the gas flow through the adsorbent trap. The pump was operated at 400 ml min^{-1} and a needle valve on the exhaust was used to provide sufficient pressure for the mass flow controller to operate.

A built-in resistance wire heated the glass tube to 400 ºC to desorb samples for injection. A second stage cryo-focusing system was used at the head of the column, with liquid nitrogen to improve the chromatography. The liquid nitrogen was held at the head for the duration of the primary injection. Thereafter, boiled water was used to desorb the samples trapped at the head of the column."

See above for for discussion of co-elution and note that as this is not intended to be a methods paper and given the method was previously published elsewhere, we do not feel the publishing this graph in ACP adds enough to the work here to justify its inclusion. However, should the reviewer/editors deem it is worthy of inclusion we would be happy to edit it and include for publication.

> *P5, calibration section: this section needs some further clarification as it is not clear how the calibration was done. How does 100-300 µl of pure bromoform equate to a concentration? Was it diluted prior to trapping? What is meant by a calibration loop? How were the 99% accuracy and 12% precision estimates derived?*

Response: We would like the thank the reviewer for bringing our attention to the ambiguity in this section and hope our description below is now clear. Attention has also been paid to calibrant sample flow and trapped volume. We apologise for the calibration error in the manuscript as originally submitted.

Remedial action: The calibration section in the methods has been rewritten and now reads as follows:

Calibration

"An external calibration method was used to verify the system performance. A custom built permeation oven was used to deliver aliquots of bromoform at varying concentrations to the trap (Wevill and Carpenter, 2004; Kuyper, 2014). A bromoform permeation tube held at 70 ºC (permeating at 343 ng min^{-1}) was flushed with nitrogen (grade 5.0, Air Liquide) at 100 ml/min. This gas mixture was continually passed through a 100 µl sample loop and exhausted through a halocarbon trap. Aliquots of 100-300 µl (1 - 3 sample loops) of the resulting permeation gas (bromoform diluted in nitrogen), were introduced to the thermal desorption unit from the permeation oven. **The sample loop was flushed for 30 s to ensure complete transport of the calibrant onto the adsorbent trap.** Calibration samples were passed through the drying trap as for air samples, thus any loss would be consistent for air and calibration methods. The calibration points were analysed using on the same temperature programme as air samples to ensure identical retention times. These were also used for the identification of bromoform.

A complete calibration curve (Fig. 2) was measured prior to the start of the experimental period. The peak area was determined from the injection of 1 - 3 loops of diluted bromoform in nitrogen gas. Peak areas were calculated through the trapezoid method of integration (Poole, 2003). These areas were computed in MATLAB. The mixing ratios of the injected loops were calculated as the number of moles injected. Each loop injection **resulted in 0.1865 ng of bromoform being loaded on the trap, based on the calibrated rate of the permeation tube (**Weville et al. 2004; Kuyper 2014**). The number of moles of bromoform on the trap was calculated. Through the air number density and the number of molecules loaded on the trap, the number of moles (bromoform) was converted to a mixing ratio. Calibration standards and air samples were run through the system independently of each other**.

The variability of the peak areas measured based on repeated loop injections was converted to a 95 % confidence interval. This confidence interval was used to show the uncertainty in the conversion of measured peak area to mixing ratio. Since the peak area is proportional to the concentration in the sample, the measured peak area is controlled through the number of injected loops and thus calculated against mixing ratio (Fig. 2).

Thereafter, a calibration point of 1-3 loops was run every 5 air samples to account for system drift. Based on a linear regression between the introduced sample and peak area response a 99 % accuracy was achieved on this system. Analysis from repeated 2 loop injections indicated a system precision of **7.4** %. **Following an analysis of the calibration curve a limit of detection of 0.21 ppt was determined for this system. “**

> *P5, Figure 2: Normally the fixed entity (mixing ratio) would be on the x-axis and the variable entity (peak area) would be on the y axis? What do the error bars represent (how many samples)? Why is the uncertainty given in the mixing ratio rather than the peak area?*

Response: The calibration is done by the injection of a known number of loops. The number of loops is an arbitrary measure however. What is of much more interest is the measured peak area and how this relates to actual mixing ratios since the peak area is related to the concentration or mixing ratio. By controlling the number of loop injections, we effectively control the peak area so this goes on our x axis. The mixing ratios are then calculated from the measured peak areas. The uncertainty displayed through the error bars reflects variations in the measured peak area from repeated injections. An example of the calibration curve calculated in this manner can be found in Wevill and Carpenter (2004).

Remedial action: As far as we can determine this figure is correct. The text has been revised to better explain the calibration curve.

> *P6, L6: “flow path” not “flow pass”.*

Remedial action: This has been corrected in the revised manuscript.

> *P6, L31: What is meant by “rapid shifts” on the 19th, 29th and 30th?*

Response: The rapid shifts referred to the rapid changes in wind direction.

Remedial action: The text has been amended to reflect this.

“The transit of weaker cold fronts caused the occasional rapid shift in wind direction …”

> *P6-7: what is the significance of wind speed?*

Response: Wind speed is important in gas measurements for a number of reasons. The bromoform sea-air gas flux rate may be approximated as a function of either the square or cube of the wind speed; e.g. Nightingale et al., (2000): $k = 0.31u^{2}(Sc/660)^{-1/2}$. The rate of atmospheric dilution also increases with wind speed. The wind speed can also be an indication of wind fetch. These factors combined play a role in determining the observed mixing ratio at any given point in time.

Remedial action: A discussion about the role of wind speed in the variation of bromoform mixing ratios has been added to the results and discussion.

> *P8, L7: "Measurements of bromoform at all ranges were recorded at CO levels below 100 ppb". If this is the case why are there no red or orange circles in this CO range?*

Response: One of the difficulties of polar plots is that the data are binned by wind direction and averaged. In this case the smaller measurements biased the few high measurements that occurred at below 100 ppb.

Remedial action: This figure has been removed and the sentence revised to be more accurate in the updated results section.

> *P8, L11: I fail to see the 2 periods of elevated ozone referred to.*

Remedial action: The time series figure has been updated and annotated to highlight the event periods. The discussion regarding ozone and in particular elevated ozone have been revised.

> *P9, Fig.6: What is the impact of boundary layer height on the measurements? This might also explain some of the variability. I am not convinced that the observed variation in ozone is sufficient to be able to get any real meaning from the analysis in Fig. 6.*

Response: The marine boundary layer height at Cape Point was calculated from twice daily radiosonde measurements made at Cape Town International airport, 60 km northeast. This is a rough approximation of the MBL at Cape Point. The height of the MBL was determined by the surface and elevated temperature inversion methods (Seibert et al., 2000; Seidel et al., 2010). The calculated heights ranged from a minimum of 91 m to a maximum of over 4000 m (Kuyper 2014).

Changes in the MBL height have in past studies been reported to influence the measured concentration of bromoform (Fuhlbrügge 2013). Despite this strong relationship reported by Fuhlbrügge et al (2013), no such relationship between bromoform and MBL at Cape Point could be established in the data presented here This could be explained due to a variety of factors. Firstly uncertainties may arise due to the approximation of MBL from Cape Town International Airport (some 60km away), as this might not be representative of conditions at Cape Point. In addition to this, if the bromoform measured at Cape Point was not locally sourced it would therefore independent of local MBL height and no effect would be expected. It could be that the MBL height as ascertained at Cape Town International airport is independent of the MBL at Cape Point and therefore not significant. At this stage we do not have enough information to separate or elaborate further on these speculations.

Remedial action: We have added information relating to the MBL at Cape Town International Airport to results and discussion sections.

> *P9, L9: What are the stated uncertainties in the reported maximum and minimum measurements and how do these differ from the somewhat lower uncertainties in the mean?*

Response: The uncertainties were calculated as a function of the precision of the measurements. The uncertainty in the mean is the standard deviation from the calculation of this value.

Remedial action: The uncertainties of the measurements have been reported as the percentages in the text rather than the calculated values.

> *P10, Fig.7: It would help if the 3 periods of interest were highlighted (shaded?) on the Figure. Can the authors say something about the very low values of bromoform on the Figure? There are a number of points very close to zero. Where does the air come from at these times? Can the authors be sure that this is not a measurement problem – it seems unlikely that values would drop to zero in a region where bromoform is generally rather high?*

Response: The values reported are around 4-5x above the calculated LOD and therefore are unlikely to be an artifact. We see similar low "background" concentrations reported in the literature making these observations consistent with similar studies elsewhere.

The calibration curve indicates that the system displayed a linear response to bromoform over a range of **0-40** ppt. An injection of 100 ***µ***l equates to **11.8 ppt**. As to the rapid changes, yes this can be expected. We see this in other regions (Pyle et al. 2011).

Based on the original calibration data, the wind direction at Cape Point when the bromoform mixing ratios were less than 5 ppt (n = 5) was predominantly from the west to northwest (245-320°). This is entirely within the background air sector, and further confirmed by radon concentration (< 250 mBq m^{-3}). These measurements typically occurred late in the afternoon/ early evening or in one case, before sunrise. This has been revised in the results and discussion using the updated figures and data.

Under the revised dataset there were 20 (14.8 %) measurements of bromoform less than 5 ppt. The wind direction was predominantly from the south (30 %), southwest and northwest (20 and 15 %, respectively). There was also a significant contribution from due east (20 %) of Cape Point. Of these measurements 4 had radon concentrations above 750 mBq m^{-3} and made up the observations from the east. The remainder of the measurements were all or marine origin (radon < 350 mBq m^{-3}).

Remedial action: The figure has been updated to include vertical lines to mark the events. See time series plot earlier.

> *P10, L4-5 and Fig.8: The wind speed associated with the higher concentrations to the NE and West seem very similar to me (one is described as "high" and the other as "intermediate to low"). I cannot really see any difference.*

Response: The reviewer is correct in their assessment. The wording of this sentence has been updated to be correct and less ambiguous.

Remedial action: The sentence and section have been revised in the updated results section. Updated polar plots of bromoform as a function of wind direction have been added to the text. The first figure shows the full dataset, while the second two show the background (WD: 120-320 °) and non-background (WD: 320-120 °) conditions of bromoform measurements at Cape Point.

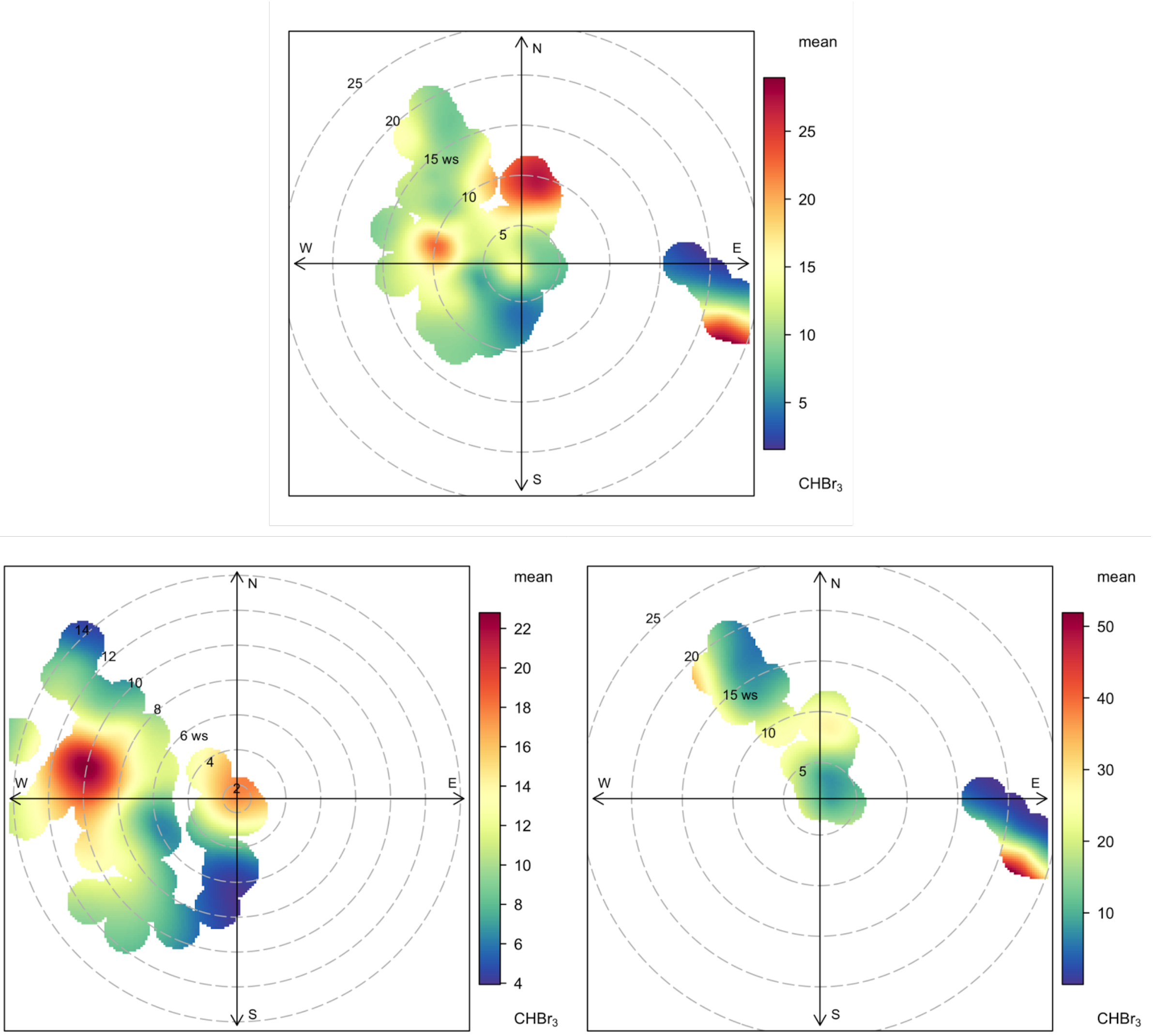

Figure: Polar plots of bromoform as a function of wind speed and direction. The top figure highlights all the data, while the lower plots show background and non-background respectively. These figures give an indication of possible source directions and distance.

> *P10, L6: This sentence needs rephrasing. I assume the authors mean that at low wind speeds the average concentration was 30 ppt and they are speculating that this is maintained by some "low level" local sources? What does low level actually mean and perhaps showing wind speed in Fig. 7 would help the reader to see this more clearly.*

Remedial action: This sentence and figure have been updated in the new results section.

> *P10, L12: "a background of low mixing ratios were observed from all wind directions". How does this relate to what was said in my previous comment? Was the "background" signal 10 ppt or 30 ppt? There are no data less than 10 ppt in the N, NE and SE sectors.*

Response: The reviewer is correct, we had introduced ambiguity through these statements. The reviewer is correct that the binned, mean data were greater than 10 ppt in the sectors N, NE and SE. The figure has been over-interpreted by the authors; especially in light of the fact that the values represented are binned means and not the actual observations.

Remedial action: The figure and text have been revised in the updated manuscript.

> *P11, episodic events: it would help to have a repeat time series for these events so the reader can see the patterns/correlations more clearly. Alternatively please put all data in one Figure (Fig 7) and perhaps some more axis markers to help distinguish between days.*

Remedial action: An updated figure 7 has been added to the text. This has also been annotated to highlight the three events.

Time series plots of the three events have also been added to the text in the results and discussion of the updated manuscript.

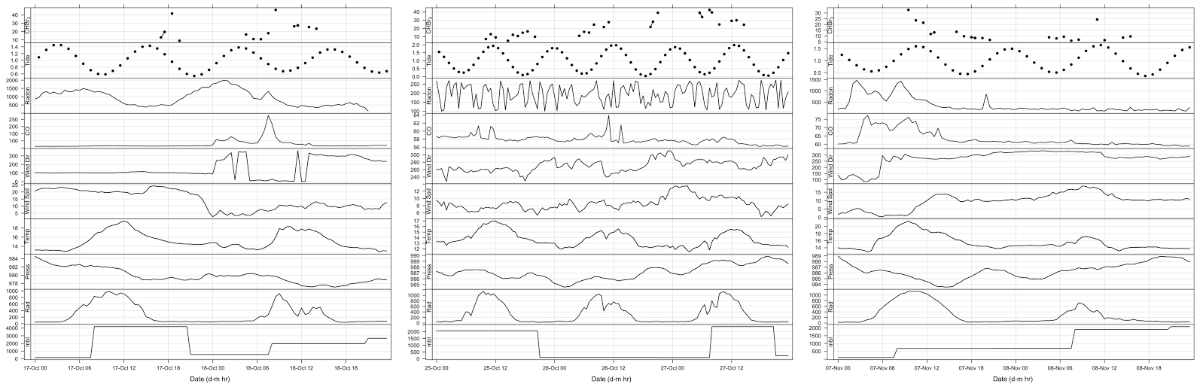

Figure: Time series sub-plots expanding the elevated bromoform event days. From left to right Events 1 - 3.

> *P11, L11 – P12, L1: The authors state that the concentration of bromoform decreased slowly between the maximum on the 18th until the 23rd. The only problem here is that there is a large gap (several days?) in the data when we have no idea what is happening. To describe this period as a single "event" is therefore a little odd.*

Response: We thank the reviewer for raising this point regarding assumptions of data over gaps.

Remedial action: The event windows have been refined to exclude the data gaps. The text has been amended to reflect that event 1 terminates with the cessation of sampling at the end of the 18th. The same applies for the third event where a data gap existed in the earlier defined window. Please also see the earlier time series plot.

> *P12-13, Event 2: The winds are predominantly from the west-north-west but the back trajectories suggest that the air is coming from the south and east. This apparent contradiction needs to be explained. In fact the trajectories in Figure 10 for Event (b) are dated November 2011, not October. Have the authors used the correct trajectories?*

Response: The reviewer is correct, the incorrect back trajectories in figure b were inserted. The authors would like to thank the reviewer for the observation.

Remedial action: The back trajectories have been updated and corrected.

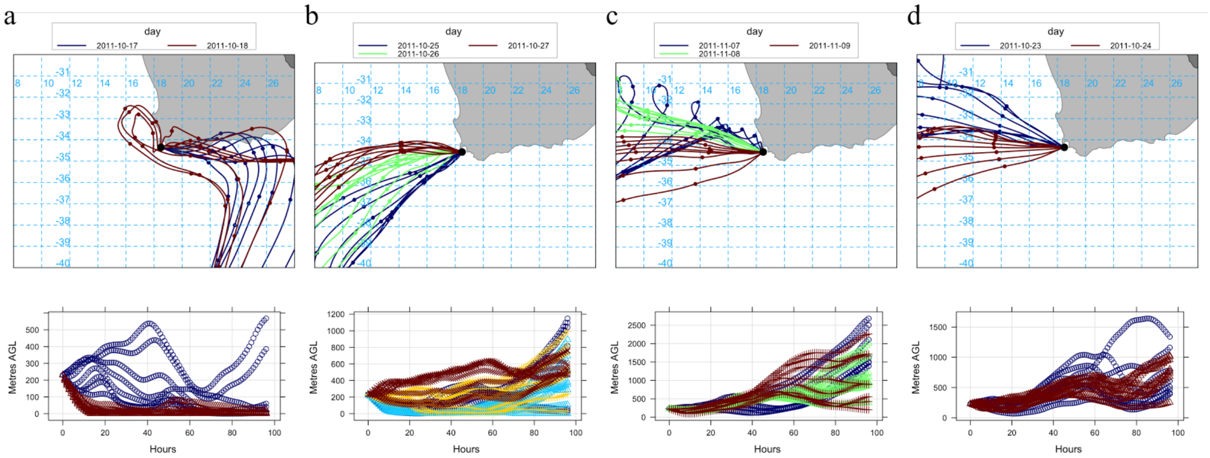

Figure: Composite daily back trajectories for the selected Events (a) E1 17 – 18 October 2011, (b) E2 25 – 27 October 2011, (c), E3 7 – 9 November 2011, (d) Background samples 23 - 24 October 2011. Trajectory heights for the events are displayed below. The colours and dates correspond respectively for each event.

> *P13, Event 3: again it is very difficult to follow the ups and downs in the various parameters. I find it hard to pick out 2 events in elevated ozone (line 11). Again there is a large gap with no data which makes linking the period more difficult. The authors cannot say that “bromoform rose to a maximum of 70.2 ppt” because there is a gap.*

Remedial action: The text has been revised to reflect the gaps in the bromoform data. The time series plot has been updated and annotated to highlight the events. Please see the time series figure above.

> *P14, Section 4.1: this section is highly speculative using such terms as “circumstantial evidence” and “tentatively appear”*

Response: Given the limited data set it is not easy to be decisive. The words ‘circumstantial evidence’ and ‘tentatively appear’ are entirely appropriate in the context of the data on which they are based.

> *P14, L14-15: If air is flowing from the Southern Ocean in a “north-westerly direction” over Cape Town, how is it possible for this air to then pass over Cape Point which is due south of Cape Town?*

Response: As a result of the Coriolis effect, anticyclones rotate in a clockwise direction in the Southern Hemisphere. A quick look at a typical synoptic chart (http://www.weathersa.co.za/observations/synoptic-charts) for a NW wind in winter in Cape Town will hopefully convince the reviewer that as the anticyclone passes south of the African continent, air flows from the Southern Ocean, over Cape Town and then down to Cape Point. These are the conditions which are typical in Cape Town winter and are those experienced in the description above.

Remedial action: The text has been revised to be more clear and accurate. The discussion has been entirely revised.

> *P14, L18-20: there is really very little concrete evidence for this anthropogenic source and its impact on the measurements at Cape Point. There is more coast directly to the north of Cape Town so even if the air was coming from this direction and picking up*

> *anthropogenic emissions, what is to say that the bromoform and CO/radon are not coming from completely different sources?*

Response: Nothing; it is entirely possible the $CHBr_3$ was entrained over the coast and then the CO over the city later. We intended to imply here that an Anthropogenic source could not be ruled out in these samples as the air had been influenced by human activities and will revise the wording accordingly.

Remedial action: Speculative text regarding source attribution has been removed.

> *P14, L21-22: how well do CO and Radon correlate for the entire period. It is hard to tell when they are on separate graphs using different axis ranges. It seems there are periods of high radon and low CO, but what about low radon and high CO? I assume this is unlikely if you assume a continental source for both, but it is very difficult to tell from separate figures.*

Response: Over the sampling period CO and radon measurements agree well. There are a couple of events where elevated CO can be observed while radon is low. These brief CO 'peaks' are potentially experimental artefacts but that discussion is beyond the scope of the work presented here. Brunke et al. (2004) show the source regions of CO and radon at Cape Point, based on numerous years of measurements.

Remedial action: The time series figure has been updated to include the radon measurements as well as the CO on the same time scale. Please see the time series figure above.

> *P14, L30: why have you not investigated the impact of tides at the local site? (see major comment above and the next comment below).*

Response: Please see the major response above. An investigation of the tides was performed. For more complete details on the tide analysis please see Kuyper, 2014. The conclusion from Kuyper (2014) were that the tides played no significant role on the variability of the measurements at Cape Point.

> *P15, L4-5: "The extensive kelp beds at CP may contribute bromoform to both the consistent baseline and extreme events observed". If this is the case, can the presence of local kelp beds not explain the entire set of measurements? Without ruling this out, the majority of the preceding discussion is surely obsolete? Where are the local kelp beds? Are they underwater or exposed at low tide? If the latter, do you see an impact of local tide time with bromoform concentration?*

Response: Please see the major response above. The kelp beds are not totally exposed at low tide. It is possible that emissions from the local kelp beds explain the majority of the variability. From the updated time series plots, it is suggested that the tide height may be a contributing factor in the elevated bromoform events, but not an independently sufficient factor. As was noted above the data presented here do not conclusively refute any of the test hypothesis A-C. The kelp beds, as the dominant bromoform source, may have also been overlooked on an event scale basis as the published emission rates were not sufficiently large enough to explain the observed elevated bromoform mixing ratios.

Remedial action: Source attribution has been removed from the discussion. The possible tidal influence has been added to the discussion, more completely. The role of tidal height and kelp exposure as a dominant source of bromoform to the atmosphere at Cape Point has been carefully explored in the revised manuscript.

> *P15, L6: what is meant by "other typical meteorological conditions"?*

Remedial action: We have added to the text:

"…such as air temperature, pressure, rainfall and global downward radiation…"

> *P15, L8: "quasi-continuous" is better than "quasi-discrete".*

Remedial action: As per the reviewer's suggestion this has been changed throughout the updated manuscript.

> *P15, L21-22, and Table 1: Why not report some median values as well as means?*

Response: None of the authors of the papers from which the values were drawn report the median values of the measurements made. We have report past results exactly as per Quack and Wallace (2003), who also report only mean values in their extensive and authoritative review.

Remedial action: The table has been updated to include more recent measurements from coastal as well as open ocean reports.

> *P15, L25-26: the evidence for an anthropogenic source of bromoform is not really apparent (see earlier comments).*

Response: The reviewer is correct and thanked for pointing this out to us. The updated figure (time series) in the results and discussion should better highlight the evidence of an anthropogenic enhancement.

Remedial action: The results and discussion have been revised as detailed above.

> *P15, L26-27: radon CO and ozone were not all elevated throughout Event 1. CO and radon were elevated at times during the period, and it is hard to say whether ozone was elevated or not. Higher ozone wouldn't necessarily be an indicator of recent anthropogenic influence. Were there no other tracers in the GC output that might help?*

Response: The only compound calibrated for at the time of the measurements, was bromoform.

Remedial action: The discussion surrounding O_3 has been revised.

> *P16, L1-7: this section is highly speculative and rather confirms that no conclusions can be drawn as to the importance of any anthropogenic source. It would be helpful if some measurements could be made near to the water processing plants to confirm the levels of bromoform.*

Response: Conducting studies into the kinetics of the bromoform production from water chlorination and the resulting budgets to the atmosphere is a whole field of research in and of itself. This however falls beyond the scope of the work presented here which sets out to try and investigate the concentrations in unpolluted baseline air from biogenic sources. The anthropogenic contribution is discussed as a potential additional source, and it may become an important factor in the rare circumstances we have evidence to show the air being sampled is anthropogenically influenced but, this is not the main thrust of the work. As noted above we do claim to conclude that there is an anthropogenic source.

Remedial action: The discussion has been revised to focus on the relationships between the bromoform measurements and the GAW measurements in background air masses. Source attribution has been limited to speculation and this has been made clear in the revised text.

> *P16, L11: I cannot easily identify a period of "moderately elevated ozone"*

Remedial action: The line has been removed. Ozone has no significant departures.

> *P16, L12: How was the ozone "biogenic in origin"?*

Response: The authors were attempting to imply that the ozone was stratospheric in origin and it was not formed from anthropogenic precursors.

Remedial action: The line regarding '*biogenic in origin*' has been removed.

> *P16, L15: "from the west" - see point earlier about the discrepancy between measured wind direction and the back trajectories.*

Remedial action: We would like to thank the reviewer again for this observation and the back trajectories have been amended accordingly.

> *P16, L16: there is no clear evidence in this analysis that supports the theory of ozone-induced bromoform release.*

Response: The authors would like to thank the reviewer for their comments on this.

Remedial action: The results and discussion have been rewritten.

> *P17, L6: the Benguela current is far to the north of CP according to Figure 1. How will this affect the concentrations at CP during Event 2? I do however agree that a study of the local kelp would be a sensible thing to do.*

Response: The Benguela Current is not exclusively far to the North, but extends all the way down to Cape Town. Moreover, with an atmospheric lifetime of 24 days it is quite possible for bromoform formed in the Northern (and more intense) Benguela to travel to Cape Point. Under north-westerly winds air is drawn from the Benguela region over Cape Town and to Cape Point. Species composition of seaweeds in the region has been discussed above. The overinterpretation of the data may have been a function of stretching all known sources to the limit to explain the elevated mixing ratios. This should be mitigated with the revised data, which is much more inline with previous measurements.

> *P17, L19: It has not been proven that the anthropogenic source of bromoform was strong during Events 1 and 2. This statement is inaccurate. In fact the whole of this final paragraph is highly speculative.*

Response: We agree with the reviewer. That line should not have appeared in the final version.

Remedial action: The discussion text has been entirely revised to indicate that this is our speculation and not a fact.

> *P17, conclusions: The whole section will need to be rewritten once the various issues above have been addressed. I do also note that the extremely high values reported from Gran Canaria were measured many years before the majority of data in Table 1, so, with due respect to the original authors, I would perhaps treat these data carefully.*

> *There have been substantial improvements in analytical technology and calibration since these measurements were obtained.*

Response: The reviewer is correct that the conclusions need to be updated along with the results and discussion sections.

Response: The conclusions have been revised accordingly with the Results and Discussion sections as per the reviewer's comments and suggestions. The Gran Canaria data will be treated with some caution.

The authors would like to again thank the reviewer for their time and effort in examining the paper. Their comments have been helpful in greatly improving the quality of the paper.

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