1 Referee 1

We are grateful for the comments and edits of the anonymous reviewer who invest time for the revision of our manuscript. Our responses were given in red ink for each comment/edit and corresponding changes were applied in revised manuscript, accordingly. Since the line numbers are updated in the revised manuscript, new line number are given in the responses.

Anonymous reviewer 1	**Authors response**
One item that would improve presentation of the data is to plot the variables in figure 6 against the measures of hydrothermal vent plumes shown in figure 4. This would allow the reader to easily link the parameters that show the presence of a plume with the data collected in those samples. Lacking that the reader is left to squint at multiple depth profiles.	Turbidity profile of PL (originally at Fig. 4) and corresponding non-buoyant plume layer boundaries are added to the Fig. 6.
The manuscript should have been checked for errors before sending it out – missing citations and a bibliography with no year information are careless errors.	Year information are at the end of each bibliographic items. End-text citation format is given by the journal and automatically generated by a reference management software. In-text and end-text citations are checked again and errors are fixed.
Line 28 ‘less abundant compositions’ – this sentence is too vague to interpret. Do you mean abundance in terms of number of elemental formulas/mass-to-charge values or abundance in terms of peak areas? (note that	Line 29-31: The sentence is updated to “In comparison to background sea water, we found that the DOM in waters directly affected by the hydrothermal plume was molecularly less diverse and 5-10 % lower in number of molecular formulas associated

as I read later the methods make it clear you are talking about number of formulas).	with the molecular categories related to lipid and protein-like compounds".
The Hawkes and Rossel papers are only appropriate citations for the alteration of refractory DOM. The work on production of dissolved hydrocarbon gases requires other sources, likely papers from Seewald and colleagues.	Line 42-45: Corresponding sentence is updated as: "Continuous exposure to extremes of temperature (up to 400°C) and low pH values either completely degrade DOM to volatile species (e.g., CO2, methane) (Lang et al. 2006) or alters originally stable, high molecular weight, refractory DOM to highly aromatic, unsaturated, oxygen poor, petroleum-like compositions in the hydrothermal vent fluids (Hawkes et al., 2015, 2016; Rossel et al., 2015, 2017)." As referee pointed out "the production of dissolved hydrocarbon gases" and related papers of Seewald and colleagues were not meant to be emphasized here. Yet, Seewald coauthored papers were cited elsewhere in the text.
Line 49 – what about the research of Arrieta et al. which does show it is available, but too dilute to be used?	Line 50-52: Sentence is updated as follows: "Considering that the deep ocean DOM is refractory (e.g., Dittmar and Stubbins, 2014; Hansell, 2013) or primarily inaccessible for organisms (e.g., Arrieta et al. 2015)…".
Line 97 'Ocean Floor Observation and Bathymetry System' – I have no idea what this is, an ROV? Camera system? AUV? CTD/rosette system? Bottom lander?	Line 100: Sentence is updated as follows for clarification: "…using a towed camera system (Ocean Floor Observation and Bathymetry System (OFOBS); Purser et al., 2019)."

Line 109 – there appear to be references to a figure or table missing.	Line 112: This was meant to refer Table 1. The error is fixed.
Line 115 – what does ‘2s’ mean ?	Line 118: Two times standard deviation. This is updated in the text.
Line 145-how do you have a sensitivity for the fluorometer in ug/l without a description of how it was calibrated to convert volts to ug/l? Also, from the results section, I don’t think you mean sensitivity here, but detection limit.	Line 147: Both, sensitivity, and the detection limits were given as 0.01 µg/L for WET Labs ECO Chlorophyll-a fluorometer User Guide. WET Labs uses the chlorophyll equivalent concentrations as the signal output which is factory calibrated against *Thalassiosira weissflogii* phytoplankton culture. Calculation for the scale factors and voltage conversions are given in the user manual which is accessible from the following URL. https://www.comm-tec.com/prods/mfgs/Wetlabs/Manuals/Eco-fluo_manual.pdf Since this information is provided in the manual, we did not think that is necessary to indicate here.
Line 184 – how did you determine which adduct to use when combining the positive and negative ion mode data? And how did you handle m/z values that are multiple adducts from a single neutral mass?	Line 185-188: Relative intensities were calculated separately in negative and positive ESI modes by normalizing most abundant ion in the spectrum. When a molecular formula has the same CHNO/S combination in both positive and negative modes, relative intensity of the negative ion was considered. Calculations were also checked by considering otherwise (positive

	ion intensities), yet the results were almost identical (e.g., relative intensity profiles (Fig.6b), diversity index (Fig. 6d), PCoA and cluster analyses (Fig. 7b-c)).
Line 197 – can you describe the percentage calculation a different way as this is not clear. How are you grouping LPD/CAR/LGN/UHC? And why is it different from the sum of CHO/CHON/CHOS? As I read later, I think the authors have done this: LPD + CAR + LGN + UHC = 100% And a separate calculation for the elemental formulas to be : CHO + CHON + CHOS = 100% However, what about elemental formulas that have multiple heteroatoms (CHONS?)	Line 202: Percentage calculations for the molecular categories were calculated as is pointed out. No elemental formulas obtained with CHONS combination in our analyses.
Line 268 'No substantial primary production occurred in the top 50 m layer of the water column, as shown by the nutrient minima and the Chl a maxima at ~40 m water depth. During the Arctic summer (from March to September), this layer gets depleted in inorganic nutrients and enriched in dissolved organic matter' – I am not certain what is data from the present project and what is speculation. Since the samples in this project were collected in Sept/Oct, where is the source for the March to September nutrients? And, if nutrients are low in the surface, that could be an indication they have already been consumed and hence that	Line 274-276: To mitigate our statement, the text has been changed and a reference has been added as follows: "At the time of sampling, the net growth of phytoplankton seems to decelerate at the top 50 m layer of the water column considering the nutrient depletion and the subsurface maxima of Chl *a* at ~40 m water depth. During the Arctic summer (from March to September), this layer gets depleted in inorganic nutrients and enriched in dissolved organic matter (Thingstad et al., 1997) ."

is not a good marker for low primary production. From the sample set they have, the authors cannot make statements about primary production, and only can provide statements about the end products of primary production.	
Line 290: ‘do not indicate any anomaly in relation to plume dispersion distant from the vent due to dilution with seawater’ – I agree with this statement, but it would be easier to see on the figures if you mark the depth of the buoyant plume on figure 3.	Line 298: The non-buoyant plume layer depths are now indicated in Fig. 3 as suggested.
Line 293 – ‘This seems contradictory to molecular changes in DOM compositions’ – at this point in the manuscript you have not discussed the DOM composition so the reader has no basis to understand this point.	Line 299: A reference tag is added to the sentence to indicate that is not mentioned yet in the main text. “This seems contradictory to molecular changes in DOM compositions (as detailed further in Section 3.3.3)…”
Line 384 ‘precluding a proper assessment of the geochemical processes influencing methane there’ – this phrasing is odd. The authors are clear about the caveats, but don’t end with statement saying you cannot do this analysis (after spending a page doing it).	Line 395: The sentence was removed from the text.
Line 392 ‘the features obtained in different modes, combining positive and negative ESI datasets provides a considerable advantage for differentiating samples.’ How? The	Line 404: A reference to Supplementary Figure 4 is added. This figure demonstrates different features that are captured by the positive and negative ESI.

previous paragraph does not provide any information about different samples.	
Line 424 ‘ DOM abundances’ is too vague – please continue to be specific and refer to the number of elemental formulas since you also have DOC concentration data.	Line 436: We now specify the term with “average relative intensity” throughout the text and figures.
Line 428 ‘CHON and CHOS heteroatom contents were maximal at the surface’ – this is a stretch. Looking at figure 6j, CHON % is higher at depth than the surface (which the authors note in the next sentence), while the range of CHOS is so wide at the surface and depth that you cannot make any such statement.	Line 442: This sentence is updated as follows “CHON and CHOS heteroatom contents were higher at the surface compared to subsurface contents up to 500 m depth (Fig. 6i-k), indicating …”
Line 436 – this paragraph is speculation and statements about global carbon use from six stations near a hydrothermal vent site only detracts from their messages about organic matter from hydrothermal vents.	Line 452: The last section of the paragraph is removed to prevent speculative statements.
Line 470 – figure 5 lacks subplots so this is an error.	Line 482: Corrected as Fig. 6a-e.
Line 525 – reference to the wrong figure again.	Line 536: Corrected as (see Fig. 6f).
Please add years to the references information.	Year information are at the end of each bibliographic items. This format is given by the BG journal.

Figure 3 – why are there lines connecting some samples and not others? I would remove the lines entirely as they do not aid in interpretation of the data in the figure.	Changed as suggested.
Figure 5: Mid-Atlantic Ridge. Also, I think I understand the plot the range of endmember values from the different systems, but the way this is plotted it appears that the other systems studied all have 1/methane concentrations of zero. I would put the comparisons to other end members to the left of the 0 value on the x-axis to avoid this interpretation. Why is Pedersen et al. 2010 discussed in the text but missing from the figure?	We modified figure 5 according to the reviewer suggestion. We did not add Pedersen et al. 2010 to the subtitle, because this report includes the discovery and the description of the vent fauna of the area but does not report methane isotope compositions. Therefore, this reference is not added to the figure but cited in the text.
Figure 6b, what is average abundance? I suspect this comes from the confusing statement in the methods (line 28), but even seeing the plot I still do not understand what is calculated here.	Confusing terms are revised and corrected at both locations (at line 28 and Figure 6b) and throughout the manuscript. Average abundance is updated as average relative intensity. Method for the calculation relative intensities are given Line 185.
Figure 7 – what is the variability for each of the PCoA axes? This information is needed to interpret the distribution of points in the multidimensional space.	Percentage of explained variance is added into both axes. The method for the calculation is added in Section 2.4, Line 212-215.
Table 1: correct to parentheses. Also when you list 'DOM' in the table here, do you	Table caption is corrected. DOM is changed to DOM composition for clarification.

mean DOC concentrations or SPE-extracted DOM? This is not clear.	

2 Referee 2

We are grateful for the comments and edits of the anonymous reviewer who invest time for the revision of our manuscript. Our responses were given in red ink for each comment/edit and corresponding changes were applied in revised manuscript accordingly. Since the line numbers are updated in the revised manuscript, new line number are given in the responses.

Line 117: Is the GF/F filter pre-combusted?	Line 120: Yes, filters were pre-combusted. This was forgotten to mention in the text and is now fixed at reviewed version.
Line 118: Are the HDPE bottles acid-washed before use?	Line 122: No, HPDE bottles are not acid washed but rinsed couple times with the sample and a brand-new bottle used every occasion. We have previously observed that there is no contamination added from the HPDE flasks for DOC and nutrient measurements
Line 124: 'the cartridges were then eluted into pre-combusted, amber glass vials with 2 ml methanol', previous studies have generally used >6ml of methanol to elute DOM from the PPL-cartridges (e.g., Dittmar et al., 2008). So, I wonder if the DOM on the PPL could be completely eluted by 2 ml of methanol?	Line 127: Volume for the elution were updated from Dittmar et al. in our experiments. We observed that using < 2 mL MeOH does not elevate the extraction efficiency. Yet, using 2 mL volume enables us to use the eluted samples without additional evaporation and resuspension steps and decrease contamination potential.
Line 367: 'the $\delta^{13}CH_4$ value of the hydrothermal fluid source to be about -36 ‰', the estimate of fluid endmember should include the error derived from the curve	Line 371: The text is updated as follows to include intercept uncertainty and R^2 values: "…we estimate the $\delta^{13}CH_4$ value of the hydrothermal fluid source to be -36 ‰ using a least squares linear regression of the entire

fitting. And, the error needs to be considered in the discussion.	plume methane dataset (R^2 = 0.59), with an intercept uncertainty of ± 3‰ at the Aurora hydrothermal field...”.
Line 398 and Figure 6b: What does ‘average abundance’ mean? The signal intensity of each formula? Please define it where it first appears in the text.	Line 406: Average abundance term is modified to average relative intensity throughout the text and was defined in Section 2.3 as follows: “Relative intensities were calculated by normalization with the most abundant ions in each mass spectrum (Kujawinski et al., 2009).”
Line 421 (Fig. 6a-d, Fig.S2) and Line 424 (Fig. 6a-d, Fig.S2): Refer to wrong figures?	Line 434: Corrected as (Fig 6a-d, Fig. 3a-e) and Line 438: (Fig. 6b-d).
Line 420-421: Changes in DOM composition seem inconsistent with the changes in nutrient and DOC. For example, the average MW, diversity index, and UHC percentage of UL-1000 are similar to that of surface water (UL-5), while DOC/nutrients (except ammonium) of UL-1000 are much lower/higher than surface water.	Line 432: This was addressed in the first paragraph of the Section 3.2, Line 298-302, by stating that compositional changes are not necessarily coupled to bulk concentrations changes. To provide a link between sections a reference tag is added in the text as “This seems contradictory to molecular changes in DOM compositions (as detailed further in Section 3.3.3)…”.
Line 460: ‘H:C≤1.0’ should be ‘O:C≤1.0’.	Line 472: Corrected.
Line 469-471: Low molecular diversity and relative abundances (average abundances?) are also observed in BG-samples at similar depths (Figure 6b,d). So, it is not sufficiency to demonstrate the influence of	Line 482: We agree to the referee that the diversity index and average relative intensities may not be sufficient to demonstrate the influence of hydrothermal intrusion in lateral transect. This was the

hydrothermal intrusion or the plume based on these data (i.e., molecular diversity, average abundance) alone.	reason why we also indicate LPD percentages to emphasize compositional differences.
Line 529-532: This explanation seems more plausible to me, as the difference in DOM compositions between NP2-samples and BG-samples is greater than that between PL-samples and BG-samples (Figure 8).	Line 541: This was added to the end of the section just for the flow of the text.
Table 1: According to the text, DOM compositions of PL-3400 and PL-3500 have been measured but they are not labeled with superscript ‘3’ in the table.	Suggested edits are corrected in the table.
Figure 7: The letter numbers of panels are inconsistent with the caption. In addition, the contributions of the PCoA1 and PCoA2 to the total variance need to be presented.	Letter numbers of the panels and the captions are corrected. Percentages of explained variances for both axis are added.
Figure 8: According to the Krevelen diagram, there are differences in DOM composition among different BG-samples, yet, the authors did not discuss the possible reasons for these differences in the current manuscript. Do the differences in DOM composition of different BG-samples indicate effects of non-hydrothermal plume processes? This information may help to distinguish the effects of hydrothermal and other processes.	This was partly mentioned in the first paragraph of the section 3.3.3 stating that there were some exceptions in clustering of the samples by stations. But we unfortunately do not have further evidence for the reasons of compositional differences between samples from same stations.

Supplementary: Figure3, 4 and 5 have wrong numbers.	Corrected.