

This paper presents a comprehensive cataloguing of common lipids (fatty acids, hydrocarbons, sterols etc) in the Beaufort Sea and surrounding region. Lipids are isolated from riverine and marine suspended particles and the upper few mm of sediments. In some cases, compound specific $\delta^{13}C$ values are also presented. The goal of the paper is to use lipid biomarker data to attribute sources to organic matter entering and accumulating in this region of the Arctic.

The study builds on several previous investigations in the region; including some that analyzed a subset of the biomarkers that were combined in the current work, and still others that applied stable and radioactive carbon isotopes. The authors should state clearly why this study improves upon previous investigations, and what new insight it offers.

The multitude of data while impressive is poorly presented and discussed. For example, statements such as “flagellates are the main contributor to SPM” or “reflecting post bloom conditions,” or “fossil carbon is the main contributor to SPM,” appeared in the text before any evidence/justification in support of such statements was provided. This gives the impression that these lipid distribution data are not being interpreted objectively. In my opinion the major shortcoming of the paper is that it lacks a clear statement explicitly addressing the assignment of certain classes of lipids to either a particular source or a process. For example, at the beginning of the discussion the authors could include a schematic, table etc that lists each relevant biomarker and its particular purported source. This table would be accompanied in the text by a description of the evidence (from the current study and previous studies) that supports each source assignment and the limitations of each assignment. [“In this paper we quantified the fossil contribution to SPM by integrating the area under the UCM (a region that occupied between x and y temperature range in the gas chromatogram). We assumed that everything in the UCM was derived from fossil organic matter, this assumption is valid because etc etc. Flagellate production was identified when the presence of 22:6 > 22:5 and 18:1 etc etc. Fresh diatoms were identified as.....We use “x-suite” of lipids to assign the detrital or refractory algal contribution...] The remaining discussion could then be organized around this initial section to make the discussion more coherent.

My overall recommendation is to consider the following:

- (1) What is the main (new) contribution of this study?
- (2) Results and discussion points are mixed in the results section. This section should simply report data.
- (3) A section that assigns particular lipids as proxies for particular source contributions should be included at the beginning of the discussion.

Specific Comments.

P.13933 Lines 5-10. Is this discussion of CPI being ~ 1 based on what is shown in Table 3? I am not as familiar with this literature, but these data suggest that there is some odd preference. How accurate is the terrestrial n-alkane calculation?

Line 15. UCM used as a fossil indicator? Be specific.

Line 17 – Does this discussion regarding the abundance of long-chain plant waxes pertain to the calculation in line 8? Or does the abundance reflect what was actually measured via GC-MS.

In general it would be helpful to clean up this section. I would first report the abundance of n-alkanes as measured directly – this is what would be normally found in a results sections. The authors can then state their assumptions for “correcting” the data for a fossil contribution, and then report those abundance data separately. The way it is currently written I can’t tell whether the calculated and measured data are both being discussed. Also, why don’t the authors simply subtract the UCM contribution (e.g., a baseline subtraction) from each n-alkane peak rather than assume that a petroleum source is contributing the entire n-alkane series. I apologize for my limited familiarity with this topic, but it seems to me that the authors are using previous studies to assume that a fossil component is present. This is then extended to interpreting the GC-MS data.

p.13936. Line 11. It is not accurate to attribute all fatty acids listed here to only a flagellate source. Just as with hydrocarbons for example, multiple sources can contribute many of these fatty acids. The authors should identify the suite of fatty acids that they think are indicative of flagellates (22:6, 18:4?). I see that they explain this later on p. 13941. They need to either move this explanation up to p. 13936 or remove the source attribution in the results section and simply report the ID and concentration of fatty acids found in each sample.

p.13937 – line 6. The conclusion that positive PC1 associated biomarkers are reflective of refractory marine and terrestrial OM seems tenuous to me. There is not much discussion in preceding sections that contributes to this conclusion. Could it be indicative of equally fresh secondary processes? In fact the attribution of +ve PC1 sources is not based on lipid composition but what comes later in the paragraph – that +ve PCI samples are from deeper depths. It seems that this discussion is backwards. I.e Line 14 -21 should come first.

Line 17-21. Here the authors point out samples that don’t fit into their previously discussed trends. Instead of identifying the lipids that make these samples unique (as would be expected in a cataloguing of results) the authors attribute sources/processes to these samples without explicitly stating their reasons for doing so. The PCA does not identify sources, it simply examines similarities and differences in lipid “profiles.”

p.13938

The $\delta^{13}\text{C}$ Results section demonstrates some of the difficulties that I experienced when trying to decipher this paper. The first two paragraphs present results on the extremely depleted $\delta^{13}\text{C}$ value of certain lipids in marine SPM. However, on line 16 the authors discount the possibility of aquatic plant input to odd, mid-chain n-alkanes because $\delta^{13}\text{C}$ values are depleted (-30 and -31 per mil in this case), and such a depletion is indicative of terrestrial origin (not aquatic origin). Given the source ambiguity of ^{13}C depleted isotope signatures – a fact the authors themselves point out several times – if it not correct to use depleted isotopic values in one case to say that only terrestrial inputs are relevant, and in another case to say that in fact, an aquatic source with an unusually depleted signature is implicated. I am sure the authors have a good reason for making this statement, but the reasoning has to be stated more explicitly. Again on line 23, the authors state that heavy isotope values for IP25 are entirely consistent with OM of planktonic origin (e.g. phytol). But it appears from the data that the values are only consistent with phytol in the sediments and not all phytol. Incidentally, “indistinguishable” appears to assume that there is no difference between a value of -17 per mil and -27 per mil (line 6). Also, it is curious that only a subset of data that appears in the discussion section is directly presented in the results - e.g., the isotopically depleted c-17 n-alkane is the first topic tackled in the discussion, yet it is absent from the results (except in the table). Again, this speaks to lack of coherence.

Discussion

p.13939. Line 14. Which lipids are considered in the “fossil alkane” category and where are these data presented? Does a value of -32.7 really contrast with the -30 per mil figure?

p. 13941 - Last paragraph of section 4.1. The preceding section did not allude to the dominance of fossil lipid at all. How did the authors arrive at this conclusion? Is this based on n-alkane concentrations? Isotope values? As far as I can tell n-alkane isotope values are not unique and concentrations are quite low compared to other lipids. Based on what is said later the authors must be referring to the “size” of the UCM “peak.” Is the UCM a definitive indicator of fossil inputs in aquatic SPM or is it simply an indicator of sample complexity. I would venture to guess that without some other indicator of high petroleum/fossil input it is difficult to definitively assign the UCM to fossil OM. If nothing else, some qualifications or justifications should be provided. It was frustrating to review this paper for exactly this reason. I found myself having to jump from one section to another over and over again to figure out whether I had missed something.

p.13942 – Why is there a big difference between SPM samples and the upper 5mm of sediments in terms of phytol- $\delta^{13}\text{C}$?

p.13943 line 18. Here is another one of those statements that seemingly come from nowhere – “post bloom conditions” – what is the basis for this statement? Line 20 – if you are in post bloom conditions would CO_2 be replete?

Line 24. Aerosols appear in the discussion for the first time. Is this really an important discussion point? The interpretation may certainly be correct but there is no independent evidence in support of this statement.

p.13944. Line 5. "Higher growth rates at depth compared to DCM." Do you mean higher growth rates for zooplankton? There are some isotope data for alcohols in Figure 4 but there is no depth information here. How different are the values? If the authors are referring to phytoplankton growth rates then I am even more confused as to why growth rates would be higher below the DCM.

Line 10-13. "herbivorous grazing on phytoplankton." Based on what we know from foodweb studies this should take place, but how is that tied to what your data show? What exactly allows you to draw this conclusion? The presence of both diatom biomarkers and zooplankton biomarkers in suspended POM, at a particular depth, does not necessarily mean there is a connection between the two or does it? Are the authors assuming that diatom biomarkers can only get to depth once they are repackaged by zooplankton grazing? I think these organisms could contribute independently to the sinking flux. Again, it is not outlandish to suggest a connection, but either sticking with the evidence or being more explicit about the conclusion would be more satisfactory. Also, this is not a big deal. I would be more than happy to let one statement like this stand. However, in the case of this paper these seemingly subjective conclusions are relatively commonplace.

Line 24. This belongs in section 4.1

p.13945 – paragraph starting on Line 9. This is the most coherent and realistic section of the discussion. In fact, I don't think the preceding discussion is really necessary. A slight expansion of this section (to include a brief statement as to why each lipid is assigned to a particular category) would make a better discussion. Alternatively, a general section that assigns each lipid to a source based on composition, other indices, and stable isotopes could precede this section (as I pointed out previously).

p.13946 – Line 16-19. Then why do you see such a big difference in $\delta^{13}\text{C}$ values between sediments and the water column?

Line 22-24 I don't understand how these things are connected?

p.13947. The results of this mass balance were used to inform the discussion about sitosterol in section 4.2. There it was out of place. Again, I reiterate, this makes the paper very hard to follow. Conclusions should only be drawn after they have been empirically (in this case) justified.

p. 13948. Line 1. I am assuming that by 'deep sediments' you mean "sediments underlying a deeper water column." All your sediment samples came from the top 5 mm?

Line 11. Does this discussion confirm or weaken your discussion point in the previous page (line 15-20) where you determine that between 55-60% of sitosterol in your “marine” sediments is derived from algal sources. These two discussions again show a lack of coherence - data from different biomarkers are interpreted individually rather than being used together to provide a unified view.

Line 28 – again “with depth” is misleading here. I would recommend “with water column depth.”

Section 4.4. There appears to be a significant amount of speculation in this section. E.g., “This is consistent with the well-known fact that picoplankton is efficiently recycled within the food web and only large phytoplankton is exported.” Isn’t it possible that flagellate-associated carbon had not yet been exported at the time that these data were collected?

p. 13951. Line 14. I am still a little unclear about whether the *terrestrial* n-alkanes are quantified based on what is measured directly (i.e., integrating the area under the peak) or only after making the petroleum-derived n-alkane correction? If the discussion refers to the latter then wouldn’t it be better to use a biomarker whose abundance has not been manipulated in this way.

P. 13952 . Line 11 etc. I am not sure that these studies are comparable.