We thank the reviewers for their thorough and constructive comments. Our point-by-point responses are *italicized.*

Response to reviewer#1

General comments

1. The authors claim that the AQY at 300 or 330 increases exponentially with salinity across different oceanic regimes (Fig. 4), arguing that salinity may be an indicator of CDOM origin (terrestrial vs. marine). However, their calculations do not seem to take into account the variation in AQY due to temperature. The irradiation experiments were conducted at different temperatures in each of the studies, for instance, at 2 – 4°C in the present study and at 21°C in the Sargasso Sea study done by Toole et al. (2003). That same study showed that a 20°C increase in temperature produced approximately a doubling in AQY. Obviously, this may modify the AQY-salinity correlation encountered, which is one of the core messages of the paper. This issue must be addressed.

   This is a good point. All AQYs are now normalized to 4 °C according to Toole et al.'s (2003) result. The correlation between DMS AQY and salinity slightly decreased after temperature correction but still remained highly significant at the UV wavelengths. Several new lines are added:
   "Note that the DMS AQYs for the Sargasso Sea, NE Pacific, and Bering Sea were all determined at 20-21°C, which were 16-19°C higher than the irradiation temperatures employed in the present study. Correction for the temperature-dependence of DMS photolysis based on the result of Toole et al. (2003) for the Sargasso Sea somewhat decreases the coefficients of determination, but the correlation between and salinity still remains highly significant ($p < 0.0001$) at the UV wavelengths (Fig. 4D,E). Salinity can thus···"

   In addition, please see the new Fig. 4.

2. There is a dearth of information regarding the description of vertical mixing and underwater spectral light attenuation (Kd) in the study area.

   - I also have some concerns regarding the measurement of CDOM in highly absorbing and scattering (turbid) waters. These issues deserve a more detailed treatment in the paper.

   Please see our response to specific comment 10.

   - It should be explained somewhere in "Materials and methods" how the Upper Mixed Layer Depth (UMLD) was calculated. This is relevant for the results of this study, as the authors themselves recognize, because they assumed that the UML was homogeneous in terms of [DMS] and optically-active substances. Using a fine UMLD criterion (e.g. < 0.05 kg m-3 density difference) is more likely to render an actively mixing or recently mixed layer. Using a coarser criterion may capture a layer that is not homogeneous in terms of [DMS], because DMS undergoes fast biological/photochemical cycling.

   The 0.05 kg m^{-3} density difference criterion was adopted, which is stated in the caption of Table 3 in the original version. This sentence is now moved to the Methods section as:

"Z_{ml} denotes the upper mixed-layer depth (UMLD, m), which was calculated using the 0.05 kg m^{-3} density difference criterion (Table 3), ···".

- The authors calculated DMS photolysis as the water-column integral straight away (Eq. 5 and 6), thus assuming that all the photons capable of promoting DMS photolysis were absorbed in the UMLD. However, they do not state clearly that the photolysis-causing wavelengths were extinguished, e.g., to < 1% of subsurface irradiance within the UML at all stations. There is only a vague sentence in P2101, L14–15. Even though spectral Kd's in the water column are not reported, they can be approximated from inherent optical properties that do appear in the paper. I used the data in Fig. 3 to make some calculations, which indicate that the **authors' assumption was correct (see specific comment on P2101)**.

  Please see our response to specific comment 6.

- In addition, the authors claim that previous studies were wrong in the way they calculated the amount of spectral irradiance absorbed by CDOM (P2108), an affirmation I do not agree with (**see detailed argumentation in the specific comments**). This should be corrected.

Please see our response to specific comment 13.

3. Non-technical aspects: I suggest that the authors revise (or reduce) the use of acronyms referred to geographical locations (MRE, MS, CB, SGT-E and SGT-W, etc). The acronyms are not always used in a consistent manner **(see specific comments)**. Replacing some of the acronyms by full words or by more explicit "labels" would help the reader.

We agree with the reviewer and made revisions accordingly. Please also see our response to specific comments 9 and 21.

- I also suggest that the authors add a sentence in both the abstract and the summary that conveys the relevance of their findings to the broad audience of Biogeosciences. Although this is speculation, I believe that the methodology used by Taalba et al. to study the relationship between CDOM quality/origin and AQYs might provide results of global significance in the future, and might have implications for the photochemical cycling of compounds other than DMS.

This is a very good point in view of the recent publication by Fichot et al. (2013) showing that the spectral slope S275-295 can be estimated from ocean-color remote sensing. A sentence is added to both the abstract and summary.
In the Abstract: "The methodology adopted here to study the relationship between CDOM quality/origin and DMS AQYs, if applicable to other ocean areas, may bring results of global significance for DMS cycling and might have implications for probing other CDOM-driven photochemical processes."

In the Summary: "This approach of linking DMS AQYs to CDOM quality/origin, if confirmed elsewhere, may lead to broad implications for studying photochemical cycling of DMS and perhaps other compounds as well.".

Moreover, we briefly discussed this point in the Results and Discussion: "The quantitative relationship between Φ^{*}_{dms} *and S275-295 identified here points to the possibility of using remote sensing to study DMS photolysis, since S275-295 can be retrieved from satellite-based ocean color data (Fichot et al., 2013)."*

Specific comments

1- Abstract L5: only in the Arctic? L10 after "salinity,...": in my view, no need to describe the mathematical function. It has no obvious interpretation! Perhaps saying "exponential form" is enough (after checking for temperature effects, general comment 1).

L5- changed to "The role of photolysis in regulating the DMS dynamics in northern polar seas remains, however, less clear" (obviously because few studies have been conducted in this region).

L10- agreed and changed accordingly.

2- Introduction P2095 L2: "... POTENTIALLY contributing to the formation of cloud..." L2– 6: A relevant work that might be cited here is "Direct Observations of Atmospheric Aerosol Nucleation, Markku Kulmala et al. Science 339, 943 (2013); DOI: 10.1126/science.1227385. Check out also "Potential for a biogenic influence on cloud microphysics over the ocean: a correlation study with satellite-derived data, A. Lana et al., Atmos.Chem. Phys., 12, 7977–7993, 2012".

Revised according to reviewer's suggestions.

3- P2096 L19 – 21: Please consider adding here a reference that documents the freshening trend in the Arctic.

The following reference is added:

Rabe, B., Karcher, M., Schauer, U., Toole, J. M., Krishfield, R. A., Pisarev, S., Kauker, F., Gerdes, R., Kikuchi, T.: An assessment of Arctic Ocean freshwater content changes from the 1990s to the 2006–2008 period, Deep-Sea Res. Part I, 58, 173–185, 2011.

4- P2097 L3: Please add something like "under shallow stratification conditions" after "summer". According to Galí et al., bacterial DMS consumption would largely dominate in stations with UMLD > 10 m.

Revised according to reviewer's suggestion.

5- Materials and methods P2099 – 2100 Equations 1, 2 and 3 (and corresponding text): I understand that "l" is the pathlength of the cell used for measuring CDOM absorbance and "L" the pathlength of the irradiation cell. However, I find these symbols a bit confusing at first sight (for instance "L" reminds me of "liter" units). Why not using "r" and "R"?

Revised according to reviewer's suggestion.

6- P2101 L14 – 15: The euphotic layer depth (which I assume is defined as the 1% penetration of PAR) is not really relevant here, given that shortwave UVA accounts for the majority of DMS photolysis. Maybe the authors mean, by saying that, that the surface waters displayed very low transparency to UV and PAR (?); so, despite the very shallow stratification encountered, the UMLD was exposed to a small fraction of subsurface PAR and even a smaller fraction of subsurface UVB and UVA.
If we calculate the total absorption coefficient (a_t) from the a_CDOM (Fig. 3B) and the quotient a_CDOM/a_t (Fig. 3C), we obtain an a_t (at 330 nm) of ~ 1.5 m-1 at Sta. 640 and 691, and ~ 25 m-1 at Sta. 697. If we assimilate a_t to the Kd (which is not exact) we obtain a 1% penetration depth for 330 nm radiation of ~ 3 m at Sta. 640-691 and ~ 0.2 m at Sta. 697. This indicates that at these three stations photochemically active radiation was absorbed at depths shallower than the UMLD. Indeed, the large absorption coefficients of CDOM at 412 nm suggest that a similar situation occurred at most stations.
In my view, this issue should be explained in more detail (perhaps in the Results section), because the estuarine and shelf waters studied seem to be very different (at least optically) from most open ocean surface waters. I suggest using the concept of "photoactive layer" (sensu Neale et al. 2003, book chapter) rather than "euphotic layer" in this context.

A very good point. We now adopted the photoactive layer concept, added the 1% penetration depth for radiation at 330 nm to Table 3, and discussed this issue in the Results and Discussion as follows:

"The photochemical turnover rate constants (kdms) in the surface mixed layer can be estimated from Eq. 6. Note that this equation requires that the depth of the photoactive layer, defined herein as the 1% penetration depth for the radiation at 330 nm ($Z_{1\%,330}$, m), was shallower than the UMLD so that the most photochemically active radiation was absorbed within the upper mixed layer. We estimated $Z_{1\%,330}$ from $a_{t,330}$, which is an underestimate of the diffusive attenuation coefficient at 330 nm due to the exclusion of the scattering coefficients of particles and water. The obtained $Z_{1\%,330}$ was always shallower than the UMLD except for three offshore stations (126, 320, 391) at which the $Z_{1\%,330}$ was slightly deeper than the UMLD (Table 3)."

7- P2102 L8 – 9: "The exponent in Eq. (5) is the photolysis rate constant..." in fact the photolysis k in Eq. 5 is only the part in parentheses that has (time)-1 units.

Now changed to "The term within the parentheses in Eq. 5 is the photolysis rate constant…"

8- L11 – 14: The authors should be more circumspect about some of these assumptions, which may have an impact on the results. In case the authors measured DMS in vertical profiles, they should mention those profiles in the text to support the 'homogeneity assumption'. See general comments.

DMS profiles were not collected. This assumption is considered valid based on a relatively fine mixing criterion adopted in this study (see response to general comment#2).

9- Results and discussion P2102 L17: Naming the stations after the name of the project alone may be confusing for the reader. Please consider using explicit names whenever possible.

We agree and did the recommended changes.

10- P2103 Is there a particular reason to use aCDOM at 412 nm? I would find more useful that the authors reported aCDOM at 330 nm, which is the peak DMS photolysis wavelength (at the water subsurface). Yet, I can assume that there is, roughly, an exponential decrease of aCDOM with increasing wavelength, so that aCDOM 412 is a good predictor of aCDOM 330.
This raises another concern regarding CDOM measurement. On one hand, aCDOM at 412 nm is typically very low in oceanic ("case 1") waters, so that aCDOM,412 is less sensitive and more error-prone than aCDOM,330 (this may be relevant for Canadian Basin and Baffin Bay stations, Fig. 2). On the other hand, the Lambert-Beer law may not hold when a solution is optically thick (absorbance > 1) and very turbid (which may have happened in samples like Sta. 697). In these cases it may be better to measure aCDOM using 1 cm cuvettes. In fact, the aCDOM,300 at Sta. 697 was ~ 20 m-1. Using Eq. 1 this corresponds to an absorbance of ~ 0.87 in a 0.1 m cuvette. Absorbance was likely higher than 1 at wavelengths < 300 nm, which might have affected the calculation of the S275_295.
Can the authors provide a convincing explanation on these issues?

There are two reasons to use aCDOM at 412 nm here: 1) the 412-nm wavelength is more relevant for remote-sensing, 2) (partly because of #1) recent CDOM-related studies in the same area all reported aCDOM at 412 nm (e.g. Bélanger et al., 2008; Matsuoka et al., 2012; Xie et al, 2012; Song et al., 2013). To facilitate comparison among different studies and explore potential remote-sensing applications by other researchers, we chose 412 nm instead of a UV wavelength, which is less or not relevant to ocean-color imaging, for ***discussing the CDOM dynamics****. We agree that aCDOM,412 is less sensitive than aCDOM,330 for measurement. However, all aCDOM,412 values in the present study (even those for the Canada Basin and Baffin Bay) are well above the instrument's lower detection limit (0.02 m^{-1}, this information is now added to the Methods). We also agree that aCDOM,313 is more relevant to DMS photolysis. That's why we presented the aCDOM/at ratio at 330 nm (table 3). We now added aCDOM,330 to this table as well.*

The absorbances of the highly colored samples from the estuary were actually determined with a 5-cm cell, which was somehow omitted in the original paper. We now added it to the new version:
"For highly colored samples in the inner part of the Mackenzie estuary, a 5-cm quartz cuvette was used to ensure that the measured absorbances fell within the upper limit of the Lambert-Beer law (i.e. ~1)."

The highest absorbances determined were those from stations 697 and S0, they both being ca. 0.56 at 275 nm or 26 m^{-1} in terms of absorption coefficient. The absorbance was thus well below the upper limit of the Lambert-Beer law. Even if a 10-cm cell used, the absorbance at 275 nm (the shortest wavelength used in this study) for these two stations was barely >1 (for all other stations the absorbances were far below 1).

11- P2103 L1 and 25: It seems there is a typo here ("MatsOUka" instead of "MatsUOka"). The work by "Matsuoka et al." does not appear in the reference list. Please check throughout.

The typo is corrected.

12- P2107 L2: Please add "LIKELY" between "anion" and "plays" in that sentence.

Revised according to reviewer's suggestion.

13- P2108 L16 – 20:
This sentence seems to imply that previous studies had an important bias: "Note that previous studies often implicitly assumed all underwater light to be absorbed by dissolved chromophores without considering the particulate colored materials, which biased the kdms estimates upward to a certain extent, depending on the fraction of light absorbed by particles".

I do not agree with the statement that previous studies assumed that all the photolysis causing radiation was absorbed by CDOM. Rather, they just assumed that ONLY the radiation absorbed by CDOM was causing DMS photolysis. The confusion here probbaly stems from the fact that previous studies used a different formulation to calculate photolysis rates in the water column, because they did not assume that all the photons were absorbed within the UML.

See, for instance, Eq. 7 in Toole et al. (2003). There, the amount of photons absorbed at depth z results from the product of scalar irradiance (Eo) and aCDOM. Eo at depth z is calculated from Eo at the subsurface (Eo-) and an exponential attenuation factor [exp(Kd*z)]. If we integrate that equation over a depth interval (z1 - z2), we get that UML-integrated Eo and thus photolysis is proportional to (1/Kd)*[1 – exp(-Kd*z2)].

The case where all photons of a given wavelength are absorbed within the UML is equivalent to setting [exp(-Kd*z2)] to zero. In this case, photolysis is proportional to (1/Kd) alone. At this point, it can be noted that both Kd and a_t have the same units (m-1), and are closely related variables (although the first is an apparent optical property, deduced from the light field, and the second is an inherent optical property). Now, note that a_t divides a_CDOM in Eq. 5 and 6, whereas Kd will divide a_CDOM once Eq. 7 from Toole is integrated with respect to z. So, the formulation of Toole et al. (2003) and that used in the present study seem to be dimensionally consistent and roughly equivalent when photochemically active photons are completely absorbed within an optically-homogeneous UMLD.

While the formulation used by Taalba and coauthors does not account for scattering (and perhaps does not need so), the formulation of Toole et al accounts for the tridimensional light field (mostly due to scattering) by using scalar irradiance (Eo). Furthermore, note that in case 1 waters (oceanic waters where most of the previous studies were done) very little light absorption is expected in the UV and blue light range responsible for DMS photolysis, as shown in Fig. 3C for Sta. 691-640. In particular, a_CDOM will frequently exceed a_particles by around 1 order of magnitude or more.

We agree with the reviewer. We misunderstood the relevant papers on this point. We now removed this statement. Our formulation does not account for scattering, since we focused on the depth-integrated quantity not photolysis at a specific depth. On a depth-integrated basis, only the downwelling irradiance matters (if the often minor upwelling irradiance is neglected) since scattering only changes the direction of light; it essentially does not alter the net amount of irradiance the water column receives.

We now included the ap/aCDOM ratio as one of the factors to explain the difference in k_{dms} between the present and previous studies:
"Moreover, while previous studies mainly surveyed clear, open-ocean waters in which CDOM dominates the absorption of UV radiation, the present study sampled waters within the Mackenzie River plume containing sizable amounts of particles which accounted for up to 30% to 65% of the total light absorption at 330 nm (Table 3). The strong competition for light by particles led to lower k_{dms} (Eq. 6)."

14- P2108 L25 – 27: In my view this assumption tends to underestimate/overestimate the photolysis rates, not k_DMS. According to Eq. 6, k_DMS is independent of [DMS] in this approach.

We agree and removed this sentence.

15- Summary P2110 L9 – 10: Yes, but check the comparison with other studies after accounting for temperature kinetic effects.

Please see response to general comment#1.

16- References please check for typos and possible formatting issues. Examples:
P2111 L16: "natural sourCes" L19: "Global Biogeochem. Cy,,"
P2112 L1: "DelValle" should be "Del Valle".

Thanks. Corrections made.

17- Tables and figures Table 1: Why not reporting the approximate depth range sampled? For instance, 0 – 0.5 m. Sampling depth = 0 m does not seem very appropriate.

Now changed to 0-0.2 m (the depth of the bucket is ~20 cm)

18- Table 3: I recommend that, besides the (model-derived) daily quantum irradiance in the 290 – 500 nm band (that causing DMS photolysis), the authors report here the total shortwave irradiance ('pyranometer irradiance', in W m-2 units) and even the UVR and PAR irradiance. This will (1) facilitate that expert readers understand the overall irradiance climate at a glimpse, (2) make the information easier to understand by the non-expert audience, and (3) allow the comparison with other studies where only total shortwave irradiance was reported.

A good point. We do not have the shortwave irradiance but PAR and UVR are now added.

19- Table 4: I suggest changing the column "Layer", which currently adds very little information, and reporting there the UMLD range in each of the studies. This would allow a better understanding of the differences among sites, that is, how much the difference in k_DMS might have been due to mixing-regulated solar exposure or to differences in quantum yields. Ideally, some information on underwater light attenuation (UV/PAR Kd) should be added too, but this information might be hard to find in some studies. In the few studies where a fixed depth horizon of 0 – 20 m was used for computations (Brugger, Hatton and Deal), a note could be added saying that the depth interval did not match the UMLD.

We now added the ranges of UMLD. As many studies do not report Kd, this parameter is not added.

20- Fig. 1: Please add "Beaufort Sea" somewhere in the map or in the caption, because it is mentioned in Fig. 2.

Now added.

21- Fig. 2: According to the current nomenclature it seems that the CFL-BS samples (Beaufort Sea) were not located in the Canadian Basin (CB). Is there a reason to give these samples a distinct nomenclature?

The caption in the original version indicates SE BS = southeastern Beaufort Sea (including both coastal and open ocean waters). To make it clearer, now changed to SE BS = southeastern Beaufort Sea, including the Mackenzie River estuary, Mackenzie Shelf, and Canada Basin. The purpose of using this nomenclature is to shorten the symbol legend embedded in the figure.

22- Fig. 4: add salinity below the x-axis of the bottom panel. If my concern about the temperature correction was true (see general comments), this figure should be modified. I suggest adding to each panel a twin panel with the temperature-corrected AQYs to see the real variability captured by salinity.

Revised according to the reviewer's suggestions. Also please see response to general comment#1.

Response to reviewer#2

General comments

1. In addition to being concentration dependent, the AQY of DMS is temperature dependent. It isn't clear how or if the authors took the temperature dependence into account when comparing AQYs from different studies (i.e., Deal et al. 2005; Bouillon and Miller 2004; Toole et al. 2003). I am not sure how much it matters for the relationship between salinity and AQY (i.e., Figure 4), but the temperature corrected AQY can be calculated using the temperature dependence determined by Toole et al. (2003).

   Please see our responses to reviewer#1's general comment#1 and specific comment#22.

2. My understanding is that scalar irradiance accounts for light from all directions. Why was scalar irradiance not used in this study?

   Yes, scalar irradiance accounts for light from all directions (i.e. taking into scattering), it is thus used for estimating photochemical rates at a specific depth. However, the purpose of our study was to estimate the k_{dms} in the upper mixed layer on a depth-integrated basis. As almost all the UV radiation was absorbed within the upper mixed layer (see our response to reviewer#1's specific comment#6), only the downwelling irradiance matters in our case (by neglecting the often minor upwelling irradiance). This is because scattering changes the direction of light but essentially does not change net amount of the irradiance that the water column receives.

3. By my definition the seasonally ice-covered Bering Sea is included in the Arctic. Arctic sea ice clearly extends well into the Bering Sea. Also, the Bering Sea is north of the July isotherm of 10°C for the warmest month of the year, which is a common definition used for Arctic waters

   We agree with the reviewer on the definition of Arctic waters and now considered the Bering Sea as part of the Arctic Ocean.

4. Please write out the abbreviations for MRE, CB, MS etc. instead of using the abbreviations throughout the text. This would make the manuscript easier to read.

   Revised according to reviewer's suggestion.

Specific comments

Introduction

23- Page 2095 line 3: "CCN increases cloud albedos: : :". What is it about CCN that increases cloud albedos? I thought all clouds had CCN.

To make it clearer, now changed to "CNN increase cloud droplet number and hence cloud albedo, thereby reducing solar radiation arriving at the earth's surface"

24- Page 2097 lines 4-7: This sentence, in particular following on the preceding one, gives the impression that these are the first measurements of the pseudo-first-order AQY in the Arctic.

The Arctic marine environment includes the seasonal ice zone of the Bering Sea where at least one previous measurement of the pseudo-first-order AQY has been made. Perhaps "Canadian" should be placed before "Arctic" in line 6 or omit "Arctic" in line 6 for clarification.

See our response to general comment#3. Now added Deal et al.'s (2205) contribution to this paragraph and revised the statement to:

"···. Deal et al. (2005) measured the pseudo-first-order apparent quantum yield (AQY) spectra of DMS photolysis in seawater from two Bering Sea stations. Their results indicate that the loss of DMS through photolysis is comparable to that by air-sea exchange in the Bering Sea in late summer. To our knowledge, these are to date the only published surveys of DMS photooxidation in the Arctic Ocean. Here we report the first measurements of the pseudo-first-order AQY spectra of DMS photolysis in Canadian Arctic marine environments covering the Mackenzie estuary and shelf, Canada Basin, and Baffin Bay. ···"

Results and discussion

25- Page 2105 line 3: UVA also dominated DMS photolysis in the Bering Sea (Deal et al. 2005). To be complete, I think this reference should be included here as well. However Bering Sea waters would not be considered "warmer waters".

Now changed to "UV-A thus dominated DMS photolysis both at the surface and in the entire water column, which is consistent with the results reported for warmer waters in the Pacific and Atlantic Oceans (Kieber et al., 1996; Toole et al., 2003, 2004; Bouillon et al., 2006) and polar waters in the Bearing Sea (Deal et al., 2005)."

26- Page 2107 line 20: Is "titled" the correct word here?

It should be "tilted". Corrected.

27- Page 2108 lines 17-18: From what I can tell, previous studies do not often implicitly assume that all underwater light was absorbed by dissolved chromophores. Your statement that previous studies do make this implicit assumption needs to be corrected or clarified. Because the chromophores involved in DMS photolysis are not known previous studies have used total absorption by CDOM as a proxy for reactant absorption.

Please see our response to Reviewer#1's specific comment#13.

References

28- Page 2114: Matsuoka et al. (2012) is cited in text but not included in references.

Now added to the references.

Tables and figures

29- Table 1: It would be more appropriate to replace the nitrate concentrations of 0.00 with "<" and the detection limit or an abbreviation (e.g., < DL) and state detection limit in the caption.

Revised according to reviewer's suggestion.

30- Table 2 caption: Should be "in an irradiation cell" or "in irradiation cells", not "in an irradiation cells".

Correct. Correction made.

31- Figure 4 caption second line: It seems like "(panel" should be deleted.

Correction made.