Referee#1

My basic concern is that authors based all discussion on the surfactants enrichments in the SML, and not on the real concentrations. It ismknown that quantity as well quality of surface films effects gas exchange. Therefore, it would be also important to present global maps for SML coverage with absolute surfactant concentrations. Namely, even high enrichment in a case of low bulk surfactant concentration (like oligotrophic oceans) would come also to low surfactant concentration in SML. And opposite, in coastal productive regions low EF still may be obtained for very high surfactant concentrations in SML.	Traditionally the SML is defined as an enrichment of parameters of interest. We have used enrichments of natural surfactants as an indicator for the presence of the SML and its intensity relative to bulk water phase. Absolute concentrations without a reference point would not be applicable to achieve the objective of this study, which is to provide a first estimate of the ocean's coverage with SML. We agree with the referee's comment that absolute concentrations are important for the assessment of air-sea gas exchange rates. For this reason we examined concentrations of surfactants in the SML among the three categories of primary production as defined in section 2.6 of the manuscript. We found that the concentrations increases with increasing primary production, and included a new section 3.4 and Fig.7 to the revised manuscript to present the results. We have also included the additional Table 4 and 5 to present statistical data on the concentrations at different wind regimes and trophic states, and at the different study regions respectively. We have also revised section 4.4 ("Implications for air-sea gas exchange") in regards to SML concentrations found in this study. As we used the same categories of primary production ranges, the global maps (Fig.8 in revised manuscript) present both the enrichment and concentration ranges of surfactants in the SML. The caption of Fig.8 has been revised accordingly.
The title is not indicative on what the authors presented.	We agree, and suggest changing the title to "Formation and global distribution of sea-surface microlayers".
ABSTRACT I do not agree wit the sentence: Â´ nGlobal maps of primary production and wind speed are used to estimate the ocean's SML coverageÂ˙ z as the authors presented surfactants enrichments and not concentrations.	We agree. We have changed the sentence to "Using our observations of the surfactant enrichments at various trophic levels and wind states, global maps of primary production and wind speed allow us to extrapolate the ocean's SML coverage".
INTRODUCTION Page 5721, line 5: reference is missing. I would suggest: Plavšić,M., Ćosović, B. 2000. Adsorption properties of different polysaccharides on mercury in sodium chloride solutions solutions.	We agree, and we have included the suggested reference to the revised manuscript.

Electroanalysis 12, 895-900.	
METHODS Page 5723, lines 1-4: I suggest to remove: "collected by a withdrawal rate of 5–6cm s 1," as it is repetition; to move: "as consistently as conditions allowed" to the first line. Reference Carlson, 1982 is unnecessary.	We agree. We have changed the sentence to "The slower withdrawal rate collects the SML with a thickness of about 50 μm (Carlson, 1982), which is consistent with experimentally determined SML thicknesses of 50±10 μm using pH microelectrodes (Zhang et al., 2003).". We would like to keep the reference of Carlson (1982) as he suggested to use a slower rate of withdrawal to collects thinner layers.
Page 5724, line 16: Please add reference after "the standard addition method." as this approach is not used in Ćosović and Vojvodić (1998). The authors should add much more details on how they have measured surfactants.	We agree. We have added the following description. "Surface-active substances were accumulated at the hanging mercury drop electrode at a potential of -0.6 V (versus an Ag/AgCl reference electrode) with stirring. Stirring time was in the range of 20 to 60 sec depending on the concentration. The frequency of a.c. voltage was 170Hz and the p–p amplitude was 10 mV. Scan rate was 20 mV s^{-1}. The out-of-phase signal of the alternating current was measured.". We have added "Sander and Henze (2005)" to provide a reference for the application of the standard addition technique.
Page 5724, lines 16-17: It is incorrectly written. If the precision is less than 10% that would mean that reproducibility was very bad.	Precision is equivalent to reproducibility. Earlier studies using the same electrochemical technique reported a typical reproducibility of 8-10% (for example B. Gašparović ,B. Ćosović, Estuarine, Coastal and Shelf Science 58 (2003) 555–566. We have changed "precision" to "reproducibility".
RESULTS Page 5728, lines 24-25: There is mistake in this sentence, probably in the value of EF "(i.e. EF>2.7)" given in line 25.	We changed the sentence as following for easier reading: "In addition, above bulk water concentrations of 600 μg Teq. L^{-1} we found only relatively low surfactant enrichments not exceeding 2.7 (Fig 5). "
Page 5728, line 27: I suggest removing last part of sentence: "that means under eutrophic conditions" as blooms are regularly happening also in the oligotrophic seas.	We agree. We have changed the sentence to "Bulk water surfactant concentrations typically exceed this level under bloom conditions.".
Page 5729, lines 11-13: I concern about the statement that higher SML enrichments led to	We agree. We have changed the sentence to "Overall, we found that low productivity and

enhanced formation of SML.	moderate to high wind speeds lead to higher enrichments of natural surfactants in the SML.".
Page 5729, line 26: I would suggest to add "surfactant enrichment" after The choosen..	We agree. We have changed the sentence to "The chosen threshold values of surfactant enrichments of 1.0, 1.5 and 2.0 are used to describe weakly, moderately and strongly SML-influenced ocean surfaces, respectively, and were obtained from the range of observed values.". This part of the text has been moved to section 2.7 in the revised manuscript.
DISCUSSION Page 5735, line 10: reference is needed after: microbial respiration.	This part has been removed from the original manuscript due to major revision of the discussion.
Page 5735, lines10-12: The authors based their discussion on domination of terrigenous OM in coastal SML, with what I do not agree for the reason of often high autochtonous OM production in coastal region.	We agree and this part has been removed from the original manuscript due to to major revision of the discussion.
Page 5735, lines12-16: This part is speculative as it is not supported by referencing proving different OM matter produced in the oceans and in coastal regions. Do really authors think that coastal SMS is really mainly of terrigenous origin and oceanic SML is of autochtonous origin.	We agree and this part has been removed from the original manuscript due to to major revision of the discussion.
In fact, whole section starting from page 5734, line 19 to page 5736, line 12 needs be rewritten to better clarify authors suggestion of this section.	We agree that is section needs clarification. We have revised this part of the discussion intensively. We have also considered the comments of referee #2 and #3 in the revised text of the discussions. Please see below for the revised section of discussions.
TABLES Table 1. If Â´ naÂ˙ z is number of non-slick samples, where is number of all samples?	We agree. We have accidentally reported the total numbers of samples in Table 1 of the original manuscript. We have revised Table 1 by changing the subscript to "total number of samples (number of non-slick samples are reported in Table 3).
Table 2. To remove word surfactant after: "Statistical data on", and also to add Â´ nand total dissolved carbohydrates (TDC)Â˙ z after: "of surfactants". Please put SAS and TDC in brackets in EF SAS and EF TDC. Correct all numbers to be with points instead of commas (same in Table 3). After CI add b as superscript, and add text like in Table 3: b Confidence interval of the mean Please uniform: sd or SD (lower or upper case)	We agree and did the changes accordingly. Our original table 2 had numbers with points and not comma. It seems the comma have been introduced during editorial work.

FIGURES Fig. 1. In the fig caption instead of LineP should be written NP, or maybe line NP.	We agree. We have changed the caption to "Fig.1 : Sampling stations. AC: Arctic; CA: California, Santa Barbara Channel; HI: Hawaii; NP: North Pacific; SI: Saanich Inlet; VI: Vancouver Island, West Coast."
Figs. 2, 3, 5 and 6: y-axe title should be designated as Surfactant or TDC to be in brackets or as subscript as it is in Fig. 4. Please make it uniform.	We agree. We have changed the y-axis title accordingly.
The colours in Figs. 4 and 5 for offshore and oceanic samples are too similar (blue and green). Maybe one of those may be black to improve visibility.	We agree. We have changed the color of symbol from green to black.
Fig. 5. Explanation for broken line is missing.	We agree. We have added the explanation of the broken line.
Fig. 6. In the fig caption replace rhombus symbol for PP with the symbol square, as it is in the fig.	In our original manuscript, the caption has the square symbol. It seems the change to the rhombus symbol occurred during editorial work.
Fig. 7. The fig caption is not good. I would suggest: Global maps of surfactants SML-enrichments.... In this case last sentence Â´ nPlease note that maps represent enrichment factors (EF) and not absolute SML concentrationÂ˙ z may be excluded. The fig is unclear. I would suggest enlarging it at least to the width of fig caption, to add black continent edges, to use darker yellow in the fig.	We agree. We have changed the figure captions to "Global maps of mean concentrations of surfactants in the SML (± 95% confidence interval of mean) and their enrichment factors (EF) for a) Apr-Jun, b) Jul-Sept, c) Oct-Dec and d) Jan-Mar. ■ 320 ± 66 µg Teq L^{-1}, EF > 2; ■ 502 ± 107 µg Teq L^{-1}, 2 > EF > 1.5; ■ 663 ± 77 µg Teq L^{-1}, 1.5 > EF > 1.0; ■ no enrichment." We also have improved the quality of the figure itself.
Technical corrections	Without referring to each comment, we agree to all the suggested corrections, except Line 5720, Line 20. The SML is very thin and writing "Despite its thickness of the SML" may be confusing as the unique feature of the SML is its thinness. The manuscript has been revised accordingly.

Referee #2

There are no relationships that would make it possible to clearly derive the intensity of enrichment in the SML. Primary production was used to distinguish the three trophic states (eu-, meso- and oligotrophic) and wind speed was used as a threshold value above which the SML is likely to be broken up	We agree that to derive clearly the intensity of enrichment in the SML our data set is too limited. We have highlighted it in the revised discussion. For this reason we present a more approximate approach categorizing the enrichments in the three major trophic states. We showed that the enrichment between the

continuously. Global mapping of the SML seemed to me the major goal of study, however, I wonder whether the current dataset allows for such global extrapolation. Furthermore I completely agree with anonymous reviewer #1 (for the reason he invokes) that is important to report concentrations of the surfactants rather than presenting enrichment factors only.	three trophic states are statistically significant different. Our study includes data from all climate zones and trophic states. Despite the approximation, our study is the first presenting global mapping of the SML based on experimental data as an initial step towards a better awareness of the microlayer to the science community. Our mapping is an improvement compared to an earlier attempt by Tsai and Lui (2003) using an arbitrary set threshold of primary production above which SML-coverage has been defined. We have included results on the concentrations of surfactants in the SML including mapping (section 3.4 and 4.4; Table 4 and 5; and Fig.8 in the revised manuscript). See our response to the referee's #1 comment.
In general the text should be more focused and particularly the discussion should be extensively edited and shortened. There is much in the discussion that does not directly refer to the data measured in the current study.	We agree that the discussion needs clarification. We have revised this part of the discussion intensively (please see below for revised text). We have also considered the comments of referee #1 and #3 in the revised text of the discussions.
Page 5721; line 14-15: If the authors make a statement like this, then I would expect them to be more specific on what the arbitrary thresholds where and why they are considered arbitrary.	We agree. We have changed this part of the manuscript as following: "However, the authors chose arbitrary threshold values (i.e., not derived from experimental data) of local primary production as an indicator for the presence of surfactant films. The threshold values of two different scenarios were 0.50 and 0.83 g C m^{-2} day^{-1}, and regions above those thresholds have been defined as being influenced by surface films (i.e., SML) (Tsai and Lui, 2003)."
Page 5721; line 28-29: For me, this an overstatement as the dataset used is mainly restricted to sites near North America with one study at Hawaii.	We have collected samples from all climate zones (tropical, temperate and polar) and additionally over a whole seasonal cycle at a selected site. Therefore we believe that our dataset is representative in regards of the objective of this study. We would like to highlight again, that we did not attempt to

	provide global maps to quantitatively assess SML effects, but to show the wide distribution of the SML in more general terms.
Page 5722; paragraph 1: To be able to judge the trophic situation by the reader, I suggest to introduce the different sampling sites more thoroughly or at least provide links or citations where information can be found on e.g. latitude /longitude, major nutrient concentrations etc.	We agree. Citations to the sampling areas/regions are provided in Table 1. We have changed the following sentence to "All sampling areas are summarized in Table 1 with further references to provide details of the sampling regions. ".
Page 5723, paragraph 1: Is there any special reason why the authors deviated from the sampling scheme of Harvey and Burzell? I assume it is because of Zhang et al., however, as written this is not that clear.	The modification of a slower withdrawal rate has been introduced by Carlson (1982) to collect thinner layers. Through the measurement of the actual SML thickness (Zhang's et al, 2003) it has been shown that the modification is valid. We have changed the following sentence for clarification:

"The slower withdrawal rate collects the SML with a thickness of about 50 μm (Carlson, 1982), which is consistent with experimentally determined SML thicknesses of 50±10 μm using pH microelectrodes (Zhang et al., 2003)." |
| Page 5724, line 1: In general I suggest to check that abbreviations in the main body of the text are properly introduced on first use (here for example CA or HI). | We agree. We have checked the text for proper introduction of abbreviations. |
| Page 5724, paragraph 1: I can only assume that the chemical measurements including Chl a were done for all stations. This should be more explicitly pointed out. How much volume was used for the different analysis? | We agree. We have added the following sentence in the beginning of section 2.4: "We have measured the concentration of surfactants, TDC and chlorophyll at all stations."

We have added additional information on sample volumes as following:

Surfactants:

"...with a hanging mercury drop electrode in unfiltered samples (10 mL) according to Ćosović and Vojvodić (1998)." |

	Total dissolved carbohydrates (TDC):
	"Prior to analysis, the samples (4 mL) were subjected to hydrolysis ..."
	Chlorophyll-a (Chl-a):
	"For Chl-a determination 200-1000 mL of bulk water (1 m and 8 m) were filtered in duplicates…"
Page 5726, line 9-11: I don't think it is necessary to explain the categorization of the different wind states in such detail. It is not used anyway as far as I could see.	We think the information on how we define the wind regimes is important for future studies following a similar approach, and therefore we prefer to keep the information as provided.
The results are confusing to me. I think with better tables (indications of which parameters where significant, more detailed captions of parameters presented) and more direct description of the tables and figures this could be improved. The final global maps are divided by seasons, however, seasonality was not clearly presented in the results section.	We agree. We have revised Table 2 for better reading. However, we have not added indications of significance to the tables to keep them illustrative. We have revised the description of the results, including indication of significance, for an easier reading, and we do not think an additional table is required to indicate which parameters differs significantly.
	The seasonality of the global maps are based on the primary production. We have obtained seasonality of total dissolved carbohydrates (major products of phytoplankton exudates) at Saanich Inlet, but not for surfactants as this parameter may include other sources. We explain this observation in section 3.3. Unfortunately, we did not have the opportunity to study seasonality at an oceanic site.
Page 5727; paragraph 1: This paragraph does not adequately describe why slick samples have been excluded from the analysis. If the mean enrichment is not significantly higher why are they excluded? How many slicks did the authors encounter? I miss references to data in tables of figures and there is no description/presentation of the frequency of rainfall events e.g.	We agree. We have changed this paragraph intensively providing more details on slick conditions and why we have excluded those samples from our analysis. We have added the number of slick samples (60) to the section 2. We plan to prepare a separate manuscript presenting data on slick formation due to its special features.
Page 5727; line 16: I am not familiar with the Teq unit. I suggest that the authors include a	The unit Teq has been defined in section 2.4 as "The total concentration of surfactants is

brief description of different Teq units for different oceanic environments to give the reader a feeling for the numbers– this might be done in the methods or in the discussion.	expressed as the equivalent concentration of the non-ionic surfactant Triton X-100 (Teq), which mimics effects similar to natural surfactants.". Basically it means that surfactant concentration has been calibrated with the model substance Triton X-100.
Page 5727; line 28: I suggest introducing the relation between phytoplankton and surfactants in the introduction more detailed, but not in several places as is the case for the current draft.	We agree to provide more information on the relation between phytoplankton and surfactants to the introduction; and have deleted the page 5727, Line 28 from the revised manuscript. We have added following text to the introduction: "High molecular polysaccharides and complex β-glucans are major excretion productions of marine phytoplankton (Allan et al., 1972; Mykelstad, 1974) and frequently found in seawater during blooms (Sakugawa and Handa, 1985). The hydrophilic polysaccharides may conjugate to hydrophobic moieties, making them surface active, and become enriched in the SML by factors of up to 3 compared to subsurface waters (Williams et al., 1986; Wurl and Holmes, 2008; Wurl et al., 2009).".
Page 5729; line 15 onwards: There is hardly any data presented in this section. The description of how the global mapping was done should go into the methods section and should be mainly discussed in the discussion section. I would delete the paragraphs on global mapping in the results.	We agree. We have moved part of this paragraph to a new section 2.7 to describe how we have extrapolated the maps.
Page 5731; line 1: This counter argument is quite weak considering that the current dataset is biased to regions north of 30N and there is no data presented to the south of the equator.	We understand that our discussion based on the assumption that enrichment processes of organic matter in the SML is similar among the three trophic states. As phytoplankton exudates are the major sources of natural surfactants, we believe that the assumption should be valid for an approximation of the global SML coverage. For example we have included data of oligotrophic conditions from the tropics (Hawaii) and polar (Arctic) regions. We have highlighted

	this in the sections 3.3 and 4.
	We included the following sentence to section 3.3: "On the other hand, the difference in surfactant enrichments was insignificant between tropical (HI) and polar (AC) regions, both characterized by oligotrophic conditions indicating that surfactant enrichment was independent of location."
I agree with anonymous reviewer #1 that the discussion should be extensively edited but also shortened. Much of what is there now, is more a review of other papers than a discussion of the presented data and findings. E.g. the paragraph on UV (Page 5734) and the 'discussion' on TEP seem out of place. At least it was not obvious to me what point the authors tried to make here.	We agree that the discussion needs clarification. We have revised this part of the discussion intensively. We have also considered the comments of referee #1 and #3 in the revised text of the discussions. Please see below for the revised discussion on enrichment at different trophic states.
Page 5738, line 7: Is there any explanation for the counter intuitive trend between surfactant enrichment in the SML and primary production?	That is part of the revised part of the discussion. Please see below
Page 5739, line 8: In the light of the presented data the conclusion is an overstatement.	We agree that further studies are required to confirm our statement. However, our data show that the SML exist at typical oceanic conditions, that means under typical oceanic wind regimes and oligotrophic conditions. In earlier studies (as cited in the manuscript) it has been shown that the presence of SML affect air-sea gas exchange and the dynamic of the carbon cycle. For this reasons, we think our study indicates the potential of the SML to affect air-sea gas exchange and biogeochemical cycles on a global context. We have changed the sentence as following: "Our study shows that the SML exists under typical oceanic conditions, that is, under typical wind regimes and oligotrophic conditions. We therefore suggest that the potential impact of the SML on air-sea gas exchange and biogeochemical cycling may be of global significance.".

Referee #3

One important basis for this study is the assumption that surfactant production is mainly driven by phytoplankton. The manuscript would clearly benefit if the authors would discuss potential surfactant production by (non-chlorphyll-a containing) bacteria as well as data on the relationship between absolute SAS / TDC concentrations and concentrations of chlorophyll-a, showing that these assumptions – although intuitive - are reasonable (especially in section 3.3 enrichment versus PP).	We agree. We have included discussion on potential surfactant production by non-chlorophyll containing bacteria in section 4.2. We could not find a clear relationship between the enrichment of SAS and Chl-a/primary production. For this reason we used the more general observation that oligotrophic waters are higher enriched than more productive waters. Using this approach provides a more approximate estimate of the ocean's coverage with the SML, but we think it is a useful approach as multiple processes (as discussed in the revised manuscript) controls the enrichment and a simple relationship is unlikely to exist. We have included that further in the revised manuscript. We have included the following sentence in Section 3.3: "Regression analysis, on the other hand, did not show a significant relationship between the enrichment of surfactants or TDC and the primary production, but the following supports our more general observation of diminishing surfactant enrichment with increasing primary productivity.".
The structure and the language of the manuscript are precise, except for the term "surfactants" as it is sometimes unclear whether this refers to SAS only or SAS + TDC. It would help to use precise terms or abbreviations throughout the manuscript as they are defined in the methods-section. Furthermore, most of the figures refer to surfactants, i.e. SAS and not SAS + TDC?!	We agree. We have replaced the term SAS with "surfactants" in the revised manuscript including tables and figures.
The manuscript would benefit from comparing results of TDC and SAS in more detail and discussing their relation also in terms of the importance of dissolved (TDC) versus total (SAS) surfactants in the SML.	We have compared the results of surfactants and TDC in Table 2, and mentioned that a seasonal trend of surfactants enrichment could not found as for TDC enrichments due to different sources (end of section 3.3). A more

	thorough discussion would be certainly interesting, but would somehow not contribute much to the objective of the manuscript. We present TDC data to show readers that the enrichment of some DOM may be indeed follow seasonal trend.
	We plan to prepare another manuscript dealing with slick formation, and discussions on the relationship between the enrichment of surfactants and TDC can be better included there as the relationship may differ between non-slick and slick surfaces.
P5720, L15 The term 'being affected' is unclear (in which way, direction, implication?).	We agree. We have changed the sentence to The maps indicate that wide regions of the Pacific and Atlantic Oceans between 30◦N and 30◦S are more significantly covered with SML than northern of 30◦ N and southern of 30◦ S due to higher productivity (spring/summer blooms) and wind speeds exceeding 12ms−1 respectively.".
P5720, L21 This range of SML thickness seems to be somewhat arbitrary, especially as it is most often technically determined by the SML samplers.	We agree. It is challenging to define the exact thickness of the SML as it depends on wave states, features of interests and depending on sampler device. We have included detailed information on the definition of the SML and its thickness as following: "The SML can be summarized as being a micro-habitat with a total thickness of between 1 and 1000 μm (Liss and Duce, 1997). Based on the literature, a SML of a thickness of 60 μm could be meaningfully used for studying the physicochemical properties of the SML (Zhang et al., 2003) and up to 1000 μm for biological properties, depending the organism or ecological features of interest. Despite the thinness of the SML,this interface may…"
P5724, L3-5 How much later were the cruise samples analysed?	We agree. Samples have been stored for up to 6 weeks. We have included this information in the following sentence.

	"On those cruises, unfiltered sub-samples were preserved with 1% formalin (final concentration) and stored at 4 °C for up to 6 weeks until analysis in the home laboratory."
P5725, L8 Please specify from which depth (1 or 8 m) these chl-a concentration were derived.	We agree. The chl-a concentration at 1 m depth have been used. We have included this information in the following sentence. "…with C_{Chl} being the measured surface concentrations of Chl-a (μg L^{-1}) at 1 m depth, D_{irr} the…"
P5727, L6 An increasing enrichment of SAS/TDC after rainfall should be another example of SML import driven by bubbles. However, the 'elevated levels of particulate loads' are unclear. Please provide data.	We agree, although we do not have data on precipitation rates before or during sampling events. However, we have changed the following sentence for clarification: "Slick samples were not consistently enriched in the presence of elevated surfactant concentrations in the bulk water during bloom conditions, but high surfactant enrichment was observed after rainfall events (maximum enrichment of 16), when wet deposition provided high loads of particulate matter to the SML, as visually observed."
P5727, L7 Romano et al (1991 EstuarCoastShelfSci, 1996 DeepSeaRes) reported quite frequent observation of slicks, at least in coastal regions. Thus, the exclusion of slick samples might overestimate SAS/TDC enrichment. Please discuss the occurrence of slicks in your study.	We are aware of the study by Romano et al (1996) by the automatic detection of slicks using photographic techniques. Romano used different categories of slick, and in our study we have excluded only strong slick formation (total absence of ripples within slick area, e.g. glassy surface). We have highlighted this information in the revised manuscript as well provide more details on the occurrence of slicks during our study (see Section 3.1). We have added the number of slick samples (n=60) to the section 2, all collected within the fjord Saanich Inlet after rainfall events frequently occurring between fall and spring in British Columbia.
P5727, L28 Poor phrasing. Please also provide references.	We agree, and have deleted the phrase on page 5727, Line 28 from the revised manuscript. We have added following text to the introduction for detailed information on the

	source of natural surfactants including polysaccharides:
	"High molecular polysaccharides and complex β-glucans are major excretion productions of marine phytoplankton (Allan et al., 1972; Mykelstad, 1974) and frequently found in seawater during blooms (Sakugawa and Handa, 1985). The hydrophilic polysaccharides may conjugate to hydrophobic moieties, making them surface active, and become enriched in the SML by factors of up to 3 compared to subsurface waters (Williams et al., 1986; Wurl and Holmes, 2008; Wurl et al., 2009).".
P5732, L15-18 Please be more precise, as it remains unclear what the implications for this manuscript would be, i.e. how does particle transformations effect SAS/TDC enrichment in the open ocean?	We agree, and we have re-phrased the sentence. Furthermore, Carlson (1993) provided evidence that constant compression and dilation of the SML can facilitate particle aggregation and condensation of high molecular weight (HMW) material from surfactants, including dissolved carbohydrates. Such enhanced particle aggregation and condensation of HMW DOM are likely to be important in the formation and stabilization of the SML (Hunter, 1980)."
P5735, L25-P5736, L1 Please correct referencing: Cunliffe et al 2009b did not report any bacterial abundances. This has been done in Cunliffe et al. 2009a. Moreover, please be aware that Fehon&Oliver counted viable cells on agar plates, which is a highly disputed method for bacterial quantification. The authors should better refer to studies using other techniques, although there is generally contradictory evidence on bacterial enrichment in the SML.	We agree. We have change the reference of Cunliffe et al 2009b to Cunliffe et al. 2009a. We have deleted the reference of Frehon and Oliver (1979) although the value of their study is certainly confirmed by the fact they have concluded from their results that bacterial communities in the microlayer are taxonomically different from underlying communities. Their conclusion has been confirmed decades later with new molecular techniques (i.e. 16S rRNA). We have added the new reference of Carlucci and Wolgast (1992) reporting enrichment of bacterial abundances using epifluoresence measurements.
Generally, the whole section, especially	We agree that the discussion needs

P5735, L17 – P5736, L14 needs to be rewritten in terms of phrasing as it is somewhat imprecise.	clarification. We have revised this part of the discussion intensively (please see below for revised text). We have also considered the comments of referee #1 and #2 in the revised text of the discussions.
P5746, Table 2 Please define abbreviations in the legend	We agree. We have revised the Table 2 and all abbreviations are now defined.
Technical corrections	Without referring to each comment, we agree to all the suggested corrections.

Revised section 4.2 (Discussion)

We found that the surfactant enrichments in the SML are higher in oligotrophic waters than in mesotrophic and eutrophic waters. The difference in those enrichments between regions with low and moderate/high primary productivity is small (EF=2.8 vs. EF=1.8) but statistically significant (p=0.01). The observation is supported by the seasonal variations in the enrichment of carbohydrates, metabolic exudates by phytoplankton, in Saanich Inlet, with the highest enrichments during seasons of low productivity and vice versa (Figure 6).

We suggest that the observation of diminishing enrichments with increasing productivity is a general phenomenon, as it has been observed in several other studies for DOM enrichment in sea- and freshwaters (Carlson, 1983; Södergren, 1987; Zhou and Mopper, 1998; Hillbricht-Ilkowska and Kostrzewka-Szlakowska, 2004; Kostrzewka-Szlakowska, 2005; Baastrup-Spohr and Staehr, 2009; Cunliffe et al., 2009). Correlations between enrichment factors and primary production or chl-*a* suffer from a lack of significance, as such simple correlations are confounded by multiple and complex enrichment processes. However, we suggest that the general observation of diminishing enrichments with increasing productivity is an obvious approach to approximate SML coverage based on primary productivity maps (section 4.3). Conditions responsible for the differences in the enrichment between the trophic states are the subject of the following discussion.

Carlson (1983) reported the first observations of diminishing DOC enrichments from oceanic to productive coastal waters. He suggested that increased solubility of potentially surface-active material with increasing DOC concentrations in bulk water could cause losses of material prone to accumulate in the SML leading to lower enrichments with increasing productivity. Different DOM composition between trophic states may not only lead to different tendencies of material to accumulate in the SML, but also determine the fate of enriched DOM. For example, dissolved lignin in open ocean waters was significantly more resistant to photochemical alteration than terrestrial lignin in river waters (Opshal and Benner, 1998). In an earlier study, we showed that transparent exopolymer particles (TEP), abundant gel-like particles in the ocean, are significantly enriched in the open ocean (chl-a < 2 μg L^{-1}) but not in coastal waters (chl-a > 5 μg L^{-1}) (Wurl and Holmes, 2008). Azam and Malfatti (2007) suggested that depending on their surface properties, gels may adsorb DOM components from the dissolved phase. For this reason, it is possible that TEPs in oligotrophic SML may draw proportionately more DOM into the SML. On the other hand, we suggest that eutrophic conditions in bulk water represent a very complex system in which DOM is more rapidly recycled, transformed, or adsorbed on abundant cell surfaces before it can reach the SML.

Carlson (1983) further suggests that differences in enrichments between oceanic and coastal waters may reflect differences in SML accumulation or removal processes. We agree with Carlson (1983) that it is unlikely that the intensity of near-surface advective and diffusive processes in oligotrophic waters (e.g. oceanic) are sufficiently greater than in meso- or eutrophic waters to cause larger enrichments in low productivity regions. Zhou and Mopper (1997) observed deposition of DOM onto the SML from the atmosphere, but there is no reason

to argue that this process would be significantly larger in oligotrophic regions. More likely, the removal of SML material through the formation of particulate matter sinking down into the bulkwater may be more effective in more productive waters. For example, Wheeler (1975) was able to observe particle formation in the SML by compression of coastal water, but not in oceanic waters. Sholkovity et al. (1978) suggested that terrestrial materials can contribute to particle formation, and such material would have been abundantly available at the sampling site in Saanich Inlet, which we are using in this study as a represetive for eutrophic conditions, in general.

Surfactant production by bacteria and/or zooplankton (i.e., non-chlorophyll containing organisms) may become more important in oligotrophic conditions. Recently, Jiao et al. (2010) highlighted the importance of the bacterial production of refractory material, whereas Kujawinski et al. (2002) provided evidence of grazing-mediated surface-active DOM production. Both processes may become the major source of surface-active material under oligotrophic conditions. Jiao et al. (2010) also pointed out that recalcitrant DOM can be released during bacterial degradation of POM, which is typically enriched even in oceanic microlayers by factors of up to 40 (Carlson, 1983).