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Interactive comment on "Seasonal variations in concentration and composition of dissolved organic carbon in Tokyo Bay" *by* A. Kubo et al.

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We thank Referee #2 for providing helpful comments and suggestions to improve and clarify the manuscript. The suggestions were carefully considered and implemented in the text. Our replies to individual comments are detailed below:

Replies to comments from anonymous referee #2 General Comments: Comment: In aquatic and marine environmental studies, dissolved organic matter (DOM) composition refers to the assessment of some aspect of DOM's chemical characteristics. For example, the contribution of carbohydrates, proteins, or lipids to total DOC concentrations, or assessing bulk DOC concentrations in different size fractions of DOC (i.e. Low and High molecular weight material). The bulk pool of DOC or DOM, however, has been classified into broad "pools of lability", which includes the labile, semi-labile, and

refractory pools. It is in my opinion that this manuscript actually assessed differences in the lability of ambient DOC rather than DOC composition. Thus, the title and all associated references to composition within the text should be changed to reflect that the authors studied DOC lability.

Author response: Thanks for your suggestion. We corrected "lability" instead of "composition" within title and text in the revised manuscript.

Comment: Another major issue has to do with the methodology of apparently not filtering surface water before conducting the DOC degradation experiments and the DOC samples from these experiments. This suggests that, for the experiments, TOC was actually analyzed, which could enhance the decay of TOC in the experiments because particles are less diagenetically altered and general more bioavailable than accumulated DOC. Thus, the BDOC and RDOC abundance and removal rate estimates would be corrupted.

Author response: Both DOC samples and degradation experiments samples were filtered on GF/F. According to the comment, we restructured the sentence in the revised manuscript.

Comment: Another problem is the way that RDOC is being defined and used throughout the text. The issue is that bottom water RDOC concentrations are significantly less than experimental RDOC concentrations after 150 days. Yes, this leads to export of "RDOC" from terrestrial sources to the ocean, but it would help if the authors better defined their lability timescales of RDOC as a whole. One could argue that, relative to bottom water concentrations, the exported "terrestrial RDOC" is actually semi-labile. Clarity would help with this confusion.

Author response: According to the comment, we added the discussion in the revised manuscript. The RDOC concentrations of the surface water were significantly higher than those of the bottom water at 06 (see Table S3 in the auxiliary material). Thus, our RDOC results likely include a fraction of semi-labile DOC. Degradation of this semi-

labile DOC fraction would occur by bacterial mineralization with longer time, photodegradation (Moran and Zepp, 1997; Opsahl and Benner, 1997; Obernosterer and Benner, 2004), aggregation (Sholkovitz, 1976; Mulholland, 1981), and/or sorption to particles (Chin et al., 1998; Kerner et al., 2003). However, the results of this study did not change significantly when DOC were divided into BDOC, semi-labile DOC, and RDOC. The lifetime of semi-labile DOC is about 1.5 years (Hansell et al., 2013), which is considerably longer than the residence time of Tokyo Bay (Takada et al., 1992). Therefore, in our analysis, there was no problem with inclusion of semi-labile DOC in RDOC. In addition, Ogura (1975) only divided DOC into BDOC and RDOC; therefore, we divided DOC in the same way to enable comparison with that study.

Comment: Lastly, the discussion sections of the paper need quite a bit of work as there is little discussion relative to other studies in other systems, including the significance of looking at DOM sources and comparison to different methodologies (e.g. isotopic vs. multiple regression using salinity and chla).

Author response: Thank you for your suggestion. According to the comment, we added the discussion in the revised manuscript. We added the sentence as follows: "The fate of terrestrial DOC in the coastal ocean and open ocean has long been the subject of debate (Hedges et al., 1997). For example, biomarkers (e.g. lignin phenols) and the stable carbon isotopic composition of DOC are commonly used to estimate the contribution of terrestrial DOC to the open ocean (Druffel et al., 1992; Hedges et al., 1997; Raymond and Bauer, 2001; Bauer and Bianchi, 2011). Lignin phenols analysis indicated that terrestrial DOC comprises only a small fraction (4–10%) of the total DOC in the open ocean (Meyers-Schulte and Hedges, 1986; Opshal and Benner, 1997; Hernes and Benner, 2006). In addition, the stable carbon isotopic composition of DOC also indicated that terrestrial DOC represents less than 10% of the total DOC (Bauer et al., 2002). As a result, most terrestrial DOC is remineralized in coastal waters, and only a small fraction is exported to the open ocean. In this study, terrestrial RDOC in the surface bay mouth accounted for less than 20% of the total RDOC (Table 5). Although

these levels were slightly higher than those reported in previous studies using lignin phenols and stable carbon isotopic compositions of DOC, they are probably reasonable given that exported terrestrial RDOC were further diluted with open oceanic water once outside the bay. Nevertheless, more complete information regarding the sources and lability of DOC are important to enable a better understanding of the fate of DOC in the coastal ocean and open ocean."

Specific Comments: Title page: Comment: Change composition to lability, as no DOM compositional analysis was conducted.

Author response: According to the comment, we changed to "lability" in the revised manuscript.

Abstract Page Comment: 10204 line 1: Abstract topic sentence is long and should be shortened.

Author response: According to the comment, we restructured the sentence in the revised manuscript.

Comment: Page 10204 line 20: Remove words "DOC", and leave as "exported mostly RDOC"

Author response: According to the comment, we remove words "DOC" in the revised manuscript.

Introduction Page Comment: 10205 line 2: Add "diverse mixture of carbon with varying timescales of lability", or something similar.

Author response: According to the comment, we added the sentence in the revised manuscript.

Comment: Page 10205 line 15: Change composition to lability (do so onward for this issue).

Author response: According to the comment, we changed to "lability" in the revised

manuscript.

Comment: Page 10205 paragraph 2: Tell us why you've studied BDOC and RDOC and the significance.

Author response: According to the comment, we added a phrase in the revised manuscript.

Materials and Methods Comment: Page 10206: Freshwater and seawater DOC samples were G/FF filtered, but the degradation experiments appear to be conducted with unfiltered surface water. If this is the case, this is problematic because particles could be contributing to the bioavailable signal, thus impacting all the rate constant calculations and estimates of RDOC vs. BDOC. Please account for this.

Author response: Both DOC samples and degradation experiments samples were filtered on GF/F. According to the comment, we restructured the sentence in the revised manuscript.

Results and Discussion Comment: Page 10208 line 25: add the word concentrations after POC

Author response: According to the comment, we added "concentrations" in the revised manuscript.

Comment: Page 10209 line 1: At this point in the paper, all we have to go on for whether water is being contaminated by sewage is DOC concentration. It would be really helpful if some other water quality indicator could be included, such as N and P concentrations, etc.

Author response: Thank you for pointing them out. We inserted table of nutrients concentration data at supplement material.

Comment: Page 10210 Section 3.2 Tokyo Bay: The Tokyo Bay bottom water DOC and RDOC concentrations are lower than those of RDOC at the end of the 150 day exper-

iments. How can both be called RDOC? The authors need to clarify these differences in lability timescales upfront.

Author response: According to the comment, we added the possibility about semi-labile DOC. The lifetime of semi-labile DOC is about 1.5 years (Hansell et al., 2013), which is considerably longer than the residence time of Tokyo Bay (Takada et al., 1992). Therefore, in our analysis, there was no problem with inclusion of semi-labile DOC in RDOC. In addition, Ogura (1975) only divided DOC into BDOC and RDOC; therefore, we divided DOC in the same way to enable comparison with that study.

Comment: RDOC Sources Page 10213 Lines 6-20: Some discussion of the % breakdown of RDOC sources and how it compares to other studies that use a similar or isotopic approach would be helpful. As it stands, this section is mainly more results than discussion. What is significant about the bay exporting terrestrial RDOC to the ocean? Is this common? How rapidly is terrestrial DOM thought to be degraded in coastal systems? What impact does salinity have on terrestrial DOM degradation? Discussion of things like this would help round out the paper.

Author response: Thank you for your suggestion. According to the comment, we added the discussion in the revised manuscript.

Figures and Tables Comment: Table 3. Define X.

Author response: According to the comment, we defined X in the revised manuscript. We changed in Table 3 and Table 4 due to moving comparison of Ogura (1975) data to section 3.3 in revised manuscript.

Comment: Figure 3. POC concentrations are not properly labeled.

Author response: According to the comment, we rearranged the figures in the revised manuscript.

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McLusky, D. S., Eds.), vol. 5, pp. 7-67, Academic Press, Sam Diego, 2011. Bauer, J. E., Druffel, E. R. M., Wolgast, D. M., and Griffin, S.: Temporal and regional variability in sources and cycling of DOC and POC in the northwest Atlantic continental shelf and slope, Deep-Sea Res. Pt II, 49, 4387-4419, 2002. Chin, W. C., Orellana, M. V., and Verdugo, P.: Spontaneous assembly of marine dissolved organic matter into polymer gels, Nature, 391, 568-572, 1998. Druffel, E. R. M., Williams, P. M., Bauer, J. E., and Ertel, J. R.: Cycling of dissolved and particulate organic matter in the open ocean, J. Geophys. Res. Oceans, 97, 15639-15659, 1992. Kerner, M., Hohenberg, H., Ertl, S., Reckermann, M., and Spitzy, A.: Self-organization of dissolved organic matter to micelle-like microparticles in river water, Nature, 422, 150-154, 2003. Hansell, D. A.: Recalcitrant dissolved organic carbon fractions, Annu. Rev. Mar. Sci., 5, 3.1-3.25, 2013. Hedges, J., Keil., R. G., and Benner, R.: What happens to terrestrial organic matter in the ocean?, Org. Geochem., 27, 195-212, 1997. Hernes, P. J., and Benner, R.: Terrigenous organic matter sources and reactivity in the North Atlantic Ocean and a comparison to the Arctic and Pacific oceans, Mar. Chem., 100, 66-79, 2006. Meyers-Schulte, K. J., and Hedges, J. I.: Molecular evidence for a terrestrial component of organic matter dissolved in ocean water, Nature, 321, 61-63, 1986. Moran, M. A., and Zepp, R.G.: Role of photoreactions in the formation of biologically labile compounds from dissolved organic matter, Limnol. Oceanogr., 42, 1307-1316, 1997. Mulholland, P. J.: Formation of particulate organic carbon in water from a southeastern swamp-stream, Limnol. Oceanogr., 26, 790-795, 1981. Obernosterer, I, and Benner, R.: Competition between biological and photochemical processes in the mineralization of dissolved organic carbon, Limnol. Oceanogr., 49, 117-124, 2004. Ogura, N.: Further studies on decomposition of dissolved organic matter in coastal seawater, Mar. Biol., 31, 101-111, 1975. Opsahl, S., and Benner, R.: Distribution and cycling of terrigenous dissolved organic matter in the ocean, Nature, 386, 480-482, 1997. Raymond, P. A., and Bauer, J. E.: Riverine export of aged terrestrial organic matter to the North Atlantic Ocean, Nature, 409, 497-500, 2001. Sholkovitz, E. R.: Flocculation of dissolved organic and inorganic matter during the mixing of river water and seawater, Geochim.

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Interactive comment on Biogeosciences Discuss., 11, 10203, 2014.