We greatly appreciate the referee's thorough and constructive comments. Our responses follow the referee's comments and are *italicized*.

Responses to referee#2

1. …While comparison of filtered and unfiltered samples provides insight regarding particle- versus CDOM-based CO photoproduction, I am not convinced that the derivation of a CO AQY for particles (based on mathematical manipulation of the AQY-filtered and AQY-unfiltered) is entirely appropriate. Stronger rationalization is needed for how/why bulk particulate and POM CO AQY were retrieved. The spectral differences in AQY-CDOM and AQY-total should be focused on rather than attempting to isolate the effect of particles alone.

   The reviewer provided more detailed comments on these concerns below. Please see our responses to his/her specific comments. As for why particulate and POM AQY were retrieved, because this separation makes it possible to compare particulate and POM photoreactivity with CDOM photoreactivity (with respect to CO photoproduction), which has a growing interest in particle photochemistry.

2. Page 16171/Line 14: Do you have any experimental evidence to support your claim that scattering had only a minimal effect? This claim warrants further discussion.

   Please see our response to reviewer#1's comment#8.

3. Page 16172/Line 5-17: There are assumptions involved in the modeling of AQY spectra from measured CO photoproduction rates that lead to uncertainty unless corrected for (e.g., changes in absorption coefficients over irradiation time due to bleaching, conversion of DOM to POM (or POM to DOM), settling of particulate matter, etc.). Furthermore, there is uncertainty associated with evaluating the wavelength dependence of CO photoproduction by fitting a curve to data points generated using cutoff filters. Without evaluating this uncertainty, it is unclear whether equation 6 is appropriate. Can the difference in AQY observed in the filtered and unfiltered samples be completely attributed to particles? Is CDOM producing the same amount of CO in presence and absence of particles? How valid is it to assume that screening/scattering effects are negligible?

   Please see our responses to reviewer#1's comments#7&10 on photobleaching, particle settling, and self-shading.
   <u>POM to DOM or DOM to PM conversion:</u> *Because CDOM absorbance did not change significantly before and after irradiation (due to the very short irradiation times), the inter-conversion between CDOM and POM should not be important. In addition, as we already stated in the original version (last paragraph on page 16179), the current study does not aim to distinguish between CO produced directly from particles and CO produced from POM-derived CDOM. This subject is certainly interesting but requires a detailed mechanistic study to elucidate.*
   <u>Suitability of AQY curve-fitting:</u> *To derive AQY spectra using the multispectral irradiation-statistic fitting approach as adopted in our study, it has been generally accepted that AQY decreases with wavelength exponentially or quasi-exponentially (see review by White et al. (2010)) for many CDOM photoprocesses (e.g CO, CO2, and ammonium photoproduction and DMS photooxidation). This is supported by*

many monochromatic irradiations which show similar AQY spectral shapes (e.g. Weiss et al., 1995; Zafiriou et al., 2003; Stubbins et al., 2011). Ziolkowski and Miller (2007) directly compared the monochromatic and multispectral irradiation approaches for determining CO AQY and found good agreement between the two methods. For our unfiltered samples, the pattern of CO production rate vs. cutoff wavelength was similar to that for the filtered samples, suggesting that particle CO AQY possesses a similar spectral shape to that of CDOM-based CO AQY. The good agreement between the measured CO photoproduction rate and the CO photoproduction rate calculated from the retrieved AQY (R^2:range 0.969-0.998, mean 0.991 for filtered samples; range 0.982-0.998, mean 0.992 for unfiltered samples) indicate that the assumed AQY form is generally appropriate for both the filtered and unfiltered samples. In fact, Estapa et al. (2012) have recently used a similar approach to derive the AQY of DOC photoproduction from POM. Admittedly, the multispectral curve fitting approach could somewhat smooth out fine structures, if any, related to CO photoproduction from chlorophylls in unfiltered samples. Chlorophyll-a has an absorption peak at ca. 675 nm, for which our experiment could not resolve since our longest cutoff wavelength was 495 nm (any potential production at wavelength>600 nm was ascribed to the wavelengths between the 495 cutoff and 600 nm). Future studies using monochromatic or longer cutoff filters are needed to resolve this issue. Several lines are added to the new version to underline this caveat (page13.line13-18)
Equation 6: *this equation states the fact that the CO production in an unfiltered sample is the sum of CO productions from CDOM and particles. It holds without the need of meeting other conditions.*
Can the difference in AQY observed in the filtered and unfiltered samples be completely attributed to particles? *As there were only two classes of substrates (CDOM and particles) for CO production, it is reasonable to assume that the difference was due to particles.*
Is CDOM producing the same amount of CO in presence and absence of particles? *Particles compete with CDOM for photons, therefore reducing CO production from CDOM. However, the procedure for retrieving the AQY already took into account this effect (see equations 5, 7, 8 in the original version).*
Scattering effect: *Please see response to comment 2.*

4. Page 16174/Lines 1-12: Particulate optical density was measured but no other data is given to quantify/characterize particulate matter (suspended particulate matter and particulate organic carbon were not measured). Is there any absorption coefficient/organic carbon data available for this region in the literature that you could use to estimate the quality and/or quantify of particulate material in your samples?

As mentioned in Section 3.3, we did not measure SPM and POC concentrations but David Doxaran's team measured these two parameters during the same cruise. We now added several lines about the POC:SPM ratio in the discussion of particle CO AQY by citing Doxaran et al. (2012) as a reference (page17.line12-page18.line5; page19.line16-21).

5. Page 16177/Line 12-Page 16178/Line 11: This section is difficult to follow. How was doing a sensitivity analysis to evaluate AQY-POM the first step in retrieving AQY-POM? The approach seems to be given in reverse of how it was applied.

This section has been greatly shortened and re-organized following reviewer#1's

comment 16.

6. Page 16179/Lines 22-25: How was this done? Did you measure POM? How do you know the new CDOM wasn't produced from POM at the same time that old CDOM was lost (bleached)?

Please see our response to your comment#3 and to reviewer#1's comment#7 and Fig S1 in supplementary material. As the photobleaching in filtered samples was insignificant, the insignificant change of a_{cdom} in the unfiltered samples implies that any inter-conversion between CDOM and POM was minor as well. Surely, some CDOM molecules must have been bleached but the extent was so small (due to the short irradiations) that it was essentially not measurable.

7. Page 16199/Table 2: Consider including a(total, 412) for comparison with a(CDOM) and a(particle).

a_t is simply the sum of a_{cdom} and a_p. The ratio of a_p:a_{cdom} is more informative for this study than the ratios of a_p:a_t and a_{cdom}:a_t. To make the table succinct, we decided not to add a_t.

8. Technical corrections
Page 16166/Line 27: Reference cited should not be given as a web address.

Thanks. We will obtain suggestion from the BG editorial staff concerning how to cite website in this journal.

Page 16198/Table 1: Temperature units (degrees C) are given for Salinity rather than Temperature.

Thanks. This error is corrected.

References cited in responses to reviews:

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Estapa, M. L., Mayer, L. M., and Boss, E.: Rate and apparent quantum yield of photodissolution of sedimentary organic matter, Limnol. Oceanogr. 57, 1743-1756, 2012.

Stubbins, A., Law, C. S., Uher, G., and Upstill-Goddard, R. C.: Carbon monoxide apparent quantum yields and photoproduction in the Tyne estuary, Biogeosciences, 8, 703-713, 2011.

Weiss, P.S., Andrews, S.S., Johnson, J.E., Zafiriou, O.C.: Photoproduction of carbonyl sulfide in South Pacific Ocean waters as a function of irradiation wavelength, Geophys. Res. Lett., 22, 215–218, 1995.

White, E. M., Kieber, D. J., Sherrard, J., Miller, W. L., Mopper, K.: Carbon dioxide and carbon monoxide photoproduction quantum yields in the Delaware Estuary, Mar. Chem., 118, 11–21, 2010.

Zafiriou, O. C., Andrews, S. S., and Wang, W.: Concordant estimates of oceanic carbon monoxide source and sink process in the Pacific yield a balanced global

"blue-water" CO budget, Global Biogeochem. Cycles, 17(1), 1015, doi:10.1029/2001GB001638, 2003.

Ziolkowski, L. A., and W. L. Miller (2007), Variability of the apparent quantum efficiency of CO photoproduction in the Gulf of Maine and Northwest Atlantic, Mar. Chem., 105, 258-270.