

## ***Interactive comment on “Contribution of dissolved organic matter to submicron water-soluble organic aerosols in the marine boundary layer over the eastern equatorial Pacific” by Y. Miyazaki et al.***

**Dr. Ceburnis**

darius.ceburnis@nuigalway.ie

Received and published: 29 March 2016

This paper is an essential contribution to marine aerosol research field as it reports on the state-of-the-art aerosol measurements in the under-researched oceanic region and I am looking forward to it being published. Ship-born campaigns are absolutely critical in areas where research infrastructures simply do not exist or hardly accessible and are superior to short-lasting airborne measurements. Stable carbon isotope analysis is a highly informing technique when it comes to carbonaceous matter origin over the ocean due to distinct isotopic signatures of marine versus continental sources. To my knowledge the technique was first applied to carbonaceous particulate matter by Chesselet et al. (1981) and pursued by many others although in the past the technique

C1

did not reveal its full resolving power due to heavily polluted Northern Hemisphere. The authors may also refer to the dual carbon isotope technique proposed by Ceburnis et al. (2011) where source contribution and source specific isotope values were constrained by the dual isotope method and being constrained marine source isotope value (-20‰) closely match the observed stable carbon values of this study (-19.8±2.0‰) which is fascinating.

I would like to comment on a few specific issues which may help to improve an otherwise exciting paper.

I am somewhat confused over the terms “open oceans” and “coastal oceans”. By definition the ocean beyond a continental shelf should be considered an open ocean. The continental shelf in the reported region R3 does not extend beyond 100 km from the coast and the shelf sea occupied only a part of R3 region. In my opinion the difference between open ocean and coastal ocean in this study was in the abundance of chlorophyll in surface waters. However, chlorophyll rich waters can be found thousands of kilometres from the coast as in the North Atlantic & Arctic (O’Dowd et al., 2004; Yoon et al., 2007; Russell et al., 2010) or the Southern Ocean (Sciare et al., 2009). Is it not better referring to “high/rich” and “low/poor” chlorophyll regions?

I suggest using the term “sea spray” when referring to primary marine particles as sea salt is only one component of sea spray (Section 3.1., line 31). Major sea salt ions should not be expected to correlate linearly with wind speed as there is a power law relationship between sea salt(spray) mass and wind speed, e.g. (Ovadnevaite et al., 2012), hence, moderate linear correlation observed. Cl depletion during certain periods was rightly attributed to acidifying species where sulfate (sulfuric acid) should have been dominant component contrary to nitrate (nitric acid) which significant presence could not be corroborated by stable carbon values (significant nitrate must be anthropogenic and consequently would have driven C13 to more depleted values). It is very likely that if sulfate was plotted against Cl/Na ratio an apparent anti-correlation was clearly revealed. Cl depletion by sulfuric acid is rarely implicated due to the ever-

C2

present nitrate, but in clean oceanic regions sulfuric acid must dominate the observed depletion.

Despite anthropogenic sources contributed negligibly to the TC mass, its contribution cannot be totally discounted in the Northern Hemisphere and it would have resulted in slightly more negative stable carbon ratio of TC as it was measured (Section 3.5). Very small contribution of anthropogenic carbonaceous matter would not compromise the results of this study as the region was predominantly clean based on cumulative evidence from all the tracers. It is hard to discount small amount of anthropogenic tracers without concurrent BC measurements as the Shank et al. study was conducted during different season and meteorological conditions. Lastly, even small amount of BC is always accompanied by a small corresponding OC mass.

Ceburnis, D., Garbaras, A., Szidat, S., Rinaldi, M., Fahrni, S., Perron, N., Wacker, L., Leinert, S., Remeikis, V., Facchini, M. C., Prevot, A. S. H., Jennings, S. G., Ramonet, M., and O'Dowd, C. D.: Quantification of the carbonaceous matter origin in submicron marine aerosol by C-13 and C-14 isotope analysis, *Atmos. Chem. Phys.*, 11, 8593-8606, 10.5194/acp-11-8593-2011, 2011.

Chesselet, R., Fontugne, M., Buatmenard, P., Ezat, U., and Lambert, C. E.: The origin of particulate organic carbon in the marine atmosphere as indicated by its stable carbon isotopic composition, *Geophys. Res. Lett.*, 8, 345-348, 1981.

O'Dowd, C. D., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., Fuzzi, S., Yoon, Y. J., and Putaud, J. P.: Biogenically driven organic contribution to marine aerosol, *Nature*, 431, 676-680, 10.1038/Nature02959, 2004.

Ovadnevaite, J., Ceburnis, D., Canagaratna, M., Berresheim, H., Bialek, J., Martucci, G., Worsnop, D. R., and O'Dowd, C.: On the effect of wind speed on submicron sea salt mass concentrations and source fluxes, *J. Geophys. Res.-Atmos.*, 117, 10.1029/2011jd017379, 2012.

### C3

Russell, L. M., Hawkins, L. N., Frossard, A. A., Quinn, P. K., and Bates, T. S.: Carbohydrate-like composition of submicron atmospheric particles and their production from ocean bubble bursting, *Proceedings of the National Academy of Sciences of the United States of America*, 107, 6652-6657, 10.1073/pnas.0908905107, 2010.

Sciare, J., Favez, O., Sarda-Esteve, R., Oikonomou, K., Cachier, H., and Kazan, V.: Long-term observations of carbonaceous aerosols in the Austral Ocean atmosphere: Evidence of a biogenic marine organic source, *J. Geophys. Res.-Atmos.*, 114, D15302, 10.1029/2009jd011998, 2009.

Yoon, Y. J., Ceburnis, D., Cavalli, F., Jourdan, O., Putaud, J. P., Facchini, M. C., Decesari, S., Fuzzi, S., Sellegri, K., Jennings, S. G., and O'Dowd, C. D.: Seasonal characteristics of the physicochemical properties of North Atlantic marine atmospheric aerosols, *J. Geophys. Res.-Atmos.*, 112, D04206, 10.1029/2005jd007044, 2007.

---

Interactive comment on *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2016-164, 2016.

### C4