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The potential impact of DOM accumulation on computed fCO_2, carbonate ion concentrations, and omega in ocean acidification experiments

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The internal consistency of measurements and computations of components of the CO_2-system, namely total alkalinity (A_T), total dissolved carbon dioxide (C_T), CO_2 fugacity (fCO_2), and pH, has been confirmed repeatedly in open ocean studies when the CO_2 system had been over determined. Differences between measured and computed properties, e.g. ΔfCO_2 (=fCO_{2meas} – $fCO_{2(AT+CT)}$)/ fCO_{2meas}*100), there are usually below 5%. Recently, Hoppe et al. (2010, Biogeosciences Discussions) provided evidence of significantly larger ΔfCO_2 in experimental setups. These observations are currently not well understood. Here we provide additional examples from phytoplankton culture and mesocosm experiments of ΔfCO_2 of up to 40%. ΔfCO_2 varied systematically during the course of these experiments. In one set of experiments a clear correlation with the accumulation of dissolved organic carbon (DOC) is evident. Culture and mesocosm experiments are often carried out under very high initial nutrient concentrations, yielding high biomass concentrations that in turn often lead to a substantial build-up DOC. DOC can reach concentrations much higher than typically observed in the open ocean. To the extent that DOC includes organic acids and bases, it will contribute to the alkalinity of the sea water contained in the experimental device. Our analysis suggests that whenever substantial amounts of DOC are produced during the experiment, standard computer programs used to compute CO_2 fugacity can underestimate true fCO_2 significantly. Alternative explanations for large ΔfCO_2, e.g. uncertainties of pKs, are explore as well, but are found to be of minor importance.