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Interactive comment on “Seasonal signatures in SFG vibrational spectra of the sea surface nanolayer at Boknis Eck Time Series Station (SW Baltic Sea)” by K. Laß et al.

Anonymous Referee #2

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The manuscript submitted by Laß et al. describes the temporal dynamics of organic material at the air-water interface, which was measured by sum frequency generation (SFG) spectroscopy, over a period of 3.5 years. With this study the authors contribute to two main fields within air-sea interface research, which have been received too little attention so far: (a) the better characterisation of organic material accumulating in the interface and (b) more insights into the temporal variability of interface properties. Thus, the study is of principal interest for the readership of Biogeosciences. The manuscript is generally well structured and has a clear language. Nevertheless, I have some concerns about the discussion and conclusions presented, which need to be addressed, before publication should be considered.

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General comments: The authors relate SFG signals from the interface, which they refer to as the nanolayer, to parameters in the underlying water column (1 meter depth) in order to discuss the relation of SFG dynamics with biological activity. The discussion lacks information about a possible discrepancy between the investigated parameters – e.g. Chlorophyll a - in the nanolayer (or microlayer as the underlying interfacial layer) and the bulk phase. Principally, it is suggested throughout the literature, that material in the interface is dominantly imported from the underlying bulk water. However, there might be short-term effects, changing this dependency (precipitation, turbulent mixing) and leading to remarkably different concentration of organic material between the interface and the bulk water. In this respect, also a discussion about wind speed affecting interfacial concentrations of e.g. Chlorophyll a and DOC given in literature would help along with indications of the wind speed conditions during the monthly sampling of the study presented. Additionally, it would be helpful for the reader if the authors could make some comments on how the SFG signals, both in vibrational modes and in intensity, differ (or not?) between the samples taken with the screen or from 1 meter water depth.

Moreover, the temporal resolution of the sampling strategy needs some thoughts, especially regarding the conclusions drawn by the authors. I suggest that only monthly sampling prevented the detection of some phytoplankton blooming events, which possibly explains the lacking spring bloom signals in 2009 and 2010. This is especially worth to mention, as in 2010 the SFG signals are highest in spring and decline throughout the year. Thus, at least for this year I don't follow the authors with the explanation of sloppy feeding being the main driver for SFG signal variability in the nanolayer. Moreover, if sloppy feeding remains the hypothesised main driver, I also follow the suggestions by the reviewer O. Wurl to have a closer look at zooplankton data available for the sampling station. As zooplankton data for this period of investigation is not available (as can be read in the authors comments), a closer look at "historical" data might help - if available. Basically each year investigated showed a different seasonal SFG signal: 2009 – one peak in summer, 2010 – one peak in spring, 2011 – two peaks in spring

and summer. Thus, sloppy feeding maybe one driver for the signal intensity in summer, but the authors need to present some ideas about driving factors for the SFG signals in spring. Other biological factors should also be considered, such as heterotrophic bacterial “blooms” (as shortly but not sufficiently discussed by the authors) and their turnover by protists. I bet that such data is also available for the sampling station.

Specific comments: p.3179, l.8-14: I also had some difficulties with this paragraph and the clarification given by the authors in response to the review by O. Wurl is definitely needed. p. 3180, l.23: check explanation for the abbreviations. The authors need to switch words. p.3185, l. 19: for the reader refer to section 4.2.2 that this feature is discussed later. p.3187, l.21: from the data presented the “generally high layer intensity in June and July” is not only restricted to these months. (See also general comments above). p.3187, l.23-25: for the air-water interface, bulk material which is settling downwards is most likely not relevant, but its degradation and the resulting production of dissolved material would be. So the authors should go a bit deeper into the discussion here, including bacterial turnover of organic material. (See also general comments above). Fig. 3: why are August samples highlighted?

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