Response to Referee #1

RC: The paper is well written with a clear structure. The methodology is well described and the results are interesting. To summarize; the paper is in good shape and will make an interesting contribution to the Atmospheric Chemistry and Physics Discussions. In order to further improve the paper, e.g., the following could be elaborated:

AC: We would like to thank the reviewer for this encouraging feedback and the constructive comments, to which we will respond in detail below.

RC: Section 2.2. and related sections Ship motions are generally described by the six degrees of freedom that a ship can experience, while the ship in the large eddy simulation (Popinet et al., 2004) is stationary. The large eddy simulation uses numerical dissipation instead of a sub-grid scale turbulence model. An uniform inflow velocity is applied at the inlet boundary. These three aspects influence the results of the large eddy simulation and therefore in turn the calculation of the uplift. Please elaborate how these aspects are accounted for in the correlations?

AC: The reviewer correctly names the main shortcomings of the LES. We have acknowledged these shortcomings in Section 2.2. The deviations of the LES results from the observations may be attributed to these simplifications, as well as unresolved structures (such as mast supports) in the model. It was, however, not the focus of this work to improve the LES model and we did not investigate quantifying the uncertainties arising from the simplifications made. Here we present two additional methods, which use 3D wind speed measurements to validate air-flow models, aside from the difference of mean velocity and mean horizontal deflection measured at two different points. These are (i) the vertical tilt of the wind vector, which is measured from the ratio of mean vertical and horizontal velocities, and (ii) the vertical uplift, which can be estimated from the frequency distribution of the observed turbulences.

RC: Section 2.5. Page 10 (9-13): It is here suggested that it may be that small-scale turbulence adjusts to the new orientation of the tilted stream lines. Small-scale turbulence is typically more iso-tropic than large-scale turbulence. It is therefore counter intuitive that it rather is the small-scale than the large-scale turbulence that is adjusted to the tilted stream lines. It may, however, be that the magnitude of the small-scale turbulence is increased in areas of increased shear as a result of a tilted air flow. It may also be that since the small-scale turbulence is more iso-tropic than large-scale, it is not so influenced by the tilted air flow. Please clarify what is meant by the word adjust in this discussion.

AC: The sentence is indeed misleading (turned out wrong) we apologise and suggest rewording as follows: “Our observation in described in Sec. 2.5 suggests that while rotation of the coordi-
nate system into the air stream is crucial to adequately measure the contribution of large eddies, it is counter-productive for the measurement of flux carried by small eddies, i.e., it may be that the small scale turbulence ($\lambda < 1 \text{ m}$) does not adjust its orientation to the new flow direction as efficiently as the large scale turbulence. Another possible explanation could be that the magnitude of small scale turbulence may be increased locally as a result of the shear in the tilted and accelerated air flow.” We did also expanded on the following “Due to the projection of the auto-covariances of the three components $u, v,$ and $w$, the momentum flux estimate is generally more sensitive to the choice of the coordinate system than the scalar fluxes (see Wilczak et al., 2001). The sensitivity of the nCo$_w$ estimate to the tilt increases in the inertial sub-range, where the auto-covariances of the three velocity components diminish slower with increasing frequency ($f^{-2/3}$).”

RC : Section 2.5. Page 11 (9-22) This description does not belong to this section. Please consider moving it to another section or make it an own section.

AC : We moved this to a new Section 3.2.

RC : Section 2.8. Page 16 (12-13): The transition of $n$ is discussed in terms of smooth and rough due to increasing wind speed. $n$ is also to a large degree dependent on surfactants, especially during low wind conditions, e.g., [Fredriksson et al., 2004; McKenna and McGillis, 2004; Zhang et al., 2013]. Please add some references regarding this phenomenon.

AC : We have added the following sentence to acknowledged this phenomenon: “The exact shape wind speed dependence of this transition has been found in to depend on surfactant concentration on the water surface (e.g. Frew et al., 2004; McKenna and McGillis, 2004; Zhang et al., 2013; Krall, 2013).”

RC : Section 3.3(3.4) Page 23 (2) wind stress definition should be with squared friction velocity.

AC : Thanks for spotting this mistake, it is now corrected.

RC : Section 4.1. Page 30 (15-16) This section discusses how the gas transfer velocity relates to the friction velocity and that buoyancy-driven processes may contribute significantly at lower wind speeds. E.g., [Fredriksson et al., 2016] discuss the transition between a gas transfer velocity mainly driven by buoyancy processes to a gas transfer velocity mainly driven by shear stress processes via the Richardson number (relates the buoyancy flux to the friction velocity). This paper can be used in the discussion regarding the range of friction velocity, where the gas transfer velocity as a function of friction velocity is valid.

AC : The results of Fredriksson et al. (2016) suggest that the influence of the buoyancy flux on air-water gas exchange may become relevant at low wind speeds, depending on the magnitude of the surface buoyancy flux. For wind speeds below $5 \text{ m s}^{-1}$, the magnitude of the surface heat flux remained below 200 W m$^{-2}$ during SOAP. Our estimates of the Richardson number ($R_i = B_{0,w} \nu_w u_{*w}^{-4}$) were below the critical value of $R_i \approx 0.004$, suggested by Fredriksson et al. (2016). However, we would like to note here, that the slope of Eq. (10) is more than a factor of two higher than typical parametrisations of shear driven gas transfer (e.g. Jähne et al., 1987) and that Eq. (10) predicts negative gas exchange rates at low wind speeds. It is therefore likely that the observed CO$_2$ transfer velocities are from the combined effect of shear driven gas transfer as well as of wave breaking and bubble-driven gas transfer. It is however not within the scope of this paper to present a physical based equation, appropriately describes these contributions.

We have modified the part of Sec. 4.1 as follows:

“Eq. (10) should not be interpreted as physical law, but rather as empirical parametrisation for the wind speed range $5 - 19 \text{ m s}^{-1}$, Extrapolation of this linear $k$ vs EC $u_*$ relationship outside of the wind speed range of the SOAP data set is not recommended, because there are physical reasons why this relationship might not hold. At lower wind speeds, buoyancy-driven processes may contribute significantly to gas transfer (Soloviev, 2007; Fredriksson et al. 2016). In fact (10) slightly underestimates the wind speed binned data for $u_{10m} < 5 \text{ m s}^{-1}$ and would predict negative $k_{600}$ for $u_* \leq 0.07 \text{ m s}^{-1}$ ($u_{10m} \leq 2.3 \text{ m s}^{-1}$). However, since our estimations of the Richardson number ($R_i = B_{0,w} \nu_w u_{*w}^{-4}$) remained below the critical value of $R_i \approx 0.004$,
which was suggested by Fredriksson et al. (2016), we do not expect significant contribution of buoyancy-driven processes to the gas exchange rates observed during SOAP. Here \( B_{0,w} \), \( \nu_w \), and \( u_{*,w} \) are the water side surface buoyancy flux, kinematic viscosity of sea water, and waterside friction velocity respectively. At higher wind speeds, wave breaking and bubble-driven gas transfer are expected to contribute to gas transfer of CO\(_2\) and other sparingly soluble gases (Woolf, 1997; Fairall et al., 2011; Bell et al., 2017). Surprisingly, there is no evidence in the SOAP data for an increase in the slope of the \( k_{0,660} \) vs \( u_* \) relationship at high wind speeds. If anything, the limited SOAP data available at the highest wind speeds appear to be biased low relative to the linear regression.

References


Frew, N. M., Bock, E. J., Schimpf, U., Haru, T., Haußecker, H., Edson, J. B., McGillis, W. R.,