

An Assessment of the Precision and Confidence of Aquatic Eddy Correlation Measurements

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ABSTRACT

The quantification of benthic fluxes with the aquatic eddy correlation (EC) technique is based on simultaneous measurement of the current velocity and a targeted bottom water parameter (e.g., O₂, temperature). High-frequency measurements (64 Hz) are performed at a single point above the seafloor using an acoustic Doppler velocimeter (ADV) and a fast-responding sensor. The advantages of aquatic EC technique are that 1) it is noninvasive, 2) it integrates fluxes over a large area, and 3) it accounts for in situ hydrodynamics. The aquatic EC has gained acceptance as a powerful technique; however, an accurate assessment of the errors introduced by the spatial alignment of velocity and water constituent measurements and by their different response times is still needed.

Here, this paper discusses uncertainties and biases in the data treatment based on oxygen EC flux measurements in a large-scale flume facility with well-constrained hydrodynamics. These observations are used to review data processing procedures and to recommend improved deployment methods, thus improving the precision, reliability, and confidence of EC measurements. Specifically, this study demonstrates that 1) the alignment of the time series based on maximum cross correlation improved the precision of EC flux estimations; 2) an oxygen sensor with a response time of <0.4 s facilitates accurate EC fluxes estimates in turbulence regimes corresponding to horizontal velocities < 11 cm s⁻¹; and 3) the smallest possible distance (<1 cm) between the oxygen sensor and the ADV's sampling volume is important for accurate EC flux estimates, especially when the flow direction is perpendicular to the sensor's orientation.

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1. Introduction

Aquatic eddy correlation (EC) is an increasingly common technique to infer fluxes across the sediment–water interface in aquatic environments. The EC method is based on the simultaneous high-frequency measurement of the vertical velocity (normal to the local streamline) and the concentration of oxygen or other constituents (Berg et al. 2003). The method resolves the turbulent fluctuations in the benthic boundary layer (BBL), and the resulting time series data are averaged to obtain a mean benthic flux (Lorke et al. 2013). Performed in the turbulent boundary layer just above the sediment and assuming constant mean current velocities and oxygen concentrations (Holtappels et al. 2013), the measured flux represents the flux across the sediment–water interface averaged over an upstream-located footprint area (Berg et al. 2007).

Originally developed to resolve atmospheric fluxes in terrestrial environments (Baldocchi 2003), the EC application to the aquatic environment has shown promising results where conventional methods for benthic flux acquisition are difficult or impossible to apply (Glud et al. 2010; Hume et al. 2011; Long et al. 2012; Berg et al. 2013; Long et al. 2013). Despite the growing number of EC applications, there are still several unresolved issues, including deployment considerations (current direction, sensor spacing) as well as requirements on sensor response time. While these topics have been discussed in previous publications (McGinnis et al. 2008; Lorrai et al. 2010), their implications have not been fully assessed or investigated.

Indeed, among other constraints, the resolution of the instantaneous fluctuations of vertical velocity and oxygen concentration requires that 1) both measurements refer to exactly the same sampling volume and 2) all “flux carrying” eddies are properly resolved (Swinbank 1951; Foken et al. 2004). However, such requirements are not always fulfilled during in situ deployments. In practice, the oxygen sensor is located outside the sampling volume of the velocity measurement to avoid interference with the acoustic Doppler velocimeter (ADV). This physical separation, if not corrected for, can produce biased flux estimates (McGinnis et al. 2008; Lorrai et al. 2010; Billesbach 2011). Additionally, a potential bias can emerge due to the slower response time of the oxygen sensor relative to the response time of the ADV. Minimizing this potential bias requires a suitable time shift of both time series (by their cross correlation), but the loss of high-frequency fluctuations can still lead to errors (Eugster and Senn 1995; Aubinet et al. 2001). Although these issues were addressed theoretically, there exist no quantitative studies evaluating the potential effects of time shift and response times on eddy correlation flux estimates in the aquatic environment.

Here, we present the outcome of an international experimental workshop held in February 2012 at the Royal Netherlands Institute for Sea Research (NIOZ Yerseke) with the aim of exchanging ideas and experience among aquatic EC users. In this study, we address the effects of time shift and sensor response time on aquatic EC flux estimates. To this end, we performed experiments under controlled conditions in a large-scale laboratory flume combined with numerical modeling. From this, we developed a list of recommendations with respect to deployment and data processing to improve precision and reliability of EC flux measurements.

2. Materials and methods

a. Flume setup

Eddy correlation measurements were performed in a large racetrack flume facility (Fig. 1) with a total length of 17.55 m, a width of 0.60 m, and a straight working section of 10.8 m. The water flow is generated by a conveyor belt system that uses a series of paddles to drive the flow [see Jonsson et al. (2006) for the flume hydrodynamic characteristics, referred to as a NIOO flume]. Ten days prior to the start of the experiments, cohesive sediment was collected from an intertidal flat at Kapelle Bank (Westerschelde estuary, The Netherlands; porosity ~ 0.7 , organic carbon ~ 1.5 wt%) (Middelburg et al. 1996; de Brouwer et al. 2000). Visible fauna were removed from the sediment that was then homogenized and deposited as a 5-cm-thick bottom layer, covering the working section of the flume (Fig. 1).

Seawater from the sampling site (salinity: 32) was used to fill the flume to a water depth of 30 cm (total water volume ~ 9 m³). The temperature in the flume hall was kept constant at 17°C and the sediment was allowed to equilibrate for 10 days, before EC experiments were conducted. The flume was kept in darkness and low artificial light levels were only applied when instrumentation was added or changed; this ensured that no phototrophic growth took place in the flume. The temperature and O₂ concentrations in the flume water were continuously monitored with two optodes (4330F, Aanderaa), one located before the flow straighteners and the other optode was positioned at the end of the straight work section (Fig. 1).

b. EC system

The EC system consisted of an ADV (Nortek) with two gain-adjustable, galvanically insulated amplifiers (McGinnis et al. 2011), each equipped with custom-built Clark-type O₂ microelectrodes (Revsbech 1989; Gundersen et al. 1998). The O₂ microelectrodes were calibrated against the air-saturated flume water and

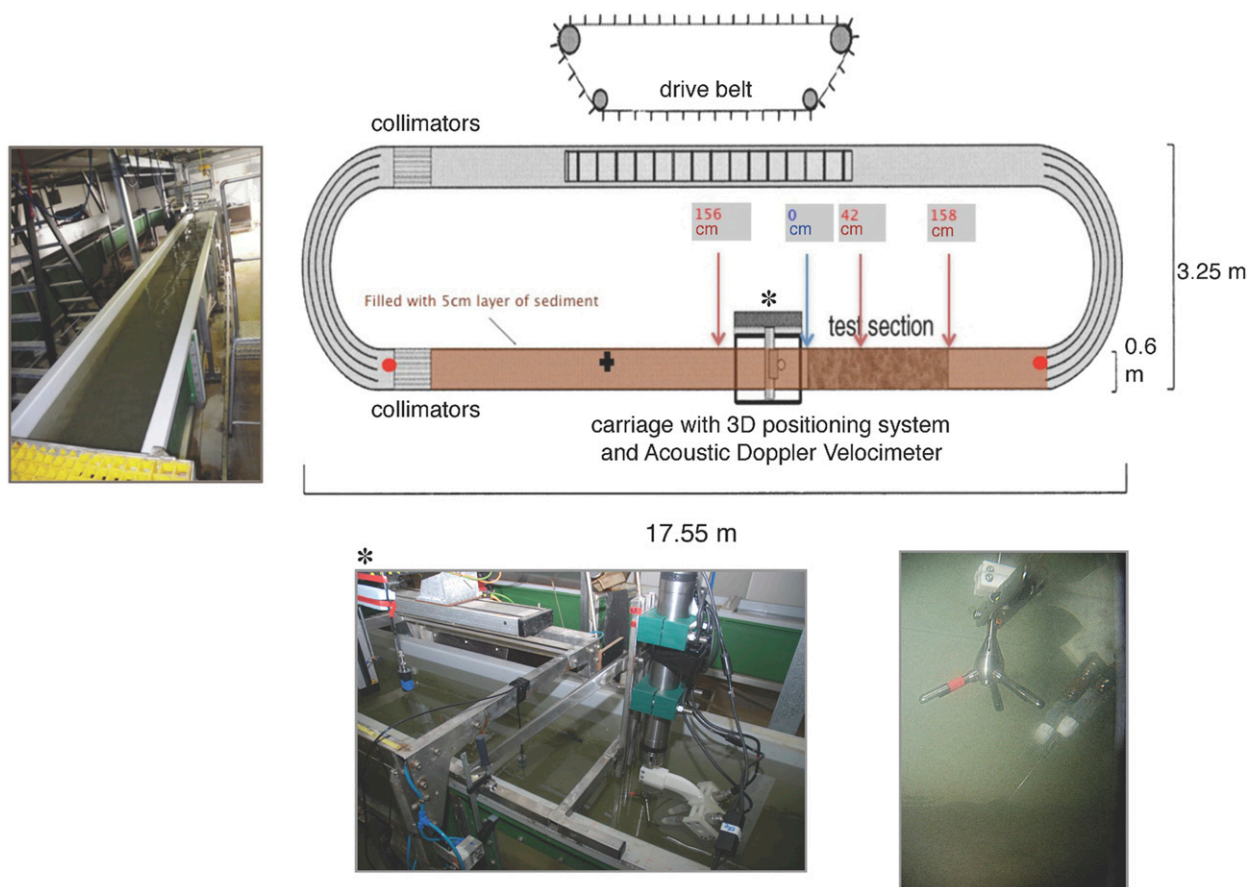


FIG. 1. Pictures of (top left), (bottom left) the racetrack flume facility at NIOZ-Yerseke and (bottom right) the EC system used for the experiments. (top right) A schematic of the flume: the dark brown shading indicates the straight working area, the red arrows indicate the location where the O_2 microprofiles were performed, the blue arrow indicates the location where EC measurements were performed (position 0), the two red dots mark the position of the optodes for monitoring the O_2 and temperature in the flume, and the cross indicates where the incubations were performed. Scheme adapted from Bouma et al. (2005).

anoxic flume water at the same temperature and salinity. The ADV sampling volume was located 15.7 cm below the probe and had an hourglass shape with diameters and height of 14 mm (Lohrmann et al. 1994). However, the sampling volume was assumed to be cylindrical with the same dimensions, from which the distance to the O_2 microelectrode tip was defined. The EC system was battery powered and suitably grounded to avoid electrical noise. The ADV stem was positioned at 7-cm depth, while the amplifiers and oxygen sensors were almost completely submerged, with the tip of the oxygen sensors and the ADV sampling volume at a depth of 22 cm (see Fig. 1).

c. Flux calculations

The O_2 concentrations and velocities were sampled at 64 Hz and subsequently averaged (simple average) to 8 Hz to improve the signal quality while still resolving the high-frequency eddies (as evaluated from the

obtained velocity power density spectrum). Velocity spikes were removed and interpolated between neighboring data points (as described in Goring and Nikora 2002) with an ADV beam correlation threshold of 70% (Elgar et al. 2005).

The measured vertical velocity (w) and O_2 concentration (C) can be separated into a fluctuating (prime) component and a mean (overbar) component as $w = w' + \bar{w}$ and $C = C' + \bar{C}$ (Reynolds 1895), respectively. The mean values were calculated by applying a moving average filter with a window length of 60 s (Moncrieff et al. 2004); this was sufficient to include the frequencies that contributed to the vertical flux (as discussed below). The O_2 fluxes (F) were calculated as $F = \overline{w'C'}$ [see Lorke et al. (2013) for a complete derivation] over each 5-min interval, which represents a good balance between a clear data visualization and high temporal resolution.

To determine the time shift needed to align the time series, C' and w' were shifted relative to one another

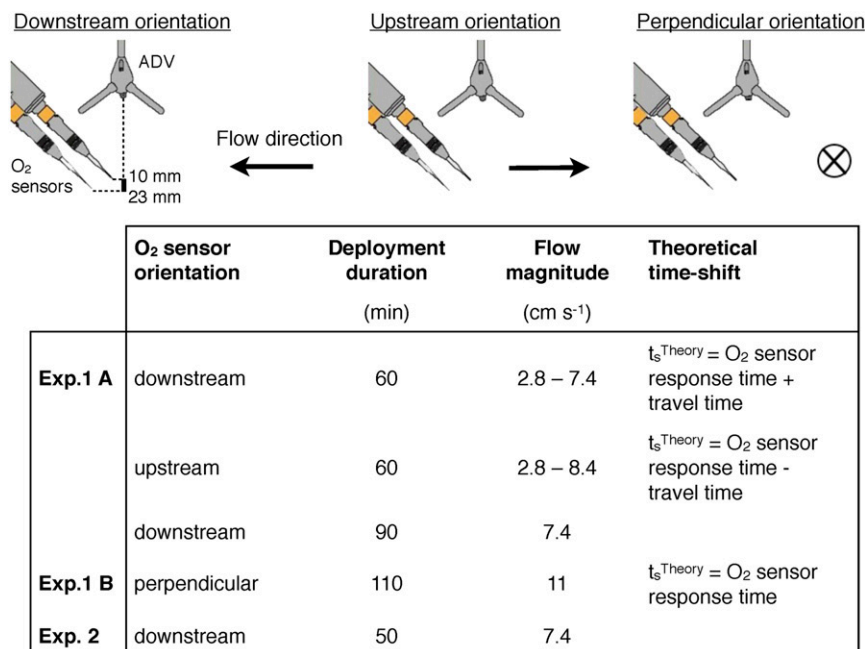


FIG. 2. (top) The EC system configuration and orientations used for the three experiments summarized here. The t_s^{Theory} is the expected displacement of the time series w' and C' , given by the sum of the O₂ sensors response and the travel time of the water parcel. An approximate estimation of the cross correlation needed for the alignment of the time series was calculated according to the t_s^{Theory} reported in the table.

using a 0.125-s step size (as defined by the 8-Hz sampling rate) over a window from 0 to 4 s for the respective 5-min interval (or sample window). The highest cross correlation (absolute value) in this window was used to calculate the EC flux for each sample. The statistical significance of the cross correlation was evaluated by calculating the probability of receiving the same correlation (i.e., the same flux) from a random dataset, by using the MATLAB 7.10.0 (R2010a) *corrcoef* function (Holtappels et al. 2013). The threshold for a significant flux was set to $p < 0.05$.

d. Experiment setup

Three specific flume experiments were conducted. Experiments 1A and 1B used two O₂ sensors with similar response times (90% response time; $\tau_{90} = 0.35$ s; stirring sensitivity = ~1%). The sensors were positioned at different distances (10 mm, referred to as “proximal”; and 23 mm, referred to as “distant”) from the edge of the cylindrical ADV sampling volume. The EC system was oriented downstream, upstream, or perpendicular to the flow direction (Fig. 2); these three configurations were expected to result in distinct and different time shifts (t_s) for the optimal correlation between w' and C' .

Experiment 2 was conducted to compare two O₂ sensors with different response times of 0.35 and 3.00 s

(for the latter, stirring sensitivity = ~0%) (Figs. 3b,c). Both sensors were positioned at 10-mm distance from the velocity sampling volume. See Fig. 2 for the deployment summary.

e. O₂ microsensor profiles and chamber measurements

Diffusive O₂ uptake (DOU) was determined from O₂ profiles across the sediment–water interface obtained using an O₂ microelectrode (tip diameter of 50 μm and $\tau_{90} < 5$ s) positioned by a motor-controlled micromanipulator. DOU was determined from the linear concentration gradient just beneath the water–sediment interface using Fick’s first law of diffusion (Glud 2008). The porosity at the sediment–water interface was assumed to be 0.7, and the molecular diffusion of O₂ was calculated from temperature and salinity based on Soetaert et al. (2012). Measurements were conducted at three different positions in the working section of the flume (0.4 and 1.5 m downstream, and 1.5 m upstream from the EC device) and at three flow velocities (18, 7.1, and 2.7 cm s⁻¹). Furthermore, the total oxygen uptake (TOU) of the sediment was determined via chamber incubations, which were performed 4 m upstream from the EC system (Fig. 1). Three acrylic chambers (\varnothing : 11 cm; height: 10 cm) were gently pushed into the

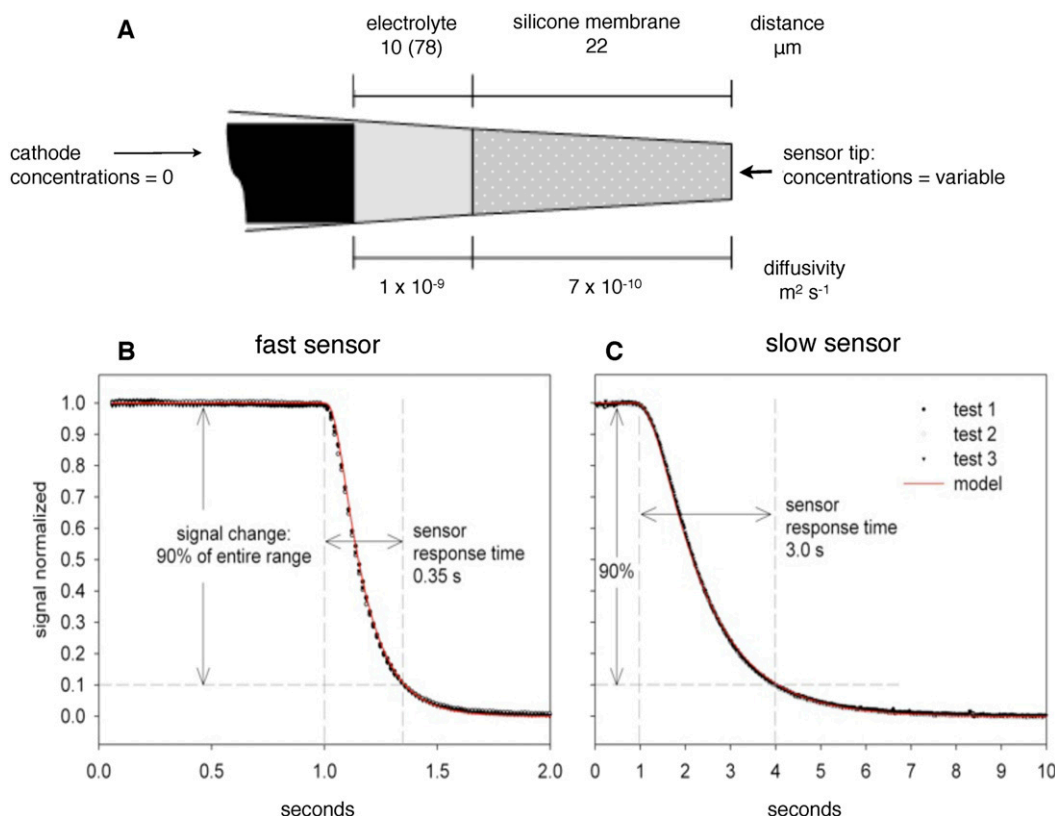


FIG. 3. (a) Schematic drawing of an O₂ microsensor, showing the dimensions and diffusivities as used in the numerical model. (b),(c) Response times of a slow (3.00 s) and a fast (0.35 s) O₂ microelectrodes used for experiment 2 in the flume. The response time is defined as the time required to τ_{90} .

sediment, and the O₂ concentration was continuously logged during the ~20-h incubations (Firesting oxygen optode, PyroScience GmbH). TOU rates were computed by linear regression of the O₂ time series as a function of time. The O₂ levels never declined more than 20% from the initial value during the incubations. Both TOU and DOU measurements were performed 4–8 days after the sediment and filtered seawater had been introduced to the flume.

f. Numerical model of a Clark-type microelectrode

To further assess the effect of sensor response time, we developed a numerical model of an oxygen microsensor using the finite element program COMSOL Multiphysics 4.3 (<http://www.comsol.com>). The O₂ flux to the cathode was described using a one-dimensional diffusion model with two domains, membrane and electrolyte (Fig. 3a) (Glud et al. 2000), with O₂ diffusivities of 0.69 and 1.00 × 10⁻⁹ m² s⁻¹, respectively (Gundersen et al. 1998). The maximum element size in either domain was 0.05 μm. The concentration at the cathode was set to zero, whereas the input concentration at the tip was determined by a prescribed time series of

realistic O₂ concentrations. The oxygen distribution across domains was calculated using a time-dependent solver. The output concentrations C_{out} of the sensor model were calculated from the modeled flux at the cathode J_{cath} (both time dependent) and multiplied by a calibration factor derived from the ratio of mean input concentrations at the sensor tip \bar{C}_{in} and the respective mean flux at the cathode:

$$J_{\text{cath}}(t) \frac{\bar{C}_{\text{in}}}{J_{\text{cath}}} = C_{\text{out}}(t). \quad (1)$$

A first step was to assess the effect of sensor response time on the fluxes measured during experiment 2. Therefore, we applied the model to simulate the O₂ flux as recorded by the fast ($\tau_{90} = 0.35$ s) and slow ($\tau_{90} = 3.00$ s) responding sensors, as in the experiment. The physical dimensions of the simulated slow and fast sensors were based on the dimensions of the custom-build sensors (Fig. 3a). The response time of the sensor models was estimated by applying abrupt changes of O₂ concentrations at their tip. The model was validated by comparing modeled and measured response times (Figs. 3b,c).

Subsequently, in order to investigate the potential bias of especially the fast sensor ($\tau_{90} = 0.35$ s), we generated an artificial time series of O_2 oscillations and used it as input into the numerical simulation. To approximate realistically high-frequency O_2 oscillations, we used a time series of measured vertical velocity fluctuations and multiplied the values with the ratio of the standard deviations of C' over w' [$\text{std}(C')/\text{std}(w')$] in order to scale down the amplitude, so that the standard deviation matches those of the measured O_2 concentration fluctuations. Afterward, the time series was multiplied by -1 for a maximum negative correlation with the vertical velocity fluctuations (i.e., a downward flux).

For our model results, the instantaneously responding sensor was considered to be the baseline flux, that is, the flux obtained considering no distance or response time difference between velocity and O_2 concentration measurements for experiment 2. With the same approach, we were also able to simulate a fast-response time sensor of 0.1 s.

g. Theoretical correction with frequency-dependent dampening correction

The sensor response time can potentially dampen the signal from turbulent oxygen fluctuations, with increased dampening occurring at higher frequencies. This leads to a signal loss that can be estimated by spectral analysis (McGinnis et al. 2008). One procedure is to apply a frequency-dependent transfer function (Eugster and Senn 1995; Gregg 1999) to the O_2 -velocity co-spectrum $S_{O_2}^{\text{obs}}$ ($\text{mmol m}^{-2} \text{s}^{-1}$) to arrive at an enhanced co-spectrum:

$$S_{O_2}^{\text{Corr}}(\omega) = (1 + \omega^2 \tau^2) S_{O_2}^{\text{obs}}, \quad (2)$$

where ω is the wavenumber [$\omega = 2\pi f$, where f is frequency (Hz)] and τ (s) is the $1/e$ ($\sim 63\%$) response time. We integrated the enhanced co-spectra to estimate the signal loss (expressed as a percentage of the flux) resulting from the slower sensor response time. The integration of the enhanced co-spectrum, however, should only be applied up to the highest frequency of the eddy contributions [e.g., estimated for a logarithmic boundary layer as in Table 1 in Lorrai et al. (2010)] and not beyond this point, as the transfer function also enhances the noise.

3. Results

a. EC measurements: Time-shift effect on measured O_2 fluxes

We compared the frequency spectra of the vertical component of the flow, measured for perpendicular and downstream orientation at the same flow velocity. These

were nearly identical (data not shown), excluding any anomaly potentially induced by the submerged instrument (e.g., flume-wall effect).

To illustrate the effect of the time shift on the EC fluxes, the results for experiments 1A and 1B are presented, including estimated time shifts and cross-correlation p values over the entire deployments (Figs. 4a–d). Different effects of time shift on the EC flux were observed according to sensor positions and orientations with respect to the flow direction (Figs. 4a–d). The calculated time shift (t_s) required for maximum correlation between w' and C' for both sensors increased with decreasing flow velocity (Fig. 4b). As expected (and according to Fig. 2), the sign of t_s reversed when shifting the sensor orientation from downstream to upstream (gray area in Fig. 4b). Furthermore, the O_2 sensor proximal to the velocity sampling volume had a less variable t_s compared to the sensor positioned at 23 mm, particularly under conditions of low flow velocity and perpendicular sensor orientation (Fig. 4b).

Oxygen fluxes measured during experiments 1A and 1B are listed in Table 1. On average the time-shift correction leads to an increase in the O_2 flux by $\sim 40\%$ and $\sim 30\%$ for the proximal and distant sensors, respectively. No significant difference is observed between the average fluxes from the two O_2 sensors (Table 1). However, as shown from Figs. 4c,d, the significance of the cross correlations increase substantially after applying the time shift, except for the 23-mm distant sensor case at perpendicular orientation. The most statistically robust correlations (with or without time-shift correction) are observed for the downstream orientation, compared to the upstream and perpendicular ones (Figs. 4c,d). The observed divergence between fluxes obtained for different sensors' orientations suggests that the applied time-shift correction procedure does not completely compensate for the biases related to the flow direction (i.e., the signal loss due to the undersampling of contributing eddies).

Reference measurements obtained by microprofiles and chamber incubations were carried out on different days at different positions in the flume and showed no significant differences (Table 2; Fig. 5), suggesting that the sediment diagenetic activity was in steady state throughout the duration of EC measurements. No significant diurnal variations of O_2 benthic fluxes were observed (the flume hall was kept dark except during the experiment setup), and O_2 concentrations along the water column and air–water exchanges appeared to be steady.

The closest agreement between EC fluxes and reference measurements ($\text{TOU} = -12 \pm 4$; $\text{DOU} = -8 \pm 2 \text{ mmol } O_2 \text{ m}^{-2} \text{ day}^{-1}$; Table 2) occurs when the EC

TABLE 1. Average EC fluxes for the three orientations of the oxygen sensors at 10- and 23-mm distance from the velocity sampling volume, before and after the time-shift correction. The increase of absolute value of the shifted flux compared to the nonshifted flux is included.

| | Nonshifted flux ($\text{mmol O}_2 \text{ m}^{-2} \text{ day}^{-1}$) | | | | | | Shifted flux ($\text{mmol O}_2 \text{ m}^{-2} \text{ day}^{-1}$) | | | | | |
|-------|---|---------------|--------------------------------|------------|---------------|--------------------------------|--|---------------|--------------|------------|---------------|--------------|
| | Downstream | | | Upstream | | | Downstream | | | Upstream | | |
| | All | Perpendicular | Increase of absolute value (%) | All | Perpendicular | Increase of absolute value (%) | All | Perpendicular | Increase (%) | All | Perpendicular | Increase (%) |
| 10 mm | -10 \pm 2 | -8 \pm 1 | -2 \pm 1 | -5 \pm 2 | -16 \pm 3 | 37 | -5 \pm 2 | -5 \pm 1 | 11 | -8 \pm 3 | -5 \pm 1 | 60 |
| 23 mm | -12 \pm 2 | -5 \pm 1 | -3 \pm 2 | -5 \pm 4 | -22 \pm 3 | 45 | -5 \pm 4 | -3 \pm 2 | 16 | -7 \pm 2 | -3 \pm 2 | 0 |
| | | | | | | | | | | | | 37 |
| | | | | | | | | | | | | 28 |

However, in this case, the cross correlation is less well defined, especially when the sensor is farther away from the velocity sampling volume (Fig. 6b). Finally, when the sensors are positioned perpendicular to the flow velocity direction, the cross correlation is irregular with no clear maximum for the distant sensor (Fig. 6c). The poorly pronounced cross-correlation peaks observed for the perpendicular orientation suggest that the magnitude of the flux is remarkably smaller than the flux obtained for the downstream and upstream orientation (Figs. 6a–c), confirming our previous analysis (Figs. 4c,d; Table 1).

Furthermore, the cross correlations for upstream and perpendicular orientations indicate that a closer sensor leads to more robust flux estimates, therefore suggesting that a time shift does not fully compensate for the 23-mm distance between sampling volumes. This is additionally indicated by the identical (and low) significance of the cross correlations before and after the time-shift correction obtained for the farther (23 mm) sensor in the perpendicular case (Fig. 4d).

Next, we compared the observed time shift (t_s^{Obs}) to the theoretical time shift (t_s^{Theory}) for each flow direction and sensor distance, for experiments 1A and 1B. The t_s^{Theory} is the expected displacement of the time series w' and C' , given by the sum of O_2 sensors' response and travel time of the water parcel (depending on the distance between the sensors, flow velocity, and direction) (Fig. 2). As expected, t_s^{Theory} and t_s^{Obs} strongly depend on the instrument orientation (Fig. 6d). The applied time-shift correction procedure (i.e., maximum cross-correlation shift) closely matches the theoretically expected time shift. However, a noticeable discrepancy (34%) between t_s^{Obs} and t_s^{Theory} is observed for the perpendicular orientation at 23 mm, for which the observed time shift also exhibits high variability (Fig. 6d).

c. Numerical model of a Clark-type microelectrode

As the time delay between w' and C' is not only caused by the traveling time but also by the response time of the O_2 sensor, we evaluate the effect of the fast and slow O_2 sensors used in experiment 2 on the flux estimation by means of numerical model simulations. The maximum cross correlation between vertical velocities and model sensor signals are obtained for time shifts of 0.2, 0.4, and 1.3 s for response times of 0.10, 0.35, and 3.00 s, respectively, at a flow velocity of 7.4 cm s^{-1} and a sensor spacing of 10 mm (Table 3). The time shifts of measured and modeled slow sensor agree, indicating that a significant part of the time shift can be attributed to the response time of the sensor.

By using the artificial O_2 time series as input for the sensor model, it was possible to test the signal loss of sensors with different response times (infinitely fast = 0 s,

TABLE 2. Flume benthic oxygen fluxes obtained between 8 and 14 Feb 2012 at different positions and flow speed by microsensor profiling and chamber incubations (EC flux results in Table 1 refer to 14 Feb 2012).

| Date | Flow magnitude (cm s^{-1}) | Position (cm) | DOU ($\text{mmol O}_2 \text{ m}^{-2} \text{ day}^{-1}$) |
|---------------|---------------------------------------|---------------|---|
| 10 and 11 Feb | 18 | 158 | 9.8 ± 1.2 ; $n = 8$ |
| 13 Feb | 7.1 | 158 | 8.0 ± 0.9 ; $n = 3$ |
| 12 Feb | 2.7 | 158 | 7.6 ± 0.4 ; $n = 4$ |
| | 2.7 | -156 | 8.4 ± 1.0 ; $n = 3$ |
| | 2.7 | 40 | 7.8 ± 1.2 ; $n = 3$ |
| | 2.7 | 158 | 7.3 ; $n = 1$ |
| | | | TOU ($\text{mmol O}_2 \text{ m}^{-2} \text{ day}^{-1}$) |
| 8–14 Feb | | (see Fig. 1) | 12 ± 2 ; $n = 3$ |

ultrafast = 0.1 s, fast = 0.35 s, and slow = 3.00 s). We compared the fluxes obtained with different response times to the fluxes obtained by an ideal (i.e., instant response) sensor (Table 3). The O_2 flux is reduced by 45% and 65% for the fast and slow O_2 sensors, respectively, while an ultrafast sensor with a 0.1-s response time would have a flux loss of 16%. Note that these results are specific to a flow velocity of 7.4 cm s^{-1} and the given flume conditions.

d. Theoretical correction with frequency-dependent transfer function

The O_2 spectra can be “rebuilt” to account for signal loss due to the sensor response time (McGinnis et al. 2008), knowing that the slower response time dampens the turbulent fluctuations, with increased dampening occurring at higher frequencies. Therefore, we applied the correction using Eq. (2) to the dataset (experiment 2) utilizing the 3-s response time sensor.

The calculated correction is shown in Fig. 7 as a function of the frequency of the O_2 oscillations.

The spectral corrections become quite substantial when sampling high frequencies with a slow sensor. However, the turbulent eddy contributions decrease with increasing frequency (as shown in Fig. 8 and discussed below), so while these corrections seem large, they are less (but still) significant when rebuilding the cospectra.

Figure 7b shows the flux underestimation for the 7.4 cm s^{-1} case (experiment 2) as a function of sensor response time, which was derived by multiplying the O_2 spectra with the correction factor as a function of frequency. The average fluxes were calculated for each hypothetical response time by integrating the flux between the limits assumed to be defined by the flux-contributing range (5–0.1 Hz; see Lorrai et al. 2010). The results are not sensitive to the lower-frequency

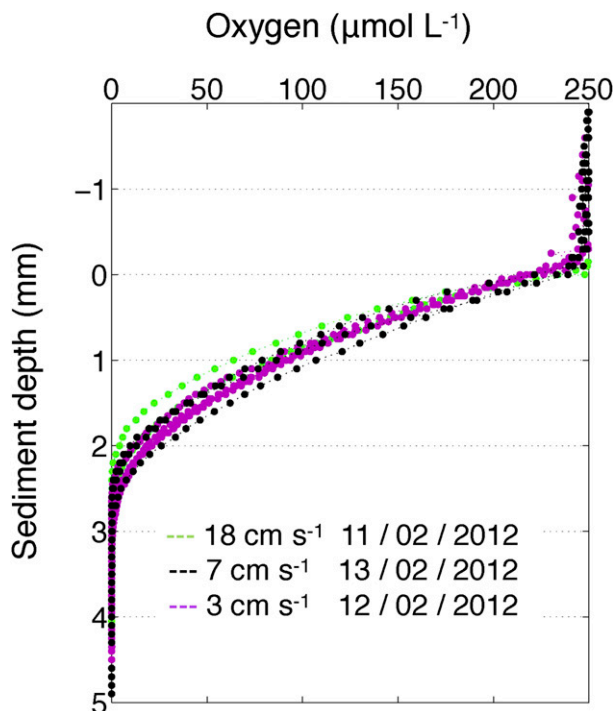


FIG. 5. The O_2 profiles performed in the flume at 158 cm (see Fig. 1) over 4 days at different flow velocities. The O_2 penetration depth ranged between 2.5 and 3 mm below the sediment–water interface.

integration limit, as the correction quickly decreases for lower frequencies; however, it is very sensitive to the high-frequency limit. The correction has a tendency to amplify high-frequency noise, so that integration beyond the high-frequency limit may introduce artefacts. Therefore, this example is only valid for the 7.4 cm s^{-1} case, as the flux-contributing eddy range (including the inertial subrange) will shift with changing velocity. For the case of the 3-s response time, we estimate that a 67% correction would be needed. This is very close to the value obtained from the O_2 sensor model results (Table 3).

4. Discussion

The assessment of turbulent benthic solute exchanges with an EC system requires instantaneous measurements of velocities and a scalar at the same point. This is practically impossible to achieve with the currently available instrumentation, that is, the combination of an ADV and a fast-responding sensor (here, we examined the most used Clark-type microelectrode for dissolved O_2 measurements). Previous work on aquatic EC measurements (McGinnis et al. 2008; Lorrai et al. 2010) has described how the flux measurements are systematically biased due to the different response time and the physical distance of the sensors. The authors introduced

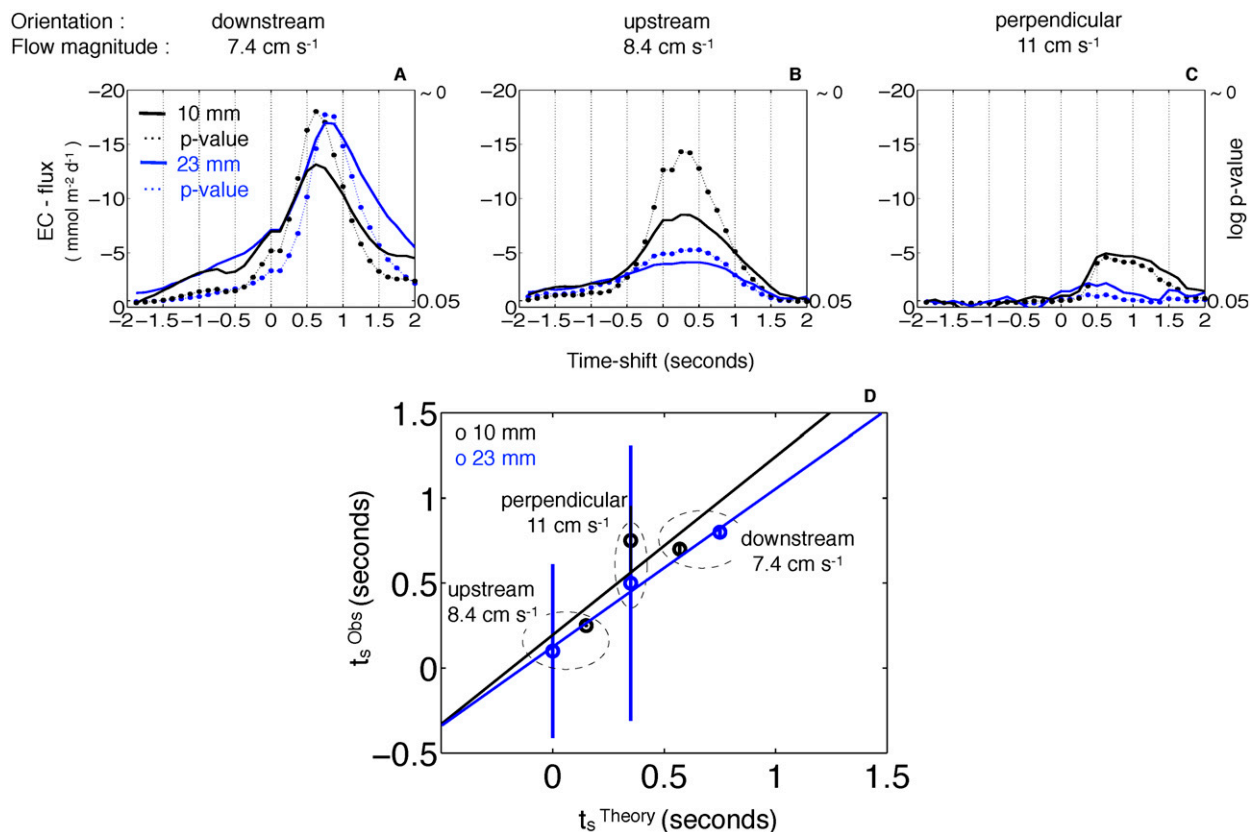


FIG. 6. (a)–(c) Cross-correlation functions between the velocity and O_2 time series (solid line) and respective p values (dotted line), for sensors positioned at 10 and 23 mm from the center of velocity sampling volume (black and blue, respectively). Data are shown for one representative sample window for each orientation at comparable flow velocities. (d) Average and error bars of observed time-shift (t_s^{Obs} , time-shift of w' vs C' needed to achieve the maximal correlation) vs the predicted theoretical time-shift (t_s^{Theory} , sum of traveling time and sensor response time) for each orientation at comparable velocities, for experiments 1A and 1B. The lines indicate linear regression.

correction procedures, however, that were based only on theoretical assumptions. We therefore designed ad hoc experiments in controlled environments (flume tank) and a model of a Clark-type microelectrode to systematically study the errors caused by sensor displacement and response times and to evaluate the correction procedures at hand.

a. Time shift, sensor displacement, and flow direction

By analyzing EC measurements in the well-constrained conditions of a large-scale flume facility, we could assess O_2 fluxes for different sensor positions (i.e., orientation and distance of O_2 sensors with respect to the center of the ADV sampling volume) at different flow magnitudes and directions. The dependency of estimated fluxes on sensor position is an aspect that has not been addressed in aquatic EC studies. While some variability of measured O_2 fluxes is expected to occur due to natural hydrodynamic conditions (Holtappels et al. 2013) and bottom heterogeneity (e.g., Rheuban and Berg 2013), our results indicate that some of the flux

variability is also related to the orientation of the sensor with respect to the flow direction. This effect is not easily resolved in natural environments, where a highly intermittent flow field and spatial variations may mask the artifacts in benthic flux estimates. However, when averaged over longer time scales, the average flux might also be less sensitive to these potential issues.

Results from the flume investigation here reveal that when the flow is parallel to the sensor (i.e., downstream or

TABLE 3. Model prediction of a slow (3.00 s), fast (0.35 s), ultrafast (0.1 s), and artificial and infinitely fast (0.0 s) O_2 sensor signal, and percentage of loss flux as referred to the 0-s response.

| Sensor response time (s) | Time shift for max cross correlation (s) | Flux modeled (mmol O_2 m ⁻² day ⁻¹) | Loss of flux (%) |
|--------------------------|--|--|------------------|
| 0 (artificial) | 0.0 | –45 | 0 |
| 0.10 | 0.2 | –38 | 15 |
| 0.35 | 0.4 | –20 | 45 |
| 3.00 | 1.3 | –16 | 65 |

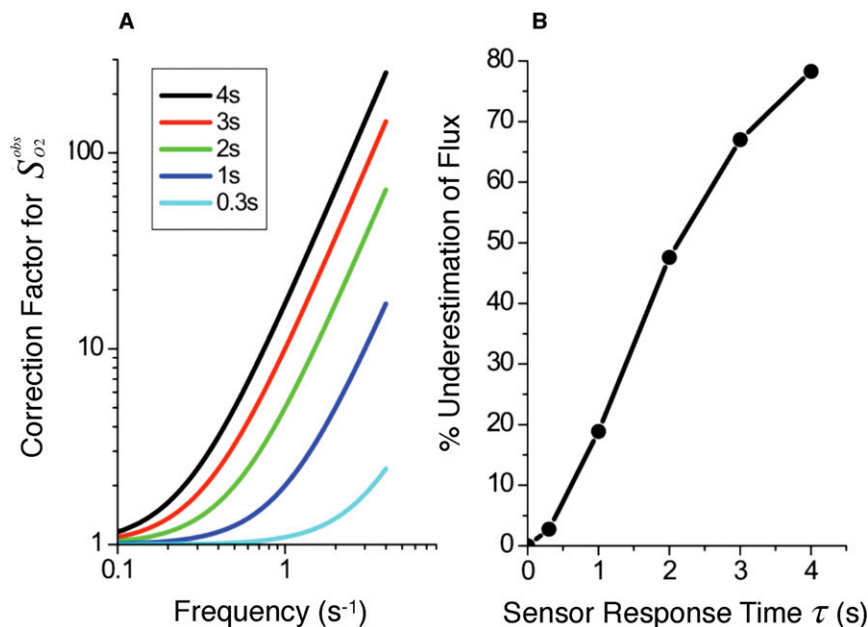


FIG. 7. (a) The frequency-dependent correction factor ($1 + \omega^2\tau^2$) applied to the O_2 spectrum. (b) Underestimation of the flux as a function of sensor response time.

upstream orientation; Fig. 2), the turbulent structures pass the O_2 and velocity sensor with a delay that depends on the distance between sampling positions for w' and C' and the response time of the O_2 sensor. However, an increasing distance between sampling volumes lowers the probability of sampling the same eddy structure, as evident by the more statistically significant correlations between w' and C' obtained in our experiments for the proximal sensor (10 vs 23 mm). Thus, it is advantageous to position the O_2 sensor as close as possible (<1 cm) to the edge of the velocity sampling volume, but taking care that it does not enter the sampling volume. If larger diameter sensors are used, these new dimensions must be considered with respect to the potential interference.

If all the orientations of the O_2 sensors with respect to the flow are considered for the flux average (equal deployment time of 60 min for downstream, upstream, and perpendicular), then we observed similar EC fluxes compared to chambers and microprofile estimates, only after applying the time-shift correction ($EC = -9 \pm 4$; $TOU = -12 \pm 4$; $DOU = -8 \pm 2$ mmol O_2 m $^{-2}$ day $^{-1}$).

However, the best agreement with TOU measurements occurs when the EC instrument is aligned downstream with or without time-shift correction, (EC shifted = -16 ± 3 ; EC nonshifted = -10 ± 2 mmol O_2 m $^{-2}$ day $^{-1}$, for sensor distance of 10 mm), while when the orientation is upstream, the flux magnitude and the significance of the correlation decrease substantially. A flux magnitude decrease for the upstream orientation was observed for both proximal and distant O_2 sensors and

holds true after applying the time-shift correction (EC shifted = -9 ± 1 mmol; EC nonshifted = -8 ± 1 mmol O_2 m $^{-2}$ day $^{-1}$, for sensor distance of 10 mm). The weaker correlation between w' and C' at upstream orientations and the reduced effectiveness of the alignment procedure could be caused by the sensors and amplifiers affecting the flow and the turbulent structures. Finally, the perpendicular orientation results in underestimated fluxes by 70% compared to fluxes measured when the O_2 sensors are oriented downstream (EC shifted = -5 ± 1 ; EC nonshifted = -2 ± 1 mmol O_2 m $^{-2}$ day $^{-1}$, for

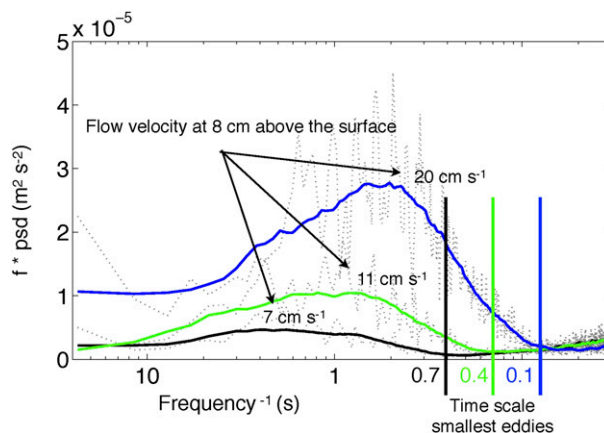


FIG. 8. Variance-preserving spectra of w corresponding to flow velocities of 7 (black), 11 (green), and 20 cm s $^{-1}$ (blue) measured in the flume at 8 cm above the surface. The curves, smoothed by adjacent averaging over 50 data points, indicate for each flow field the lifetime range of turbulent eddies.

a sensor distance of 10 mm). Flow directions perpendicular to the sensors line are indeed more challenging, given the higher probability for the ADV and O_2 sensors of measuring decoupled signals. Moreover, at perpendicular orientation, the travel time alignment is not applicable, so that the highest frequencies contributing to the flux (i.e., eddy lengths smaller than the distance between the sensors) are more likely to be missed. Clearly, the more robust correlations obtained for the proximal sensor confirm that a closer distance reduces this decoupling effect. Note, however, that such a poor correlation is not necessarily expected in the field, because of a much larger variability in the magnitude and direction of the flow. However, these results do suggest that the instrument should ideally be aligned in the main direction of the flow.

Given that flume walls have a negligible boundary layer thickness (~ 1 cm) and we did not observe any anomaly in the velocity frequency spectra during the entire experiment, we exclude the possibility that the poor cross correlation at perpendicular orientation is caused by sidewall effects. However, the dimensions of the racetrack flume are appropriate only for the scales of the hydrodynamic processes corresponding to the higher-frequency turbulent structures, which represent a restricted range of field applications.

In summary, the flow direction appears as an important factor influencing EC flux measurements. The time-shift correction does not lead to a uniform flux estimations between downstream, upstream, and perpendicular orientations, especially because the latter case cannot be fully corrected because of the undersampling of flux-carrying eddies. For downstream configurations, the time-shift correction can correct this bias, although increasing displacements increase the mismatch of the spatial overlay of corresponding velocity–concentration pairs and may constitute a weak point of this method, especially under nonuniform flow conditions. However, if all three orientations from our flume data are equally considered in the flux average, then the time-shift correction improves the confidence and precision of the obtained flux estimation up to 30% (Table 1). Applying a time shift is thus a convenient procedure, and it was confirmed that it corresponds closely to the sum of traveling time and O_2 sensor response time (discussed below) for the case of downstream and upstream orientation of the EC system.

b. Flux underestimation due to microsensor response time

Similar to what has been assessed for the atmospheric application of EC (see Foken et al. 2012), we demonstrate that for aquatic EC, corrections must be introduced when the maximum frequency response of a sensor is less than the highest frequency of the

turbulent eddies responsible for the flux (McGinnis et al. 2008). By applying a 1D model to simulate O_2 diffusion in a microsensor and imposing a vertical velocity signal with the same amplitude of the O_2 signal, we were able to estimate the fluxes measured by microsensors with different response times. The simulations showed that an O_2 sensor with a 3.00-s response time (thus, 10 times slower than sensors usually used for EC measurements) leads to an underestimation of 65% of the true flux (i.e., no response time and no signal dampening) and a sensor with a 0.10-s response time results in a 15% loss for the conditions evaluated (7.4 cm s^{-1}). This indicates that high-frequency signal loss, intrinsic to any electrochemical electrode measurement, cannot be corrected by the time-shift procedure alone. The dampening effect on vertical solute transport estimations by EC can be significant, depending on the frequency of the turbulent processes involved. At our flume conditions, a sensor with a τ_{90} of 0.4 s would resolve nearly the complete flux-contributing eddy ranges for a flow velocity of $<11 \text{ cm s}^{-1}$ (measured at 8-cm height); however, the sensors are not sufficiently fast for flow velocities $>20 \text{ cm s}^{-1}$ (Fig. 8).

In summary, at lower flow velocities, the signal loss is less relevant because there are fewer flux contributions from high frequencies. Conversely, the portion of missed turbulent contributions becomes increasingly relevant with slower-responding O_2 sensors and increasing velocity.

Using the theoretical time scales of flux-contributing eddies (approximated, e.g., in Lorrai et al. 2010), it is possible to have an a priori estimate of the spectral range that is affected by sensor response time and correct it with the theoretical approach described here. This procedure is therefore recommended when turbulence regimes appear critical with respect to the O_2 sensor response time.

5. Conclusions and recommendations

Our flume experiments revealed that a closer distance between the ADV sampling volume and the O_2 sensor leads to more statistically significant correlations between w' and C' , thus to a better EC flux estimation. We showed that applying a time-shift correction improves the precision of the EC flux estimates and that the correction is particularly successful when the O_2 sensors are oriented downstream.

We found that when the current direction approaches perpendicular (Fig. 2) to the O_2 sensor-to-measuring volume line, then the likelihood that the signals are decoupled increases compared to other current directions (i.e., upstream or downstream). The effect is particularly exaggerated when the physical distance

between the sampling locations is increased, which translates into poor correlations and underestimated fluxes. For example, a somewhat high turbulence regime (e.g., friction velocity $> 0.01 \text{ m s}^{-1}$) combined with a sensor separation distance of 2 cm can lead to flux underestimation approaching 80%. Therefore, if the flow direction is expected to change during the deployment of an EC system, then the O_2 sensor tip distance from the velocity sampling volume should be as small as possible ($< 1 \text{ cm}$). Accordingly, one may also consider discarding the fluxes obtained for orientations other than downstream/upstream.

Slow sensors may also lead to signal loss. We demonstrate how the response time of the O_2 sensor is crucial to capture the O_2 fluctuations defined by turbulent processes that ultimately govern the benthic O_2 fluxes. Therefore, the error associated with a response time of the O_2 sensor depends on the turbulence levels, which define the eddy time scales that contribute to the vertical flux.

The spatial scale of this experiment setup is not entirely representative of most natural environments, where advective processes can lead to extremely variable and complex interactions with the seafloor (e.g., unsteady advection due to internal or surface gravity waves). Thus, compared to EC measurements in natural systems, our results are likely to be more sensitive to signal losses at the higher frequencies of the inertial subrange, resulting in flux underestimations.

Nevertheless, a greater underestimation of the flux is expected for any EC application when using a sensor with a response time of more than 0.2 s (the usual recommended τ_{90} for an EC measurement). In our case, we already observed a 15% signal loss from a sensor with $\tau_{90} = 0.1 \text{ s}$ for velocities of 7.4 cm s^{-1} ; thus, the reliability of the EC measurements in flows much higher than 20 cm s^{-1} (i.e., with frequency ranges $> 10 \text{ Hz}$) should be carefully considered. In settings with lower current speeds on the order of $2\text{--}10 \text{ cm s}^{-1}$, this effect is of less importance. Generally, it is beneficial to assess the O_2 sensor response time in relation to the expected flow prior to any in situ deployment.

Here, for the first time, we systematically investigate the bias in aquatic EC measurements resulting from 1) the physical separation between the O_2 sensor tip and the velocity sampling volume and 2) the O_2 sensor response time. The results promote awareness of these potential sources of measurement artifacts for field measurements, and allowed us to assess a potential EC flux underestimation resulting from these factors and to provide guidelines for deployment design and data treatment.

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