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Earth and Space Science



RESEARCH ARTICLE

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Key Points:

- We developed a portable instrumentation platform for measuring benthic fluxes in shallow aquatic environments by relaxed eddy accumulation
- Simultaneous measurements of dissolved oxygen fluxes by aquatic eddy covariance showed the reliability of the conditional sampling technique
- This work paves the way for exploring the dynamics of biogeochemical cycles in aquatic ecosystems at hourly time scales

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A New Technique for Resolving Benthic Solute Fluxes: Evaluation of Conditional Sampling Using Aquatic Relaxed Eddy Accumulation

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Abstract Solute exchange between the sediment and the water column strongly influences water quality in aquatic ecosystems. Among the techniques available for in situ observation, only aquatic eddy covariance (AEC) can resolve the hourly dynamics of benthic fluxes, but only for few substances like oxygen. In micrometeorology, relaxed eddy accumulation (REA), a technique based on high-frequency sampling conditioned by current direction, overcomes this limitation. We developed a portable prototype instrumentation platform to transfer REA to the sediment-water interface of shallow aquatic ecosystems. In order to evaluate the sampling system, we configured the prototype for measuring oxygen fluxes and comparing them to simultaneous AEC measurements. In a river and two artificial lakes, at depths 1–3 m and 0.2 m above the sediment, we measured 15 oxygen flux pairs in fully developed turbulence during 8–15 min periods, ranging from -150 to 50 $\text{mmol m}^{-2} \text{day}^{-1}$. The good agreement between REA and AEC fluxes ($R^2 = 0.94$) shows the reliability of the conditional sampling system. REA represents a promising technique for exploring the sediment contribution to biogeochemical cycles in aquatic ecosystems.

Plain Language Summary Microbial activity at the bottom boundary of aquatic ecosystems, such as lakes, rivers and coastal areas, influences their water quality. Microorganisms decompose organic matter and release nutrients and greenhouse gases to the overlying open water. In shallow aquatic ecosystems, the release from the sediment surface can vary rapidly, especially with currents, but so far, no technique succeeds in monitoring dynamic fluxes at hourly timescales for a broad range of substances. In micrometeorology, relaxed eddy accumulation is a well-established technique for monitoring the turbulent exchange of all kinds of compounds between land and atmosphere (including gas, aerosols and particles). Air is sampled conditionally for downward or upward motions and accumulated in separate containers. Concentration differences between sample pairs provide information on the fluxes. We developed a portable prototype, transferring this technique to quantify the exchange of nutrients and other compounds at the bottom boundary of shallow aquatic ecosystems. For its evaluation, we adapted the prototype to measure dissolved oxygen fluxes. The agreement of its measurements with fluxes measured by aquatic eddy covariance, our reference, shows the potential of this technique. It can now be used to quantify the exchange of major ions and of other substances including dissolved gases.

1. Introduction

The exchange between sediment pore water and the water column, that is, benthic fluxes, plays a major role in regulating the water quality and the ecology of aquatic ecosystems (Wang et al., 2020) and contributes to global biogeochemical cycles. The sediment-water interface is a hotspot of organic matter decomposition: nutrients and greenhouse gases produced during the mineralization of organic matter in anoxic pore water diffuses toward the water column. The benthic fluxes of a variety of dissolved substances have been explored, but their spatial variability and temporal dynamics remain poorly understood.

The organic matter content in the sediment, its biological activity, the concentration gradient at the water-sediment interface and near-bed currents are the main drivers of benthic fluxes (Lorke & MacIntyre, 2009). Over fine

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sediments particularly, currents reduce the thickness of the diffusive boundary layer, the thin lowest part of the benthic boundary layer and the bottleneck for solute exchange (Lorke & Peeters, 2006). Near-bed flows vary in magnitude and direction in response to external forcing, including tides (Reimers et al., 2020), wind-generated flows in lakes (Wüest & Lorke, 2003), and discharge variations in rivers (Kang & Lee, 2020). These variations modulate oxygen benthic fluxes at time scales of several hours (Chipman et al., 2016), or less for flow rate changes (Koopmans & Berg, 2015).

Existing techniques are unable to measure most benthic fluxes at the hourly timescale. Benthic chambers yield diffusive fluxes and underestimate fluxes in highly permeable sediments exposed to strong currents (Berg et al., 2013). Concentration gradient measurements across the sediment-water interface require stable diffusive boundary layer and hydrodynamic conditions (Davison et al., 1991); they are representative for a small sediment surface area. On the contrary, aquatic eddy covariance (AEC, Berg et al., 2003) has a typical footprint area of 100 m², since flow velocity and concentration are measured within the turbulent boundary layer (Berg et al., 2022). As a main limitation, it requires high frequency sensors (typically 10 Hz) and which do not perturb the flow. They are only available for suspended matter (the velocimeter itself, Kawanisi & Yokosi, 1997), oxygen (Berg et al., 2003), temperature and electrical conductivity (Crusius et al., 2008), nitrates (Johnson et al., 2011), hydrogen sulfide (McGinnis et al., 2011), hydrogen ions (Long et al., 2015) and fluorescent dissolved organic matter (Hu & Hemond, 2022).

Micrometeorologists overcame this difficulty with the relaxed eddy accumulation technique, in which the fast and separate sampling and sample accumulation of turbulent updrafts and downdrafts replace the fast concentration measurements of eddy covariance (Businger & Oncley, 1990). A simulation of REA fluxes by reanalyzing AEC measurements from a riverine lake showed the theoretical feasibility of adapting REA to the sediment-water interface (Lemaire et al., 2017). The challenges are the same as at the land-atmosphere interface. A first one is to isolate a small turbulent vertical flux perpendicular to a large horizontal advective flux over the sediment. This requires a precise alignment of the current velocimeter with the vertical and the high-frequency separate sampling of updrafts and downdrafts. A second one is to conserve accumulated samples correctly before analysis and to measure their concentrations with sufficient analytical precision.

This work addresses the first challenge and evaluates in situ the reliability of an aquatic REA conditional sampling system. The article presents a portable prototype for shallow aquatic environments, its configuration for benthic oxygen fluxes, their comparison to AEC reference fluxes, and a discussion of the technique potential range of use.

2. Theoretical Basis

Assuming a steady-state mass balance, negligible divergence of horizontal advection, that the sediment surface is the only source or sink of fluxes and homogeneous over the measurement footprint, the vertical turbulent flux of a solute can be formulated as the covariance of the fluctuations of the vertical velocity (w') and of the solute concentration (C') at any location in the turbulent boundary layer:

$$F = \overline{w'C'} \quad (1)$$

where the overbar denotes a temporal average and the prime a turbulent fluctuation obtained by Reynolds' decomposition (e.g., $w' = w - \bar{w}$). Equation 1 is directly used in AEC (Berg et al., 2003). In order to capture the entire range of flux-contributing eddies, measurement frequencies are often at least of 8 Hz. The absence of significant flux contributions at the highest resolved frequencies in the velocity and dissolved oxygen co-spectra, indicates that the sampling frequency is sufficient (Lorke et al., 2013). At the low-frequency end of the measured spectrum, the flux averaging time has to be much longer than the largest eddy time scale:

$$\tau = h_{\text{BBL}}/u \quad (2)$$

where h_{BBL} is the height of the benthic boundary layer in a stratified flow as in a lake or the water height in an unstratified flow, as in a river, and u^* is the current friction velocity. The averaging time is determined during post-processing and ranges from 5 to 30 min (Long, 2021).

REA, a micrometeorology technique, is based on a high frequency sampling conditioned by the direction of turbulent velocity fluctuations (Businger & Oncley, 1990). It relies on the same assumptions about matter transport as

eddy covariance. Air is pumped at a fixed flow rate during successive flux averaging periods. Depending on the vertical velocity direction, fast-switching valves collect updraft and downdraft samples in separate compartments. Their average concentrations at the end of the flux averaging period, \overline{C}_1 for updrafts and \overline{C}_d for downdrafts, along with the standard deviation of the vertical velocity σ_w and a dimensionless empirical coefficient b , yield the flux:

$$F = b\sigma_w(\overline{C}_1 - \overline{C}_d) \quad (3)$$

In order to increase the concentration difference and improve the flux quantification limit, smaller eddies of vertical velocity lower than a velocity threshold w_0 are discarded; this also preserves the switching valves from fatigue. The value of the coefficient b has received considerable attention. With the assumption of cospectral similarity, it is independent of the transported compound. It can be estimated experimentally by combining Equations 1 and 3, either with simultaneous eddy covariance and REA measurements (Pattey et al., 1993), or by the reanalysis of eddy covariance data to simulate the average concentrations of updrafts and downdrafts (Businger & Oncley, 1990). Without velocity threshold, the value b_0 of the coefficient is close to its theoretical value for joint Gaussian distributions of velocity and concentration, 0.63 (Wyngaard & Moeng, 1992). By reanalyzing eddy covariance data, Ammann and Meixner (2002) estimated an empirical expression for the decrease of the coefficient b with the velocity threshold w_0 , with coefficients b_0 , b_∞ and a depending on atmospheric stability:

$$b = b_\infty + (b_0 - b_\infty)\exp\left(-a\frac{w_0}{\sigma_w}\right) \text{ for } 0 \leq \frac{w_0}{\sigma_w} \leq 2, \quad (4)$$

Similar simulations at the sediment-water interface with AEC data yielded the following values for dissolved oxygen: $b_0 = 0.60$, $b_\infty = 0.152$ and $a = 1.35$, with a large interquartile range of 0.4–0.8 for b_0 (Lemaire et al., 2017). Both in the atmosphere and in aquatic environments, simulated REA fluxes showed best agreement with eddy covariance fluxes for a threshold velocity w_0 of $0.75\sigma_w$.

3. Materials and Methods

3.1. REA Prototype

Inspired from atmospheric REA setups (Bowling et al., 1998), the prototype addresses requirements concerning its stability on the sediment surface at a water depth up to 5 m, high frequency conditional sampling at a stable flow rate, correct separation of updraft and downdraft samples, the accumulation of a sufficient volume for chemical analysis (200 mL) during each of flux averaging periods of 30 min over deployments of several hours, and sample conservation before recovery from the accumulation compartments and laboratory analysis.

In order to avoid frame vibrations within the turbulent frequency range, the instruments are fixed to a topographic tripod (Noss et al., 2018). During each flux averaging period, an acoustic Doppler velocimeter (ADV Vector, Nortek AS, Figure 1) measures the three-dimensional current velocity at 8 Hz above the sediment surface and triggers the REA control system programmed in LabVIEW. The computer recovers velocity data and rotates the measured velocities to compensate for the imperfect leveling of the ADV with the sediment-water interface, then extracts the turbulent fluctuation of the vertical velocity w' (this processing is detailed in Section 3.3). If w' is larger than the velocity threshold w_0 , the fast response 2/2 solenoid valve corresponding to its direction opens and lets water flow through one of the 2.5 mm diameter perfluoroalkoxy alkane sampling tubes. Their inlets are syringe needles of 0.5 mm diameter, located 10 mm outside and at the height of the mid-point of the ADV measurement volume (14 mm height, 14 mm in diameter). Separate sampling tubes avoid mixing updraft and downdraft samples, especially since the flow in the tubes is laminar and not turbulent as in atmospheric REA.

In order to ensure the stability of the sampling flow rate of small volumes, water is not pumped as for atmospheric REA, but is moved by the pressure difference between the hydrostatic pressure at the inlets of the sampling tubes and the atmospheric pressure in the accumulation compartments, connected to the atmosphere through venting tubes. Manual valves clamp the sampling tubes to reduce the flow rate, which increases with water depth, to reach around 2 L hr⁻¹ for the desired accumulated volume.

If concentration differences between upward and downward sample pairs remain above the quantification limit, the samples are intended to be analyzed for all kinds of solutes commonly monitored in surface waters, beginning with major ions. In order both to avoid external contamination and to limit the contact of sampled water with

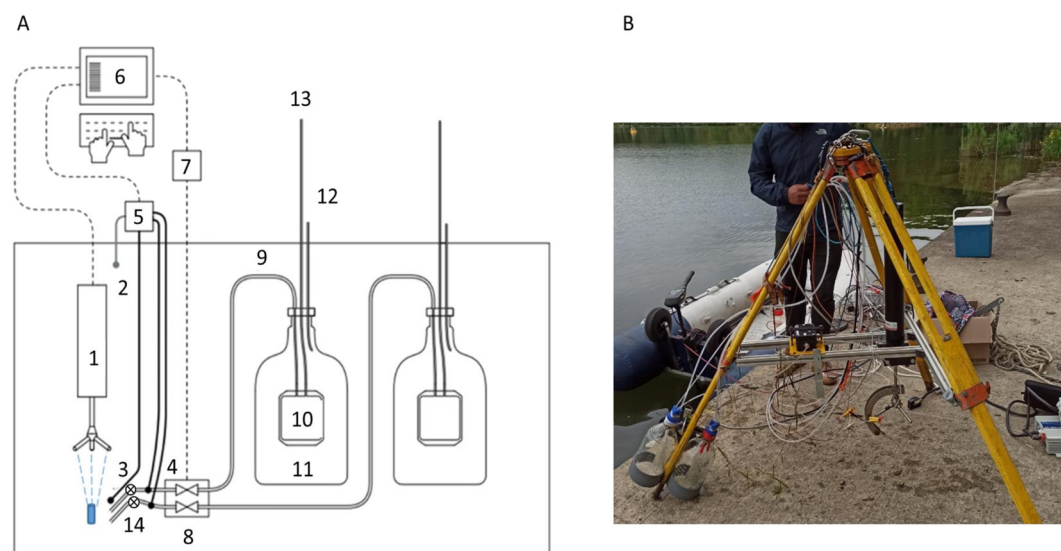


Figure 1. (a) REA and AEC setup for aquatic environments: (1) acoustic Doppler velocimeter with its measurement volume in blue; (2) temperature probe; (3 and 4) oxygen probes for AEC and inside each sampling tube; (5) oximeter; (6) control system; (7) on/off control module; (8) solenoid valves; (9) sampling tubes; (10) accumulation bags; (11) accumulation bottles; (12) venting tubes; (13) sample recovery tubes; (14) valves controlling sampling flow rate; (b) photograph of the setup.

air, which could cause organic matter degradation or reduced substances oxidation, the samples are accumulated in 250 mL polyolefin perfusion bags (inspired by Bowling et al. (1998)), inserted in 2 L bottles. The change in the sampled water composition should be negligible during flux averaging periods of less than 30 min, since the expandable bags are located underwater, at temperature and light conditions comparable to the sampling point.

Immediately after each flux averaging period, recovery tubes are opened and the samples are collected and refrigerated before chemical analysis. The velocity threshold is updated for the next flux averaging period ($w_0 = 0.75\sigma_w$).

3.2. Adaptation of REA Method for the Technical Evaluation of the Sampling System

AEC is the natural reference method for the evaluation of aquatic REA, like eddy covariance for atmospheric REA (Baker, 2000; Oncley et al., 1993). In this study the instruments for REA and AEC were fixed to the same frame (Figure 1).

Among the fluxes measurable with AEC, dissolved oxygen is easier to handle than, for instance, water temperature, which would require thermal insulation of the whole system. But the measurement of aquatic fluxes of dissolved gases is challenging, due to potential gas exchange in the accumulation bags and recovery tubes between water and air, the latter being necessary to maintain atmospheric pressure at the sampling tube outlet and a stable flow rate.

In situ measurements with fiber optodes avoid sample conservation and transportation before laboratory analysis, but the potential contact with the accumulation bags during sample recovery could damage them. We have therefore slightly downgraded the REA setup for the evaluation of the conditional sampling system: 50 μm diameter high speed micro-optodes (OXB50-UHS, PyroScience GmbH, response time <0.3 s) continuously monitor the oxygen partial pressure of water flowing through the sampling tubes, and not in the accumulation bags. Averaging concentrations measured during sampling mimics the mean concentrations in the bags. The distance between the sampling tube inlets and the oxygen probes is short, around 10 cm, in order to limit the attenuation of concentration fluctuations between successive samples during their intermittent transport through the tube.

For AEC, a 430 μm micro-optode (OXR430-UHS, PyroScience GmbH, response time <0.3 s) was used. The three optodes, plugged to an oximeter (Firesting, PyroScience GmbH) measured at 4 Hz. Since the oximeter deactivates its temperature measurements when it measures oxygen concentration at high frequency, water

temperature is measured before every flux averaging period to prepare the conversion of partial pressures into dissolved oxygen concentrations using a temperature-dependent Henry coefficient (Sander, 2015).

Unlike for AEC, the duration of flux averaging periods has to be fixed before REA sampling. They lasted 8–15 min and were longer in low-energy than in high-energy conditions.

3.3. Quantification of Oxygen Fluxes

3.3.1. Processing of Velocity Measurements

Wahl (2000) recommended to replace by interpolation ADV velocities with a signal-to-noise ratio below 15 dB or with a correlation between transmitted and received pulses below 70%. Velocity outliers are commonly filtered with the Phase-Space Threshold method (Goring & Nikora, 2002). But to drive REA water sampling, we could only use information that are provided by the ADV continuously, namely the beam correlation. At a given time step, if the ADV measured with low beam correlation, no water was sampled for REA. After the campaign, these bad quality velocities were replaced by linear interpolation for the AEC flux computation.

3.3.2. Reorientation of the Coordinate System

The prototype for aquatic REA is portable and designed for depths below 5 m. When it is placed on a sediment either imperfectly flat or susceptible to differential settling, the coordinate system of the current meter is to be aligned with the vertical. The double and triple angle rotations and the planar fit are the most common methods (Lorke et al., 2013; Wilczak et al., 2001). In our setup, both for AEC and REA, we used the double rotation, which requires shorter velocity timeseries than the planar fit. The control system calculates the mean current (\bar{u}) direction and cancels the mean vertical speed (\bar{w}) with successive rotations by angles α around the z -axis and β around the x -axis, using the average velocity components (\bar{u} , \bar{v} , and \bar{w}).

For eddy covariance, either atmospheric or aquatic, the rotation angles are computed after the campaigns from the velocities of each sampling period or of a range of successive periods. For REA, angles have to be determined before sampling air or water. For atmospheric REA, the angles are often computed once, after the anemometer is fixed on a “flux tower” (Pattey et al., 1993). Sarkar et al. (2020), who used portable atmospheric REA equipment, determined the angles at the beginning of each campaign from a velocity measurement period without air sampling, then used for each flux averaging period the angles computed from the velocities measured during the previous one. But during the development phase of our prototype, we alternated between velocity measurement periods without sampling, later called “prototype alignment periods,” and flux averaging periods of the same duration. In the following, “velocities” will refer to aligned velocities.

3.3.3. Extraction of the Turbulent Velocity Fluctuation

Another challenge for measuring turbulent fluxes is the presence of non-turbulent motions like waves (seiches, boat passage) (Berg et al., 2022; Long et al., 2015). They can temporarily drift the mean vertical velocity and perturb REA sampling. To prevent these low frequency velocities from adding a periodic advective contribution to the computed fluxes, the turbulent fluctuations of vertical velocity and concentration are extracted by subtracting running averages. For AEC the extraction on the entire data set occurs after the campaign, with a centered running average of 60 s after Berg et al. (2003). For REA, the control system applies a 30 s back-looking running average to each measured velocity. We checked that AEC fluxes did not change significantly when switching from a 60 s centered average to a 30 s back-looking average.

3.3.4. Data Synchronization for AEC

For AEC, the synchronization between oxygen and velocity time series compensates for the delay due to the distance between velocity and oxygen probes and the response time of the latter. The usual strategy is to maximize the flux computed from time series shifted one relatively to the other within a 2 s search window (Fan et al., 1990; McGinnis et al., 2008). We used a wider search window of 5 s, since imperfectly synchronized computers acquired the measurements.

3.3.5. Computation of REA Oxygen Fluxes

In the adapted prototype, concentration is measured continuously in the sampling tubes. After the campaigns, to count only once the concentration of each water sample, only the concentrations measured while the water was

flowing through the corresponding sampling tube are included in the upward or downward simulated average concentration. The attenuation of fluctuations between the sampling tube inlet and the oxygen probe is neglected. Shift tests within a 2 s window showed that it was useless to synchronize the oxygen and valve opening sequences to account for the time lag between the two computers, since it did not substantially change the REA flux.

The accuracy of the oxygen probes is 0.8 mmol m^{-3} at 95% of saturation. In usual conditions in rivers ($\sigma_w \approx 0.01 \text{ m s}^{-1}$), the REA flux uncertainty, $200 \text{ mmol m}^{-2} \text{ day}^{-1}$, is often larger than the flux (Equation 3). A compensation of any calibration offset between probes drastically improves the accuracy of the measurement of concentration differences: before every flux averaging period, the valves are opened for 90 s, sampling the same water on both lines and yielding the bias between the optodes, $\overline{C_{\text{comp}}}$. It is used to compute the REA flux during the following flux averaging period:

$$F = b\sigma_w \left(\overline{C_{\uparrow}} - \overline{C_{\downarrow}} - \overline{C_{\text{comp}}} \right) \quad (5)$$

where the coefficient b is computed with Equation 4 and the coefficient values for oxygen.

3.4. Selection of Valid REA Measurements

Fotiadi et al. (2005) proposed a large set of quality criteria for atmospheric REA. Here we only used three. The first criterion, about good quality velocity measurements, is that the correlation between the transmitted and received pulses of the ADV be sufficient, above 70%, during more than 80% of the flux averaging period (Guseva et al., 2021).

The second criterion, about developed turbulence, is that we observe a turbulent inertial subrange in the power spectrum of the aligned vertical velocity, namely the $-5/3$ slope in logarithmic scales between power and frequency.

The third criterion, about the prototype alignment with the vertical, is that the absolute value of the mean aligned vertical velocity ($|\overline{w}|$) remains below the threshold velocity w_0 . It should vanish after double angle rotation but a non-zero value can persist, especially when current direction changes between prototype alignment and flux averaging periods.

We also computed the friction velocity u to quantify the turbulence level during each flux averaging period (Weber, 1999):

$$u = \left(\overline{u'w'^2} + \overline{v'w'^2} \right)^{1/4} \quad (6)$$

We did not use it for selecting measurements, but for estimating a turbulence threshold from the selection results.

3.5. Measurement Sites and Campaigns

REA requires both fully developed turbulence and measurable concentration differences between accumulated samples, in other words large benthic fluxes. Laboratory flumes ensure developed turbulence but it is difficult to create a vertical oxygen flux over a large surface area (Donis et al., 2015). Estuaries are dynamic due to tides (Koopmans & Berg, 2015), but during prototype development we preferred nearby research observatories with permanent technical staff for support and an easy access to water. We also did more campaigns in lakes than in rivers. In a river, benthic fluxes are often low due to the constant washout of sediments; flow rate varies mainly during high waters, when measurements become difficult; suspended matter quickly damages the AEC oxygen fiber; and a mass balance between upstream and downstream concentrations can yield benthic fluxes. On the contrary, lakes have a rich biogeochemistry, with large benthic fluxes and quickly varying turbulence conditions, which make the measurements both more challenging and promising.

3.5.1. River With Stable and Intense Turbulence

We measured oxygen fluxes in July 2020 in the Petit Morin River (modulus $3 \text{ m}^3 \text{ s}^{-1}$) at Courcelles-sous-Jouarre (70 km east of Paris, 48.939°N , 3.149°E). The water depth was 0.8 m and the measurement point was located in the middle of the river for homogeneous turbulence and fluxes, and in a straight section to avoid secondary currents, that is, the helicoidal flow resulting from turbulent shear stress on the river banks (Wetzel, 2001). The river bed was rocky and partly covered by macrophytes. The average current velocity at measurement height (0.2 m) was 0.09 m s^{-1} and the friction velocity $5 \times 10^{-3} \text{ m s}^{-1}$.

3.5.2. Shallow Lakes for a Compromise Between Turbulence and High Benthic Fluxes

The challenges for REA in shallow lakes are that turbulence should be sufficient and that the prototype should be aligned with the vertical in the absence of a stable current direction. High turbulence is rare, especially in summer when biological activity is intense but when thermal stratification decreases the energy transfer from the wind to near-bed currents. We investigated two shallow closed lakes during windy days with sufficient turbulence; the wind speed measured at the nearest Météo France weather stations ranged between 3 and 8 m s⁻¹ during the measurements.

The first site is a reservoir at the experimental platform PLANAQUA (CEREEP-Ecotron, Saint-Pierre-lès-Ne-mours, France, 48.283°N, 2.670°E). The artificial lake, located on a plateau and far from obstacles, is well exposed to wind. It is rectangular, 126 m long, 15 m wide, and 3 m deep. The sediment is sandy, homogenous and flat. The lake is oligotrophic; a liner isolates it from groundwater. We measured fluxes in the middle of the lake from a footbridge on 14 October 2020, during 15 min flux averaging periods. The average current speed at measurement height (0.15 m) was 0.04 m s⁻¹ and the friction velocity 0.01 m s⁻¹.

The second site, Lake Champs-sur-Marne (20 km East of Paris, 48.863°N, 2.597°E, Piccioni et al., 2021), is a former sand pit, fed by runoff and groundwater. Its surface area is 0.12 km² and its average depth 2.5 m. The lake is strongly eutrophic and undergoes recurrent algae blooms. Trees around the lake and on an island partially shelter it from wind. During the five days of measurements in July 2020, the lake was thermally stratified only during day time, with a vertical temperature difference of 1.5°C. Macrophytes and algae covered the sediment. We measured at two points, at shallow depth (1.2 m) from the lake shore (10 min flux averaging periods) and from a platform close to the middle of the lake (2.7 m depth, 8 min flux averaging periods). At shallow depths, the sediment was flat and the current velocity at the measurement height (0.15 m) varied from 4 × 10⁻³ to 7 × 10⁻³ m s⁻¹ and the friction velocity from 0.5 × 10⁻³ to 1.5 × 10⁻³ m s⁻¹. At larger depth, the bathymetry was irregular with a slope of 11°. The current speed at the measurement height (0.20 m) varied between 1.5 × 10⁻³ and 9 × 10⁻³ m s⁻¹ and the friction velocity from 0.8 × 10⁻³ to 6 × 10⁻³ m s⁻¹.

4. Results

Among the 33 REA measurements conducted at four different locations, 15 records fulfilled the criteria of good velocity data quality, developed turbulence and adequate alignment: 11 out of 24 in Lake Champs, one out of six in the artificial lake, and all three in the river. For 10 periods in Lake Champs, the power spectra of the vertical velocity did not present the -5/3 slope, indicating too low turbulence. During these periods, the maximum friction velocity was 0.5 × 10⁻³ m s⁻¹. In the same site, the prototype alignment was insufficient during three other periods ($|\overline{w}| > w_0$). In the experimental lake, ADV beam correlation was too low for five out of six periods due to technical difficulties.

The 15 flux values range from -150 to 50 mmol m⁻² day⁻¹ (Figure 2a). The flux direction is the same for all REA and AEC flux pairs. Positive fluxes indicate the production of oxygen by photosynthetic microalgae or macrophytes covering the sediment at the experimental lake and in Lake Champs (Figures 2c, 2d, and 2f), and negative fluxes correspond to the uptake of oxygen by the sediment in the river and in Lake Champs (Figures 2e and 2f).

Due to the low number of values, we combined them for statistical analysis. The 15 flux pairs measured by AEC and REA are in good agreement. The slope of the linear regression between both flux series is 0.96, with a coefficient of determination R^2 of 0.94 (Figure 2a). The mean absolute error of 12 mmol m⁻² day⁻¹ between the 15 paired fluxes represents 27% of their mean absolute AEC flux. The ANOVA test shows no significant difference in their median and variance ($p = 0.97$), nor the Student t -test any significant mean difference between flux pairs ($p = 0.84$).

The 11 REA oxygen fluxes measured in Lake Champs are partially correlated to the friction velocity ($R^2 = 0.54$, Figure 2b).

REA fluxes varied less in time in the river than in Lake Champs, with a coefficient of variation around 20% over 1 hr (Figure 2e) versus up to 100% over 3 hr (Figure 2f). Fluxes changed direction during the same day in Lake Champs (Figure 2f). AEC fluxes exhibited similar features (not shown).

5. Discussion

The fraction of successful REA measurements may appear low, but it helps understand the field of use of the technique. A turbulence threshold can be deduced from the flux averaging periods excluded due to the absence of an inertial subrange in the velocity power spectra, a friction velocity u of 0.5 × 10⁻³ m s⁻¹. It is comparable

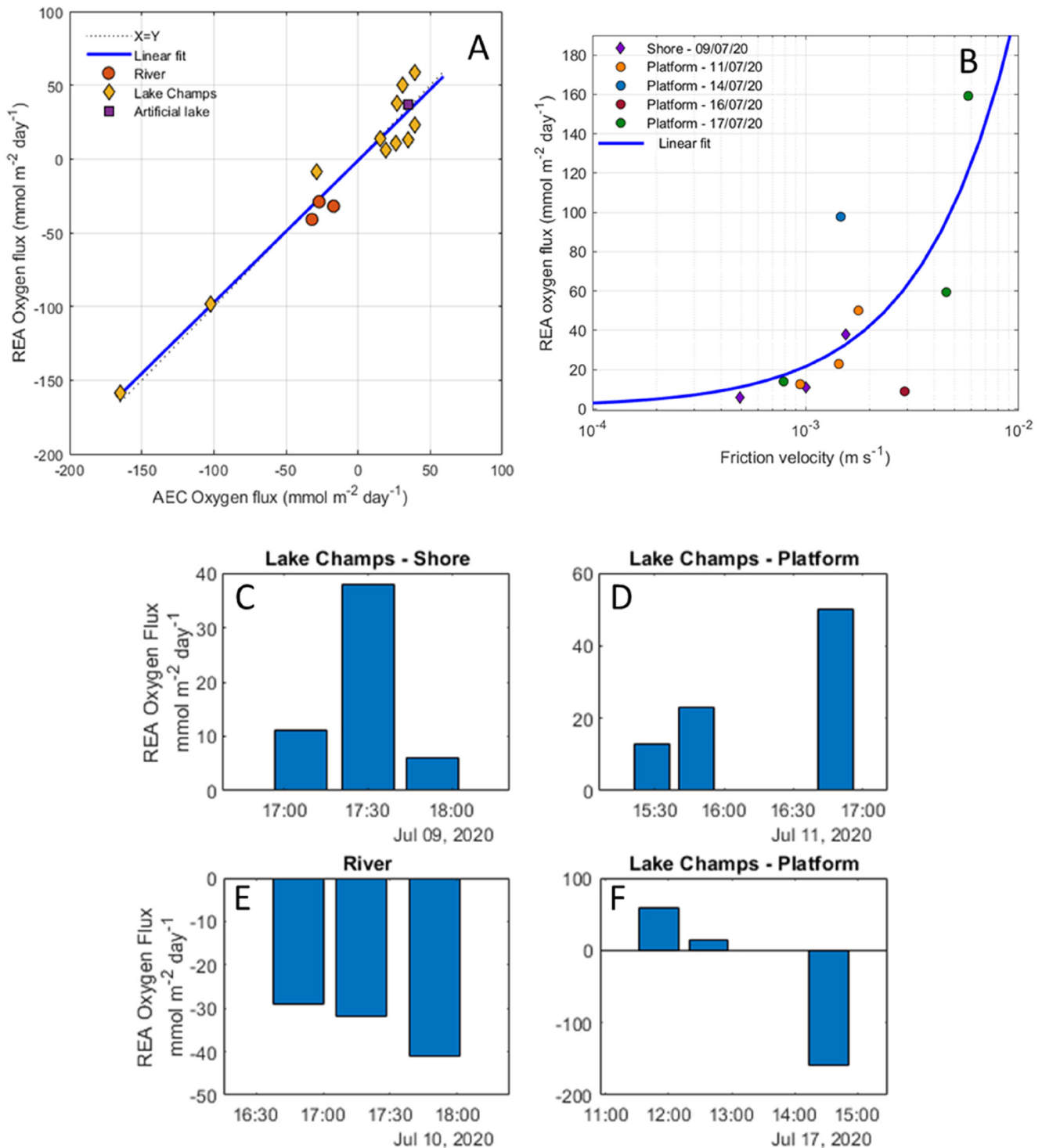


Figure 2. (a) Linear regression of the 15 REA and AEC oxygen flux pairs (equation $y = 0.962x - 1$; $R^2 = 0.94$). (b) Linear regression of the absolute values of the 11 REA fluxes measured in Lake Champs, close to the shore and from the platform close to the center, versus friction velocities (equation $y = 20.6 \times 10^3 x - 1$; $R^2 = 0.54$). (c)–(f) Time series of fluxes measured at the different sites over 1–4 hr in Lake Champs and in the river (the three single measurements are not shown).

to the lowest friction velocity at which Murniati et al. (2015) measured AEC oxygen fluxes in a riverine lake, $0.4 \times 10^{-3} \text{ m s}^{-1}$. Brand et al. (2008) measured AEC oxygen fluxes in low turbulence conditions in a deep lake with internal seiches, at minimum current speed and standard deviation of the vertical velocity of 5×10^{-3} and $0.3 \times 10^{-3} \text{ m s}^{-1}$, respectively; the minimum values of our measurements that respected the quality criteria are

comparable, 1.5×10^{-3} and 10^{-3} m s⁻¹. Absolute REA oxygen fluxes are correlated with the friction velocity (Figure 2b), as is often reported for absolute AEC fluxes (McGinnis et al., 2014).

Several factors can explain the slight discrepancy between REA and AEC fluxes, a relative mean absolute error of 27% and a regression line slope of 0.96 instead of 1. First, low current speeds cause large relative uncertainties in the mean velocity components and on the prototype alignment, on the running average used for extracting velocity fluctuations, on the standard deviation of the vertical velocity, on the velocity threshold for sampling, on the coefficient *b*, and finally on the REA flux.

A second possible source for the discrepancy is Equation 4, used to compute the empirical coefficient *b*. Here measurements were too few to recompute the coefficient *b*, whereas the reanalysis of more AEC data sets in different environmental conditions could reduce the uncertainty on the coefficients of the equation and help verify flux similarity between elements like oxygen, heat and particles (Lemaire et al., 2017).

A third point is whether continuous measurement of oxygen concentration in the sampling tubes correctly represent the average concentration in the accumulation bags. We limited the attenuation of concentration fluctuations in the tubes by keeping the distance between their inlet and the probes small. The compensation addresses the drift of the oxygen probes: the compensation term remained stable during most measurement campaigns, except when we regularly displaced the prototype for adjustments as in Lake Champs, with a variation by up to 5 mmol m⁻³ within a day, above the probe accuracy of 0.8 mmol m⁻³ at 90% saturation.

A fourth contribution to the discrepancy is the different velocity processing for REA and AEC, especially with rotation angles computed during a separate prototype alignment period for REA contrary to the very sampling period for AEC. We obtained comparable angles for both methods in the river, but very different in the lakes for most flux averaging periods, probably due to the absence of a main current direction. In order to investigate the impact of these different rotation angles, we compared AEC fluxes computed both with velocities processed for REA and for AEC: they are highly correlated (regression line of slope 1.01, $R^2 = 0.92$, normalized mean absolute error of 31%, data not shown), showing that REA velocity processing does not affect the flux significantly.

The prototype could be improved: in order to increase the frequency of REA flux measurements, an automatic multiple sampler could reduce the sample recovery time at the end of each flux averaging period, and the control system could update the alignment angles from the measurements of the previous flux averaging period or continuously (von der Heyden et al., 2021). This would probably be easier than to align the equipment at the beginning of each campaign with the planar fit method, which requires several hours of velocity measurements before sampling (Wilczak et al., 2001).

Among potential fields of application, REA should contribute to cover the knowledge gap about the dynamics of different benthic fluxes at the scale of a few hours in shallow lakes, beyond the dynamic of oxygen fluxes. We observed an intense dynamics of oxygen fluxes, with a change in flux direction during a day in Lake Champs (Figure 2f), probably due to the successive prevalence of primary production and sediment consumption.

Another field of application are rivers, where a defined main current direction and intense turbulence provide ideal hydrodynamic conditions. Oxygen flux values remained relatively constant over the short campaign of 1 hr (Figure 2e). Benthic fluxes could change over a day-night cycle, as observed by Berg et al. (2013), or when the flows vary, as in estuaries (Koopmans & Berg, 2015). The width of the river should be larger than the width of the measurement footprint, 6.5 times the measurement height (*h*) (Berg et al., 2007). REA could prove particularly useful over sediments unsuitable for other techniques, like gravel beds.

After an adaptation of the prototype for larger deployment depths, REA could also follow benthic flux dynamics in the coastal zone, where the flow direction can be expected to be stable for most of the circa 6 hr durations of flood and ebb cycles.

6. Conclusions

The aim of this work was to present an aquatic prototype of relaxed eddy accumulation and, among the different challenges for the adaptation of the technique at the sediment-water interface, to evaluate the reliability of this conditional sampling method. We configured the aquatic prototype for oxygen to avoid that gas exchange affect the measurements. The agreement with oxygen fluxes measured with aquatic eddy covariance, an established technique, proves the reliability of the sampling system in rivers and shallow lakes. Since the sample conservation

and recovery systems are widely used for water sampling in the environment, the prototype is ready to measure benthic fluxes of nutrients and other solutes with accurate analytical methods. It would require adaptations for deeper depths or for dissolved greenhouse gases or microplastics. This technical evaluation paves the way to explore the spatial-temporal influence of turbulence on benthic fluxes and the contribution of the sediment to local biogeochemical cycles in aquatic ecosystems.

Data Availability Statement

The AEC and REA data and flux estimates are available on a repository at <https://doi.org/10.17632/ntvxg7w28v.1> (Calabro et al., 2023).

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