Accounting for surface waves improves gas flux estimation at high wind speed in a large lake

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Abstract. The gas transfer velocity ($k$) is a major source of uncertainty when assessing the magnitude of lake gas exchange with the atmosphere. For the diversity of existing empirical and process-based $k$ models, the transfer velocity increases with the level of turbulence near the air-water interface. However, predictions for $k$ can vary by a factor of 2 among different models. Near-surface turbulence results from the action of wind shear, surface waves and buoyancy-driven convection. Wind shear has long been identified as a key driver, while recent lake studies have shifted the focus towards the role of convection, particularly in small lakes. In large lakes, wind fetch can however be long enough to generate surface waves and contribute to enhance gas transfer, as widely recognised in oceanographic studies. Here, field values for gas transfer velocity were computed in a large hardwater lake, Lake Geneva, from CO\textsubscript{2} fluxes measured with an automated (forced diffusion) flux chamber and CO\textsubscript{2} partial pressure measured with high frequency sensors. $k$ estimates were compared to a set of reference limnological and oceanic $k$ models. Our analysis reveals that accounting for surface waves generated during windy events significantly improves the accuracy of $k$ estimates in this large lake. The improved $k$ model is then used to compute $k$ over a one-year time-period. Results show that episodic extreme events with surface waves (6 \% occurrence, significant wave height $> 0.4$ m) can generate more than 20 \% of annual cumulative $k$ and more than 25 \% of annual net CO\textsubscript{2} fluxes in Lake Geneva. We conclude that for lakes whose fetch can exceed 15 km, $k$-models need to integrate the effect of surface waves.

1 Introduction

Lakes are universally regarded as significant sources of CO\textsubscript{2} to the atmosphere, however, the accurate quantification of the magnitude of such emissions remains to date challenging (Cole et al., 2007; Tranvik et al., 2009; Raymond et al., 2013). CO\textsubscript{2} fluxes can be directly measured with floating chamber or eddy covariance systems (Vachon et al., 2010; Vesala et al., 2006). However, both approaches have their own constraints. The former suffers from limited time and space integration (from...
minutes to hours, and centimetres to metres; Klaus and Vachon, 2020), while the latter remains technically difficult and can be influenced by non-local processes (entrainment from the shore or advection; Vachon et al., 2010; Esters et al., 2021). Long-term direct flux measurements are thereby mostly restricted to small lakes (Huotari et al., 2011) and fluxes remain mostly estimated with models. CO₂ fluxes at the surface of lakes operate through a net diffusive transport, therefore obeying the Fick’s first law:

\[ F = k \alpha \Delta p \text{CO}_2 , \]  

(1)

where \( F \) (mol m⁻² s⁻¹ but often expressed as μmol cm⁻² h⁻¹) is the CO₂ gas flux, \( \alpha \) is the CO₂ solubility coefficient (μmol cm⁻³ μatm⁻¹), \( \Delta p \text{CO}_2 \) is the gradient of partial pressure of CO₂ (pCO₂) between the water and the atmosphere corrected for altitude (μatm); and \( k \) is the gas transfer velocity (cm h⁻¹).

Lake carbon emissions are therefore primarily driven by the gradient of partial pressure of CO₂ between the surface lake water and the atmosphere, but the gas-transfer velocity controls the rate of CO₂ exchange across the lake-atmosphere interface. Assessing the amount of lake CO₂ emissions to the atmosphere has been a major issue, starting with the Cole and Caraco (1998) seminal paper, with debates regarding both the representativeness of the measurements and the optimal conceptual model for air-water gas transfer (e.g., MacIntyre et al., 2001; Borges et al., 2004). As recent developments in sensor technologies allow continuous and accurate measurements of aqueous CO₂ concentrations, the gas transfer velocity remains, to date, the main source of uncertainties, which hinders attempts to achieve full carbon budgets (Dugan et al., 2016) or to quantify greenhouse gas emissions by lakes, at local, regional, or worldwide scales (Maberly et al., 2012; Raymond et al., 2013; Engel et al., 2018).

\( k \) is inherently tied to turbulent mixing within the surface boundary layer, which enhances the diffusive gas exchange by renewing the surface mass content (Zappa et al., 2007). At the lake-atmosphere interface, turbulent mixing is the product of wind shear (\( k_w \)), buoyancy flux (\( k_b \)), and wind-driven surface waves, whose effect can be split into wave action (\( k_a \)) and wave breaking (\( k_b \) or \( k_b' \)), the latter producing air bubble and water spray (Fig. 1; Wiester and Lorke, 2003, Soloviev et al., 2007). Regarding the prominent role of wind action on surface turbulence, first quantitative models have empirically scaled \( k \) to wind-speed (referredenced at a 10 m height; \( U_10 \)), as a proxy for the level of wind-driven turbulence (Fig. 1; Cole and Caraco, 1998; Crusius and Wanninkhof, 2003). The parameterizations of the \( k \)-wind relationships vary between authors (e.g., Klaus and Vachon 2020), as a likely consequence of the local characteristics of the lakes used in the calibration datasets (Table 1). Yet, all studies suggested a polynomial relationship between \( U_10 \) and \( k \) with an exponent larger than 1. Further development of empirical models integrated the lake surface area as a second parameter in the \( k \)-wind relationships, to account for the role of the fetch length for wind action (Vachon and Prairle, 2013). Generally, empirical wind-based models tend to underestimate fluxes, especially at low-wind speed (e.g., Schubert et al., 2012; Heiskanen et al., 2014; Mammarella et al., 2015) where turbulent mixing through buoyancy flux is expected to take over wind shear. Besides, these empirical models require a proper
calibration each time they are applied in a new system with different characteristics, i.e., a new set of lakes and/or meteorological conditions (Klaus and Vachon, 2020), hence limiting their universal applicability.

**Figure 1**

In parallel to empirical wind-based models, process-based models attempt to link \( k \) directly to near-surface turbulence. The surface renewal model (SRM) is one of the first, and still most widely used, theories (Danckwerts 1951; Lamont and Scott, 1970) with \( k \) depending on the product of the turbulent kinetic energy dissipation rate (\( \varepsilon \)) and the kinematic viscosity of water (\( \nu \)), both to a power of one quarter as follows:

\[
k = a_1 (\varepsilon \nu) \frac{1}{\text{Sc}^{1/2}},
\]

with \( a_1 \) a calibration constant parameter and \( \text{Sc} \), the Schmidt number. Recently Lorke and Peeters (2006) and Katul and Liu (2017) demonstrated that this relationship, to which different approaches converge, can be seen as a universal scaling. As opposed to the practical empirical models presented above, process-based models have the potential to predict \( k \) using the turbulent dissipation rate over a wide range of environmental conditions extending beyond those encountered in the calibration dataset (Zappa et al., 2007). As for lakes, SRM \( k \)-models have so far considered the friction velocity at the water side (\( u_{\text{wat}} \)) and the turbulence created by thermal convection using the buoyancy flux at the surface (\( B_g \)) (Fig. 1; Eugster et al., 2003; MacIntyre et al., 2010; Read et al., 2012; Tedford et al., 2014; Heiskanen et al., 2014). Noteworthily, the SRM approach leads to \( k \) being related to \( u_{\text{wat}} \) (or \( U_{10} \)) to the first order (Wanninkhof, 1992; Lorke and Peeters 2006; see also Material and Methods section), while empirical models described above predict a higher order polynomial relationship. This inconsistency is tentatively solved in oceanography by adding another source of gas exchange associated with wind-waves whitecaps. Early gas flux parameterization already accounted for wind and buoyancy-driven turbulence together with surface waves (Fig. 1; Woolf et al., 1997; Soloviev et al., 2007; Fairall et al., 2011). Yet, the buoyancy-driven contribution can often be neglected in oceanography, and recent efforts have been dedicated to a better parameterization of the bubble enhancement term (Fig. 1; Deike and Melville, 2018). In lakes, wind fetch can be long enough to generate surface waves (Wanninkhof, 1992; Frost and Upstill-Goddard, 2002; Borges et al., 2004; Guérin et al., 2007), thus implying that surface waves could be a significant driver of \( k \) and subsequent CO\(_2\) fluxes (Schneider et al., 2013; Vachon and Prairie, 2013). Insofar, the role of surface waves has been essentially accounted empirically in lake \( k \)-models, through the polynomial scaling to \( U_{10} \) in wind-based models, and most often neglected in studies using process-based parameterizations (mainly SRM) (e.g., Read et al., 2012). While this approximation may be appropriate for small-shielded lakes, it is likely to be insufficient in larger, long-fetched lakes.

Herein, we aim to identify the most adequate \( k \)-model for Lake Geneva, a large clear hardwater lake in the Swiss Alps, to assess \( k \) values over a full annual cycle. We compare the performances of different models of gas transfer velocity, in their original or slightly modified published formulations from the limnological and oceanic literatures. This set of models includes different levels of complexity, ranging from empirical models integrating wind speed and lake size, to process-based models including wind shear, convection, and surface waves. Continuous \( \Delta p \text{CO}_2 \) measurements by in situ automated sensors and CO\(_2\)
fluxes, obtained from a new generation of automated (forced diffusion) flux chamber, were collected during specific periods of intensive field survey covering a wide range of natural conditions. Empirical $k$ values computed from chamber data are then compared to outputs from the different $k$-models. Owing to the size of Lake Geneva, we anticipate that models accounting, implicitly or explicitly, for the four key exchange drivers (i.e., wind shear, convective mixing, wave action and bubble formation) will show the highest accuracy and precision in their estimation of $k$ and that a precise integration of surface wave effects in such a large system should enhance model predictions. Thereafter, the relative distribution of these components is computed over a full year and analysed in the scope of the temporal variability of the gas transfer velocity. Finally, we expect that extreme wind and associated wave events should contribute disproportionately to accumulated $k$ values over the year. In such a case, episodic weather events could generate large CO$_2$ fluxes over very short timescales that should be accounted for when computing annual CO$_2$ emission budgets.

2 Material and Methods

2.1 Study Site

Lake Geneva is a peri-alpine lake defining part of the Swiss-French border, at 372 m above sea level (46° 26’ N; 6° 33’ E). Its surface area (582 km$^2$) and its maximum depth (309 m) make it the largest freshwater body in Western Europe, with a volume of 89 km$^3$ (Fig. 2). Lake Geneva is monomictic. The two prevailing winds are nearly opposed and come from southwest and northeast respectively (Fig. 2). The lake water has been monthly or fortnightly surveyed from the late 1950’s (OLA-IS, AnaEE-France, INRAE of Thonon-les-Bains, CIPEL, Rimet et al., 2020). Surface CO$_2$ concentrations, as computed from the routine temperature, alkalinity, and pH measurements (Stumm and Morgan, 1981), show a typical seasonal cycle with high, supersaturated values during winter mixing and values below saturation in summer (Perga et al., 2016).

2.2 Field data at LéXPLORE

All field data were collected from the LéXPLORE platform, a 10 m by 10 m pontoon equipped with high-tech instrumentation and installed on Lake Geneva in 2019. LéXPLORE is moored at a 110 m depth, 570 m off the northern lake shore (Fig. 2).

On LéXPLORE, local weather conditions (air temperature, wind speed and direction, relative humidity, short wave radiation and atmospheric pressure) were continuously recorded (10 minutes intervals) by a Campbell Scientific Automatic Weather Station. Lake surface temperature was measured every minute at 50 cm-depth using a Minilog II-T (Vemco, resolution 0.01° C). Partial pressure of water surface CO$_2$ (pCO$_2$) was also measured at 50 cm-depth during specific surveys (see flux measurement) using a miniCO$_2$ sensor (Pro-Oceanus Systems Inc.) with an accuracy of ± 30 ppm. Values of pCO$_2$ in ppm were converted into μatm following the basic equation correcting for altitude (Russell and Denn, 1972). We therefore assume that the concentration and the temperature are homogeneous over the first 50 centimetres.
Fetch distance (m) from LéEXPLORE to the lake shores considering wind direction was computed using data from the Federal Office of Topography online portal (Swisstopo-geoportal: geo.admin.ch). The position of LéEXPLORE is particularly relevant for this study as the fetch ranges from ~0.5 km to ~30 km for the two prevailing winds. Significant wave height, $H_s$ (in m) was computed after Hasselmann et al. (1973) according to:

$$H_s = 1.6 \times 10^{-3} \cdot U_{10} \cdot (\text{Fetch} \div g)^{1/2}$$

where $g$ is the gravitational constant. This variable $H_s$ is defined as the average height of the highest one-third of the waves (crest to trough) corresponding to the thickness over which the wind can push laterally (Wüest and Lorke, 2003). This equation is equivalent to the formulation by Carter (1982) that is more widely used in the oceanic literature. Simon (1997) tested the model for significant wave heights in Lake Neuchâtel (a lake close to Lake Geneva) with a fetch distance of 9 km. These results showed that the significant wave height in this lake was consistent with this oceanic formulation. However, Simon (1997) highlighted that the Joint North Sea Wave Project (JONSWAP) wave breaking parametrization did not hold for winds greater than 5 m s$^{-1}$ producing faster wave breaking and with a higher probability in the case of not fully developed surface waves. Such lake waves are characterized by steeper slopes that favour their wave breaking and wave action (Wüest and Lorke, 2003).

The net CO$_2$ flux at the lake-atmosphere interface, $F$, was directly measured with an automated (forced diffusion) floating CO$_2$ flux chamber (eosFD, eosense: environmental gas monitoring; Risk et al., 2011; Fig. A1) originally developed for soil flux studies. The flux chamber had a detection limit close to 0.05 μmol cm$^{-2}$ h$^{-1}$ and measured $F$ every 15-minutes in summer and 30-minutes in winter for battery-saving purpose. The standard floating chambers require quiet surface conditions (e.g., Cole et al., 2010; Vachon et al., 2010; Bastviken et al., 2015), thus limiting studies from low to moderate wind speed conditions. One typical problem with floating chambers arises from the possible atmospheric leakage under rough surface (Fig. A2a). To work around this problem, Vachon et al. (2010) advise to create 10 cm long-edges entering the water (Fig. A2b) and this design also reduces artificial turbulence generated by the chamber’s walls at surface. A second typical issue with this method is potential flux enhancement by artificial (chamber-generated) turbulence. This was also studied in Vachon et al. (2010), who demonstrated that the overestimations by this effect can be as high as 1000 % at low wind but less than 50 % when the wind speed exceeds 4 m s$^{-1}$ in large lakes. At even higher wind speed, this overestimation should further decrease because the surface water turbulence becomes much greater than that produced by the floating chamber. Thereby, our flux chamber was specifically conceived to increase stability under calm and windy conditions and limiting artificial turbulence, but we do not exclude a bias at low and moderate wind (Fig. A1, Fig. A2c).

Regarding the operation of the cosFD, it has two independent cavities: one for the chamber and one for the atmosphere (Fig. A1). These are connected to the same CO$_2$ sensor by a pump which sends at regular intervals (about 20 s) either the chamber
gas or the air gas to the sensor and then completely flushes the chamber cavity according to the programmed measurement timestep (15-minutes or 30-minutes). The advantage of this new instrument is therefore to have a constant monitoring of the chamber’s variation, but also of the atmosphere. In addition, the use of the same CO₂ sensor for the two measurements limits the need for intercalibration between CO₂ sensors. We tested the performance of the floating chamber by comparing the standard deviation of the CO₂ concentrations of the atmosphere and in the chamber estimated from two separated cavities (Fig. A1; Risk et al., 2011). We did not observe any difference in the standard deviation between high and low wind conditions (Fig A3), suggesting that the measured fluxes remained reliable at high wind speed without leakage of the chamber.

We assessed the performances of our flux chamber during 5 specific periods over the annual cycle (i.e., 13th–14th June 2019, 27th–28th August 2019, 1st–5th October 2019, 18th–20th December 2019, and 20th–26th February 2020). To select the most robust dataset to compare with \( k \) estimates derived from models, we discarded flux data that were below the detection limit, as well as CO₂ gradients that ranged within the uncertainty of sensors (i.e., \( \pm 20 \) ppm for air and \( \pm 30 \) ppm for water leading to \( \pm 50 \) ppm) (Fig. A2). Accordingly, we were able to retain the most robust data points during the following deployment periods: 18th–20th December 2019 and 20th–26th February 2020. Finally, all these field data were standardized at a 1-hour timestep.

### 2.3 Computed \( k \) values from field data

\( k \) values (cm h⁻¹) from field observations (\( k_{obs} \)) were computed from the gas transfer velocity equation:

\[
k_{obs} = \frac{F}{(\alpha \cdot \Delta p CO_2)},
\]

where \( F \) is the measured CO₂ flux (\( \mu mol \) cm⁻² h⁻¹), \( \alpha \) is the gas solubility coefficient (\( \mu mol \) cm⁻³ μatm⁻¹), which depends on the measured water temperature (Wanninkhof, 1992). \( \Delta p CO_2 \) is the differential of pCO₂ measured at 0.5 m below the surface (pCO₂-water) and pCO₂: at saturation (pCO₂-sat) in ppm measured from the flux chamber corrected by altitude (μatm). Noteworthily, the chemical enhancement factor (Wanninhof and Knox, 1996) was not considered in this equation since the fluxes retained corresponded to conditions of moderate pH (i.e., < 8). \( k_{obs} \) was then standardized in \( k_{600} \) using the dimensionless Schmidt number (\( Sc \)) of CO₂ by the equation: \( k_{600-obs} = k_{obs} \cdot \left( \frac{600}{Sc} \right)^{-1/2} \) (600 for freshwater standardized at 20°C).

### 2.4 Models for air–water gas transfer velocity

After years of debate, a consensus begins to emerge on the relationship linking \( k \), intensity of turbulence and \( Sc \) (Eq. 2), even when starting from different physical assumptions (see Katul et al., 2018). In this study, we selected six parameterizations widely used in limnology and oceanography combining specific calibration characteristics (Table 1). We first show that they can all be expressed following Eq. (2) for wind shear and convection, despite their different formulations. Then, we develop the effects of surface waves from oceanic models and adapt the wave action for a large lake. The final lake model integrating wave effect is ultimately calibrated using our field data (Table 1).
2.4.1 Wind shear stress

We start with the case where near-surface dynamics are driven by a weak to moderate wind, in absence of heat exchange. In this case, the contribution of surface waves can be neglected and the wind stress ($\tau_w = \rho_{\text{air}} \cdot C_{i0} \cdot U_{10}^2$, where $\rho_{\text{air}}$ is air density and $C_{i0}$ is the drag coefficient at 10 m) is equal to the tangential shear stress ($\tau_w = \rho_{\text{wat}} \cdot U_{*\text{wat}}^2$, where $\rho_{\text{wat}}$ is water density). The relationship between $\epsilon$ and the sheared velocity on the water side, $U_{*\text{wat}}$, is then derived from a law-of-the-wall scaling for the velocity profile: $\epsilon = U_{*\text{wat}}/Kz_0$ with $K$ being the von Kármán constant ($= 0.41$) and $z_0$ the thickness of the diffusive boundary layer. This relationship leads to:

\[ k_{NB} = a_1 \cdot \left( \frac{\nu U_{*\text{wat}}}{\kappa z_0} \right)^{1/4} Sc^{-1/2}, \]  

(5)

The challenge is then to define $z_0$. Tedford et al. (2014) followed an ad hoc observational approach and chose $z_0 = 0.15$ m, as the shallower depth where $\epsilon$ was measured. In contrast, theoretical studies linked $z_0$ to the thickness of the diffusive or viscous sublayer (~0.1–1 cm). In line with theory, we scale this layer as $z_0 = \nu / U_{*\text{wat}}$ (Wüst and Lorke, 2003; Lorke and Peeters, 2006) with $\nu$ as a constant value. Taking $\nu = 114$ (Soloviev et al., 2007), the thickness of this layer typically ranges from 0.04 to 0.14 m under a wind regime of 10 to 1 m s$^{-1}$. We therefore modify Eq. (5) as to compute the interfacial (no-bubble, NB) exchange coefficient:

\[ k_{NB} = a_1 u_{*\text{wat}} \left( \frac{1}{\kappa} \cdot \frac{\nu}{z_0} \right)^{1/4} Sc^{-1/2}, \]  

(6a)

or

\[ k_{NB} = a_1 \left( \frac{\rho_{\text{air}}}{\rho_{\text{wat}}} \right) C_{i0} \left( \frac{U_{10}}{\kappa} \right)^{1/4} Sc^{-1/2}, \]  

(6b)

These equations show that the SRM formulation (Soloviev et al., 2007; Read et al., 2012; Table 1 and Fig. B1) is analogous to the COAREG flux algorithm (Fairall et al., 2011), and the formulation used in Deike and Melville (2018) with the sheared velocity on the atmosphere side, $u_{*\text{atm}}$ (Table 1: DM18). Indeed, when equating the expression by Deike and Melville (2018):

\[ k_{NB} = A_{NB} u_{*\text{atm}} \left( Sc / 600 \right)^{-1/2} = A_{NB} u_{*\text{wat}} \left( \frac{\rho_{\text{wat}}}{\rho_{\text{atm}}} \right)^{1/2} \left( Sc / 600 \right)^{-1/2}, \]  

(7)

with (6a), we find that the coefficient $a_1 = 0.29$ in Soloviev et al., (2007) and Read et al., (2012) is essentially equivalent to the coefficient $A_{NB} = a_1 \left( \kappa C \right)^{-1/4} \left( 1 / 600 \right)^{1/2} (\rho_{\text{wat}} / \rho_{\text{air}})^{-1/2} \approx 1.5 \times 10^{-4}$ in Deike and Melville (2018) (Fig. B1), which in turn was found equal to the coefficient of $A = 1.5$ in Fairall et al. (2011). These results agree with Lorke and Peeters (2006) who derived a unified relation for interfacial fluxes (air-water and water-sediment) through a linear relationship of $u_{*\text{wat}}$ to $k$, especially at the bottom interface where shear is the only relevant process. Furthermore, Equation 6 has a similar (i.e., quasi-linear) wind-$k$ relationship as the data-driven parameterization from VP13 but cannot explain the higher order polynomial relationship reported in CW03 and CC98.
2.4.2 Convection

A second source of dissipation at the surface is the convection ($\varepsilon_c$) resulting from surface cooling. In the SRM formulation, only the negative buoyancy flux is considered when this term directly enters into the turbulent kinetic equation as production term. The combination of wind shear and free convection near a boundary is described by the Monin-Obukhov similarity theory (MOST) with a general form derived from a turbulent kinetic energy balance (Lombardo and Gregg, 1989; Tedford et al. 2014):

$$\varepsilon(z) = \varepsilon_6(z) \left( C_u + C_c \frac{\varepsilon}{\varepsilon_{LMO}} \right),$$

where $L_{MO}$ is the Monin Obukov length scale defined as $L_{MO} = u'_w B_0/\kappa B_0$, including $\varepsilon(z) = c_u \cdot \varepsilon_u + c_c \cdot \varepsilon_c$ in Eq. (2) that can be rearranged as:

$$k_{NB} = a_4 \left( \varepsilon_u (c_u + c_c \cdot B_0/\varepsilon_{atm})^{1/4} \right) S_c^{-1/2},$$

where $a_4$ ranges in the literature from 0.2 to 1.2 (Soloviev et al., 2007; MacIntyre et al., 2010; Tedford et al., 2014; Heiskanen et al. 2014; Winslow et al., 2016), $c_u$ from 0.84 to 1 (Winslow et al., 2016) and $c_c$ from 0.37 to 2.5 (Wyngaard and Coté, 1971; Tedford et al., 2014). Hereafter, we use the following set of values: $c_u = 1$ and $c_c = 1$. Fairall et al. (2011) used an essentially equivalent approach but formulated in terms of a Richardson number to describe the partitioning between dissipation from convection and wind shear, expressing the wind shear in terms of the air-side friction velocity:

$$R_f = B_0 u'^2 / u_{atm},$$

which can be integrated into (9) as:

$$k_{NB} = a_4 \left( \frac{\rho_{atm}}{\rho_{water}} \right)^{1/2} u_{atm} \left( \frac{c_u}{c_c} \right) \left( 1 + \frac{R_f}{R_{f,c}} \right)^{1/4} S_c^{-1/2},$$

with $R_{f,c} = c_u \rho_{atm} / c_c \rho_{water}$. The details of this demonstration can be seen in Soloviev and Schlüssel (1994).

2.4.3 Wave action

The effect of surface waves is commonly implemented in oceanography but barely considered in limnology. All process-based models rely on the same parameterization of energy dissipation by wind shear and convection. They however differ in how they parameterize energy dissipation by wave action and wave breaking.

The contribution of the wave action ($\varepsilon_w$) is, accordingly, added as a third source of turbulence (Fig. 1). In the presence of surface waves, the balance between $\tau_f$ and $\tau_c$ does not hold anymore. Therefore, Soloviev and Schlüssel (1994) added a corrective factor, $\varphi$, using the Keulegan number ($Ke = u'_w / f(g \nu)$), in order to decrease the component $\tau_f = \tau_u \cdot \varphi$ where $\varphi = 1/(1 + Ke / Ke_c)$, with the critical Keulegan number ($Ke_c$) defined in Soloviev and Lukas (2006). As a result, the equation for shear-driven dissipation $\varepsilon_u(z)$ is:

$$\varepsilon_u(z) = \varepsilon_6(z) \left( C_u + C_c \frac{\varepsilon}{\varepsilon_{LMO}} \right).$$
\[ \varepsilon_{u}(\varepsilon) = \frac{\varepsilon_{\text{sat}}}{k_{\text{cu}}} \cdot \theta^2 , \]  

Following this step, the turbulent kinetic energy dissipation rate from wave action \( \varepsilon_{w} \) is added and defined with the Keulegan number by Soloviev et al. (2007) as:

\[ \varepsilon_{w} = \alpha \left( \frac{\nu}{k_{\text{cu}}} \right)^{1/2} \frac{\delta_{\text{R}}^{3/2} \rho_{\text{atm}}}{(1 + \varepsilon_{\text{R}}/\varepsilon_{\text{sat}})^{3/2}} \frac{\varepsilon_{\text{sat}}}{\mu_{\text{atm}}} \]  

where \( C_{T} = \left( \varepsilon_{\text{R}}/H_{s} \right) \) \( \psi \) is the surface roughness scale from the water side and the \( C_{T} \) value is set as a constant at 0.6 (More details in Soloviev et al., 2007). This definition does not hold for closed basins because, in the case of incompletely developed waves, the dissipation of energy from wind shear transmitted to the waves is not fully redistributed in the water body (Simon, 1997). Hence, for the application in Lake Geneva, we followed Terray et al. (1996) who defined a varying \( C_{M} \):

\[ C_{M} = 1.38 \cdot 10^{-4} U_{10}^{2.66} \left( \frac{\nu_{10}}{C_{P}} \right)^{2.66} \],

where \( C_{P} \) is the peak speed of the wave spectrum defined in Deike and Melville (2018) according to Toba (1972, 1978). This leads to \( C_{T} \ll 1 \). This allows to increase the effect of \( \varepsilon_{w} \) (inversely proportional to \( C_{T} \) in Eq. 12) on \( k \). Here, we used this formulation to adapt the S07 ocean model for a large lake (closed basin). Henceforth we refer to the adapted uncalibrated and calibrated models as \( SD21 \) and \( SD21-fit \) respectively (Table 1). Finally, these three terms of \( \varepsilon_{w} \) \( (\varepsilon_{w}, \varepsilon_{w}, \varepsilon_{w}) \) can be added before computing the SRM (Eq. 2) for determining \( k_{\text{no bubble}} \).

### 2.4.4 Bubble enhancement

Additional deviations from the linear relationship to \( U_{10} \) are explained by the gas transfer resulting from bubbles and sprays during wave breaking. This mechanism is accounted for by adding a \( k \)-bubble \((k_{b})\) term to the already mentioned \( k_{\text{no}} \). Soloviev et al. (2007) used the empirical \( k \)-bubble parameterization from Woolf et al. (1997):

\[ k_{b} = \frac{W}{c_{1} \left( 1 + \frac{1}{1 + 0.5 \cdot W_{0}} \right)^{1/2}} , \]

where \( W \) is the fractional whitecap coverage only expressed as a function of wind \( (3.84 \cdot 10^{-6} \cdot U_{10}^{2.44}) \) and \( O_{s} \) is Ostwald gas solubility. This formulation does not take wave height into account (Fig. B1). Nevertheless, a recent study (Deike and Melville, 2018) performed a new numerical process-based parameterization for gas transfer velocity from bubble enhancement considering \( H_{t} \) through the following equation:

\[ k_{b} = \frac{A_{b} \cdot \beta^{3/2} \cdot \theta^{3/2} \cdot \rho_{\text{atm}}^{1/2}}{\Delta_{\text{atm}} \left( \frac{H_{t}}{6.5} \right)^{1/2}} , \]

where \( A_{b} \) is an empirical factor with dimension \( (= 10^{-3} \text{~m}^{-2} \text{~s}^{-1}) \) and \( \beta \) defines by the ideal gas constant \((R)\), the surface water temperature \((T_{w})\) and, CO₂ solubility coefficient in freshwater \((\alpha)\) (Reichl and Deike, 2020). Then, the gas transfer velocity is
expressed as a sum of no-bubble $k_{\text{NB}}$ and bubble $k_b$ components (Table 1: S07, DM18, SD21, and SD21-fit) following Keeling (1993) and Woolf et al. (1997, 2005). Our adapted model for lake includes a refined parametrization of the wave action term $e_w$ from S07 along with the bubble term from DM18. For these reasons, the model will be called SD21 in the rest of the manuscript. In addition, for the appellation SD21-fit, the $a_1$ parameter of Eq. (2) and the $A_B$ parameter of Eq. (15) were fitted to the $k_{\text{obs}}$ observations ($a_1 = 0.33$ and $A_B = 3 \times 10^{-5}$ m² s²).

With this review of existing and adapting parameterizations, we show that (i) there is a discrepancy between SRM-based model with shear stress as the only energy source and empirical parameterizations with polynomial (order > 1) wind-based relationship. Such a discrepancy is tentatively resolved by adding the effect of convection and surface waves. (ii) We further highlight that most fitting parameters from the different SRM-based models are in good agreement. (iii) We finally recall that it is possible to provide a unifying parameterization of $k$ with SRM model including wind shear, wind-induced waves, and convection with only a few input parameters such as $U_{10}, B_0$, and Fetch.

3 Results

3.1 Observed and predicted $k$

After quality check, our dataset contains 94 discrete CO₂ flux observations. We first assess the representativeness of our sampling by comparing the survey-specific and annual distributions of the three main inputs for $k$-models: $U_{10}$ (all models), $B_0$ during convective periods (T14, S07, SD21 & SD21-fit) and $H_s$ (S07, DM18, SD21 & SD21-fit) (Fig. 3: Temporal evolution of these three terms in Fig. C1). From 13th June 2019 to 12th June 2020, the average wind speed over Lake Geneva is 2.9 m s⁻¹ with a mode at 2.5 m s⁻¹; very low wind speeds (< 1 m s⁻¹) are encountered 12 % of the year, while high- (> 5 m s⁻¹) to very high (> 10 m s⁻¹) wind events represent 15 % and 2 % of the year, respectively. The sampling surveys covered the full annual range of $U_{10}$. Average and modal values of $B_0$ over the year are close to 0.25 $10^{-7}$ m² s⁻³. However, the sampling covered only the lowest 50 % of the annual distribution and under-samples conditions of potentially strong convection. Considering that the dissipation by buoyancy flux, as parameterized in the process-based models, is already well known in the literature and that it is not the central point of our study, we posit that the under-sampling of $B_0$ is therefore not expected to significantly affect our analysis. The predicted modal $H_s$ value is 0.15 m over the year. Events of high $H_s$ (> 0.4 m) represent 6 % of the year, with a maximum $H_s$ of 1.1 m. As for $U_{10}$, the surveys covered the full range of annual $H_s$.

Figure 3

Obliquely based $k_{\text{obs}}$ are shown with their error bars corresponding to the uncertainties of the pCO₂ in air and in water (± 50 ppm) in Fig. 4a. We notice that all the measurements with a wave height > 0.4 m were observed for wind speeds > 5 m s⁻¹ and the corresponding $k_{\text{obs}}$ are located above the linear function (i.e., from a linear regression against wind shear velocity; Fig. B1) scaling $k_{\text{obs}}$ to $u_*$ (i.e., first order relationship). We then compare the $k_{\text{obs}}$ observed during the specific surveys to the
values computed with all $k_{60}$ models throughout the annual cycle, in relation to $U_{10}$ (Fig. 4b-c). Table 2 provides the root-mean square errors (RMSE) for all model estimates compared to $k_{obs}$ during the flux surveys, (i) for the full dataset (All Wind), and split (ii) for low wind ($< 5 \text{ m s}^{-1}$, LW) and (iii) strong wind conditions ($\geq 5 \text{ m s}^{-1}$, SW). The three empirical wind-based models only depend on wind (Fig. 4b). Both $CC98$ and $CW03$ were originally calibrated for small lakes, using a mass balance calibration method (Table 1). However, they lead to divergent gas transfer velocities, particularly above $5 \text{ m s}^{-1}$, illustrated by a RMSE for SW as high as 22.8 cm h$^{-1}$ for $CC98$ while $CW03$ performs better (RMSE SW = 12.8 cm h$^{-1}$). Furthermore, both models underestimate $k_{60}$ at low wind (Fig. 4a), with a higher deviation for $CW03$ (Table 2). The $k$ values predicted by $VP13$ are closer to those of the process-based models that explicitly integrate wave actions ($S07$ and $DM18$) (Fig. 4bc), demonstrating that lake size integration in the empirical model captures at least part of the wave action on $k$. Performances of $VP13$ at strong winds (RMSE SW = 12.7 cm h$^{-1}$) were better than those of the ocean-derived models integrating surface waves (RMSE SW = 13–15.9 cm h$^{-1}$). However, $VP13$ shows a positive offset during calm periods, along with the highest RMSE of the set of models at low wind speed (Fig. 4b; Table 2).

Figure 4
The process-based models (Fig. 4c) provide different $k_{60}$ values for a given wind speed, owing to the integration of additional environmental components (i.e., the varying drag coefficient, the convective mixing in $R12$ and $T14$ as well as the effect of waves in $S07$, $DM18$, $SD21$ and $SD21$-fit). All process-based models are similar at low winds as they share a common physical basis for parameterization of wind shear and convection. Therefore, they lead to similar RMSE (2.9–3.5 cm h$^{-1}$) under such conditions, where surface waves are negligible (Table 2). Divergences occur at higher wind speeds. $T14$, initially developed for small lakes with limited wind exposure, performed the worst (RMSE: 19.8 cm h$^{-1}$). This increased $k$-underestimation at high winds can be attributed to (i) dissipation by wave action and bubble formation not considered (in $R12$ and $T14$), and (ii) to the use of a constant $z_{f}(\theta_{1}) = 0.15 \text{ m}$ in the $T14$ model (Eq. 5). This approximation of the diffusive layer is consistent with low wind speed but is almost one order of magnitude too large under strong wind speed. Other process-based models, designed for greater wind range ($> 10 \text{ m s}^{-1}$), integrate surface waves and, as a result, lead to better estimates than $R12$ and $T14$ (RMSE: 10.4–15.9 cm h$^{-1}$). However, the ocean wave model of $DM18$ shows lower performances at strong winds than $CW03$ and $VP13$ (Fig. 4bc). Finally, the specific fit parameterization of the $SD21$-fit model improves the performance at high wind speeds by ~30 % (RMSE = 10.5 cm h$^{-1}$), outperforming all the other methods.

Table 2

3.2 Surface wave integration
We herein scrutinize how those varying parameterizations ultimately alter the shape of relationship between $k_{60}$ and $U_{10}$. In $R12$ (Fig. 5a), wind is only included through wind shear, resulting in a linear relationship between $k_{60}$ and $U_{10}$, as already anticipated. Adding the wave action (no bubble) through the $S07$ parameterization (Fig. 5b) does not lead to any significant departure from the minimal $R12$ model. Adapting wave action by decreasing $C_{T}$ (for the no-bubble term) leads to a departure from the wind shear linear relationship for $H_{s} > 0.4 \text{ m}$ (Fig. 5c).
Then, adding the $k$ bubble term related to wave breaking of $DM18$ further increases this deviation from the linear $k_{oo} - U_{10}$ relationship but also scatters $k_{oo}$ estimates for a given $U_{10}$ (Fig. 5d). Finally, the fitting with observationally based $k_{oo}$ improves the estimation for strong wind (Fig. 5e; Table 2). Given the range of wind fetch from ~0.5 km to ~30 km, the contribution of waves varies for a given wind speed depending on the fetch, as evidenced by the scattering of the parameterized $k_{oo}$ for a given $U_{10}$ (Fig. 5f). A significant modification of $k_{oo}$ by wave action and wave breaking occurs for a fetch length > 15 km and $U_{10} > 5$ m s$^{-1}$ (Fig. 5f), in the case of Lake Geneva, generating wave of $H_{s} > 0.4$ m (Fig. 5e).

**Figure 5**

As compared to $k_{oo}$ estimated by $R12$ (Fig. 5a), the $SD21$ and $SD21$-fit models provide $k$ estimates that are 20–50 % higher for $U_{10} = 10$ m s$^{-1}$, respectively, and 40–70 % higher for $U_{10} = 15$ m s$^{-1}$. Therefore, adapting the surface waves, through the change of the wave action for incompletely developed waves and the fitting to observed data encountered in local lake conditions, leads to better performances of $SD21$ models. $SD21$-fit reached the lowest RMSE at all wind speeds and is thereafter used as a reference for the modelling of the annual gas transfer velocities.

**3.3 Annual cumulative gas transfer velocity and the effect of extreme conditions**

We show above that models accounting for surface waves better represent the non-linear increase in $k_{oo}$ at high winds. Because high-wind events remain rare, we test whether a better representation of $k_{oo}$ during rare, high-wind events affects the local estimates of $k_{oo}$ over a full year. To this end, cumulative sums of hourly $k_{oo}$ were computed over a full annual cycle (13th June 2019 – 12th June 2020) for all $k$-models (Fig. 6). The annual dynamics such as the annually averaged $k_{oo}$ were compared using $SD21$-fit as a new reference model.

**Figure 6**

Cumulated $k_{oo}$ computed for $SD21$-fit shows some episodic steep increases between December and March, due to the winterly prevalence of high wind events (winter average wind speed, 3.25 m s$^{-1}$, was greater than the summer mean, 2.55 m s$^{-1}$, by 25 %) and greater significant wave height (winter average wave height, 0.15 m, greater than the summer value, 0.10 m, by 50 %) (Fig. C1). The average hourly $k_{oo}$ by the $SD21$-fit model is $7.3 \pm 7.4$ cm h$^{-1}$ (mean ± se, Fig. 6a). Periods of high-winds, although accounting for 15 % of data points, contribute 44 % of annually cumulated-$k_{oo}$ in the $SD21$ and $SD21$-fit models (Fig. 6b), while the periods of high waves ($H_{s} \geq 0.4$ m) accounting for only 6 % of data points, contribute to more than 20 % of annually cumulated-$k_{oo}$. The wind-based models are those for which cumulative $k_{oo}$ diverges the most from the $SD21$-fit reference model, with the lowest annual averaged $k_{oo}$ for $CC98$ and $CW03$ (3.9 ± 2.7 and 4.8 ± 9.3 respectively) and the highest for $VP13$ (9.9 ± 6.1). These divergences arise from the low performances of these models at low wind regimes (Fig. 4a, 6b; Table 2), which represent 85 % of annual data-points. All the other process-based models have relative dynamics of cumulatve $k_{oo}$ similar to that of the $SD21$-fit model and end up with annually averaged $k_{oo}$ that are 15 % lower than for the $SD21$-fit. The representation of $k_{oo}$ at low wind speeds is similar for all process-based models, and the divergence arises from the representation of the rarer high wind speed episodes, which contribute to 43–46 % of annual cumulative $k_{oo}$ (Fig. 6b).
4 Discussion

The history of k-models, simulating the gas transfer velocity for surface waters, dates back from the early 1990’s. k-models have been developed and tested in small lakes sheltered from winds (e.g., Crusius and Wanninkhof, 2003; Tedford et al., 2014), large lakes under low to moderate wind speed (Vachon and Prairie, 2013), and oceans (e.g., Soloviev et al., 2007; Fairall et al., 2011; Esters et al., 2017; Deike and Melville, 2018). While the effects of surface waves on k can be neglected in small lakes, we question herein whether this assumption holds for large lakes such as Lake Geneva, in which surface waves are frequently observed (Fig. 2; Fig. C1). We evaluated the performance of different experimental-based and process-based models to estimate $k_{600}$ in the large Lake Geneva. We show that integrating the effect of wave formation at high wind speeds and long fetch better represents the sharp increase of the $k_{600}$ values during such episodic windy events.

4.1 Choice of k-models

Wind-based models have been long known for misestimating $k_{600}$ at low wind speeds (Eugster et al., 2003; MacIntyre et al., 2010; Erkkilä et al., 2018). Consistently, wind-based models showed the lowest performances for Lake Geneva, especially at low wind speeds (CW03 and VP13), which resulted in large discrepancies in annually averaged and cumulative $k_{600}$ over the full year. They are however easy to compute, require few inputs (only $U_{10}$) and, remain by far the most used to estimate lakes CO$_2$ emissions worldwide (e.g., Raymond et al. 2013). Another possibility is to broadly adopt process-based models. The presented process-based models require input data that are today more easily accessible: wind speed, heat flux and wind fetch (i.e., distances from the shore) routinely acquired at high-frequency in many lakes. The development of R packages such as Lake Metabolizer (Winslow et al., 2016) in which the calculations of process-based models are implemented, also alleviates their computational difficulty. Both increased data availability and computational tools should foster the use of process-based k-models, which hold great potential to obtain more accurate global $k_{600}$ estimates.

The analysis of the models adapted from the existing literature to account for the effect of lake surface waves, SD21 and SD21-fit (Fig. 5d-e-f), showed that the wave contribution to k becomes significant for $H_s > 0.4$ m, corresponding, for Lake Geneva, to winds blowing at $5 \text{ m s}^{-1}$ from the southwest where the fetch length is maximal (> 15 km) with respect to the measurement site. A significant contribution to the gas transfer velocity by surface waves is expected in lakes where $H_s > 0.4$ m is not infrequent. Wave heights beyond this threshold value of $H_s$ are frequently encountered in lakes of similar or greater sizes than Lake Geneva (6 % of annual time at the LEXPLORE platform). In the Great Lakes of North America, Hubertz et al. (1991) showed that the mean wave height of all these lakes were > 0.4 m in summer and close to 1 m in winter with a maximum of up to 5 m. $H_s > 0.4$ m can also form over elongated lakes of smaller size, such as smaller Swiss Lakes (e.g., Lake Neuchâtel, Lake Bienne) (Amini et al., 2017). Since SD21-fit is a process-based model integrating the four main processes in a mathematically coherent way, we would expect that it can be applied to such lakes experiencing $H_s > 0.4$ m and improves the accuracy of $k$ estimates. Because waves can physically damage in-shore and off-shore infra-structures, many large lakes benefit...
from wave forecasts. $H_s$-data from those forecasting systems (e.g., National Data Buoy Centre – NOAA, Wave Atlas from SwissLakes.net; Amini et al., 2017) could allow testing whether the $SD21$-fit models can be applied to those lakes and whether $k_{NB}$ and $k_B$ through $a_1$ and $a_2$ need to be recalibrated or fitted to the local context if flux measurement data are available, as for this study. Energy dissipation during high-wave events increases the gas-transfer velocity well beyond the linear relationship derived for wind shear alone. We therefore expect that computed gas fluxes at the air-water interface should be significantly improved by the integration of surface waves into the $k$-models.

### 4.2 Implication of four components on the annual $k$ estimation and the annual CO$_2$ fluxes

#### 4.2.1 Seasonal and hourly distribution of $k$$_{CO2}$

Converting $k_{600}$ to $k$$_{CO2}$ using the Schmidt number (Wanninkhof, 1992) highlights the importance of water temperature in gas exchange dynamics. Indeed, the seasonal distribution of the cumulative $k_{600}$ is ~20 % and ~30 % for the warm (spring and summer) and cold (autumn and winter) seasons, respectively. Once the temperature-effect accounted for, this distribution increases to 26.1 % for the summer and decreases to 24.9 % for the winter but remains unchanged for spring and autumn. While $R12$ only use wind shear and convective terms, the selected process-based model ($SD21$-fit) allows a decomposition into the four main drivers of the gas transfer velocity, hence paving the way to a better understanding of the implication of those processes throughout an annual cycle.

**Figure 7**

Wind shear remains the dominant component of the gas exchange velocity over the different seasons (Fig. 7a). The annual contribution of surface waves (wave action and bubble formation) is limited to 9 to 10 % of the cumulative $k$ in Autumn and Winter. The contribution of buoyancy flux at surface to $k$ is even smaller for both models ($R12$ and $SD21$-fit) at this seasonal scale. Yet, both the buoyancy flux and the surface waves can significantly increase $k$ during episodic events, during which they can contribute disproportionately to $k$ at hourly (up to 80% for convection) and daily (up to 25 % for surface waves) time scales (Fig. 7b). Several studies have emphasized the disproportionate contribution of episodic mixing events on annual flux, bringing CO$_2$ back to lake surfaces such as after ice break in dimictic lakes (Karlsson et al., 2013; Finlay et al., 2019) or during fall mixing on a eutrophic deep lake (Reed et al., 2018). Process-based $k$-models integrating both the buoyancy flux and the wind-induced waves offer the opportunity to mechanistically investigate how much those episodic events contribute to annual emissions through short-term modifications of the gas exchange velocity.

#### 4.2.2 Consequences on the choice of $k$-model on the seasonal to annual CO$_2$ flux estimation

We produced coarse estimates of monthly CO$_2$ fluxes with the objective to scale the effects of wave integration at seasonal and annual scales. Monthly fluxes were computed based on $k$-estimates at LeXPLOR from the different models at hourly timestep and the monthly average of water temperature and recorded pCO$_2$ at the lake surface (OLA-IS, AnaEE-France, INRAE of Thonon-les-Bains, CIPEL, Rimet et al., 2020; Perga et al., 2016) as well as a constant pCO$_2$ in the atmosphere (400...
μ atm). For months for which surface pH > 8.4, k-value were computed with and without considering the chemical enhancement (CE; Wanninkhof and Knox, 1996) (Table 3). The dependency of the chemical enhancement factor on k (Wanninkhof and Knox, 1996) might generate a further uncertainty in estimated CO₂ fluxes (a greater k coming up with a lower chemical enhancement factor).

As predicted by the Fick’s law, the highest outgassing fluxes occur in fall and winter, when water mixing brings CO₂ up to the lake surface, while low up-taking gas fluxes occur in spring and summer, when primary production depletes surface CO₂ below saturation. However, annual estimates of net CO₂ outgassing vary from 14.7 to 37.1 mmolC m⁻² d⁻¹ (Table 3) depending on the k-model used for computation. Consistently, differences between model estimates are relatively low in summer since both the ΔpCO₂ gradient (100-200 μ atm) and wave occurrence are limited. Adding CE during spring and summer months causes an increase in influx of the order from 5 to 128 % depending on the k-model used. For example, using of CC98 in summer leads to 93 % increase of the estimate influx (as compared to fluxes without CE), while the increase is only 39 % for the SD21-fit. This chemical enhancement factor would deserve more attention in future studies especially for high pH lakes and summer seasons. Yet, the variability introduced by the CE at the annual scale remains low (~10 %) as compared to that introduced by the choice of the k-model. Estimated fluxes are also strongly dependent on the chosen k-model in winter when both ΔpCO₂ (475 μ atm) and surface waves occurrence are higher (Table 3; Fig.7). Therefore, while high wave events represent only 6 % of the total surface waves occurrence (Hₛ > 0.4 m), an incomplete consideration and description of their contribution may lead to an annual flux underestimation of about 20–25 %.

The weak contribution of convection is at odds with observations in small lakes, but not unexpected, since large lakes are exposed to stronger winds, such that wind shear-driven εₚ often outpaces convectively driven, εₜ (Read et al, 2012). However, the limited impact of the buoyancy flux on k does not rule out its contribution to CO₂ exchange. Indeed, convective mixing plays a central role in the deepening of the mixed layer allowing the export of the CO₂ stored in the hypolimnion towards the surface during the cold period and thereby controlling the pCO₂ gradient (Zimmerman et al, 2020) and the observed wintertime outgassing. Altogether, both surface oversaturated CO₂ concentrations (as a result of convective mixing) and wind-induced waves are more relevant in fall and wintertime for the monomictic Lake Geneva, leading to most of the annual outgassing during this season (Table 3). As for many monomictic lakes, these seasons drive most of the annual CO₂ budget of Lake Geneva (Perga et al, 2016), while they usually correspond to those where direct measurements are the scarcest. An improved quantification of k-values through SRM-models including wind-induced waves should contribute to refining the overall estimation of large lakes contribution to regional CO₂ emissions.

Table 3

4.3 Wind and wave field on Lake Geneva and their impact on the spatial integration of kₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚₚportion de l'ajout de CE aux mois de printemps et d'été peut causer une augmentation de l'ordre de 5 à 128 % de l'estimation des influx, dépendant du modèle k utilisé. Par exemple, en été, l'augmentation s'élève à 93 % de l'estimation des influx (par rapport aux influx sans CE), tandis que l'augmentation est de 39 % pour le modèle SD21-fit. Ce facteur d'amélioration chimique mérite plus d'attention dans les futures études, notamment chez les lacs à pH élevé et les saisons d'été. Toutefois, la variabilité introduite par le CE à l'échelle annuelle est faible (~10 %) par rapport à celle introduite par le choix du modèle k. Les estimations des influx sont également fortement dépendantes du modèle k utilisé en hiver, lorsque les gradients ΔpCO₂ (475 μ atm) et les vagues océaniques sont plus élevés (Table 3; Fig.7). Par conséquent, bien que les événements de vagues représentent seulement 6 % de la fréquence totale de vagues (Hₛ > 0.4 m), une prise en compte incomplète et non décrite de leur contribution pourrait entraîner une sous-estimation annuelle de 20–25 %.

La faible contribution de la convection est en contradiction avec les observations dans les petits lacs, mais il s'agit d'une observation attendue, car les grands lacs sont exposés à des vents plus forts, tels que le vent shear-driven εₚ qui souvent prime sur le vent convectif driven, εₜ (Read et al, 2012). Toutefois, l'impact limité du flux de densité sur k ne retire pas son contribution à l'échange de CO₂. En effet, la mélangée convective joue un rôle central dans le profondissement de la couche homogène, permettant l'exportation du CO₂ stocké dans le limon hypolimnion vers la surface pendant la période froide et en cela contrôlant le gradient pCO₂ (Zimmerman et al, 2020) et l'expulsion de CO₂ pendant les périodes d'hiver. Ensemble, les concentrations de CO₂ sursaturées à la surface sont plus pertinentes en automne et hiver pour le lac monomictique Lake Geneva, ce qui conduit à la majorité des émissions annuelles de CO₂ pendant cette saison (Table 3). Comme pour de nombreux lacs monomictiques, ces saisons conduisent à la plupart du budget annuel de CO₂ du lac Lake Geneva (Perga et al, 2016), bien qu'elles correspondent généralement à celles où les mesures directes sont les plus rares. Une amélioration de la quantification des k-values via des modèles SRM-ayant en compte les vagues induites par le vent contribuerait à affiner l'estimation finale de la contribution des grands lacs à l'émission de CO₂ régionale.
Therefore, the question of the extrapolation of the model to the whole lake remains essential. Herein, we showcase two snapshot situations of high-wind to (i) illustrate how process-based models could enable spatially resolved estimates of $k$-values, and (ii) exemplify how much $k$ can vary as a result of the spatially variable wave and wind-fields during a single episode. Two events of high and similar wind speed ($11 \text{ m s}^{-1}$) but different directions (NE: 2020.03.30 08:00 and WS: 2020.02.10 06:00; Fig. 8a-b) were extracted from the $0.01^\circ$ hourly resolved numerical weather model of the Swiss Federal Office of Meteorology and Climatology (COSMO-1, MeteoSwiss). The two fetch distances from both prevailing wind directions (NE, SW) were then measured at each pixel of the grid ($n = 583$), from which the wave height field was mapped (Fig. 8c-d) considering Equation (3) and the two wind grids. The maps were qualitatively consistent with previous studies on wind waves for Lake Geneva using the spectral wave model (SWAN) for wave height (Amini et al. 2016). Spatially resolved $k$-values were computed from the fetch and wind grids, using the wind-based model CC98, the process-based model without lake wave implementation R12 and the SD21-fit model containing the lake wave parametrization.

Taking $H_l > 0.4 \text{ m}$ as a threshold for the significant effect of wave on $k$, the two prevailing winds show opposite waves response, with long fetch and higher wave heights affecting either the North or the South shores for respectively the WS and NE winds. In both conditions, more than 60% of the full lake surface area experience $H_l$ values $> 0.4 \text{ m}$, leading to $k$-values as computed by SD21-fit as high as $68 \text{ cm h}^{-1}$ (Fig. 8g). The Eastern part of lake experiences the lowest wind speeds and wave heights in both situations, as a consequence of the orographic effect of the Alps surrounding the Grand Lac.

Both the range and the mean of estimated $k$-values increase with the increasing model’s complexity. Accounting only for wind speed, through the wind-based model CC98, leads to a spatially integrated $k$ value for $15.7 \text{ cm h}^{-1}$ (range 1-22 cm h$^{-1}$). $k$-values computed from R12, accounting for both the windshear and buoyancy flux, are on average 55% greater ($24.3 \text{ cm h}^{-1}$) with a moderate effect on the range of spatial variability. Finally including surface waves results in a spatially averaged $k$-value more than doubled as compared to the $k$-value computed from wind speed only ($35.5 \text{ cm h}^{-1}$), with a variability that is almost three times greater (range 0-68). Noteworthily, the spatial average of $k$-values computed by the wind grids (Fig. 8g Diamonds) is equivalent to the average of $k$-values computed from the spatial mean fetch (NE: 9.5 and SW: 9.3 km; Fig. 8g cross) in these specific weather conditions. The application of an average fetch would thus be relevant to estimate a spatially averaged $k$-value.

For all models, the integration of spatially resolved wind-fields may improve the accuracy of $k$ at the lake scale but accounting for wind only would underestimate both the average gas piston velocity and its spatial variability. A better understanding of wave behaviour in large lakes, using different approaches such as field and laboratory measurements, new physical models, and technical development, would therefore improve the accuracy of gas exchange estimates at the lake-water interface, at both the temporal and spatial scales. Further estimates of lake-scale CO$_2$ fluxes would require to also account for the spatial variability of CO$_2$. The question of the spatial variability of the ΔCO$_2$ is still open and remain difficult to analyse at high
Investigations of the four main processes generating the gas transfer velocity in the large Lake Geneva demonstrated the importance of considering surface waves during episodic windy events responsible for more than 44% of annual cumulated \( k_{\text{WB}} \). The in-depth study of the behaviour of the process-based models has enabled to underscore their consistent predictions at low and strong wind, especially considering the new combination and adaptation model, \( \text{SD21-fit} \). This last model significantly improves the estimation of CO\(_2\) flux when these three thresholds appear in the field: \( U_{10} > 5 \text{ m s}^{-1} \), Fetch > 15 km, and \( H_s > 0.4 \text{ m} \), making it applicable in a wide range of lake sizes. Furthermore, \( \text{SD21-fit} \) is assembled on solid theoretical bases coming from limnological and oceanic literature and allows to analyse the distribution of these four main terms (\( k_u, k_c, k_w, \) and \( k_b \)) across a variety of time scales depending on the kind of study.

To conclude, the development of high resolution gridded atmospheric models such as COSMO-1 is therefore an asset for future estimates of the gas transfer velocity and the spatial heterogeneity of lake biogeochemical cycles. Moreover, this study sheds light on the complexity of large lakes located at the interface between small, sheltered lakes and the open oceans, thus experiencing a combination of processes relevant for both small and large systems. The possibility of using process-based models in a fairly simple way with few inputs to improve the precision of the gas transfer velocity and therefore the gas flux should be supported in future research. In addition, this approach is very promising regarding long-term trends of CO\(_2\) emissions from lakes, as well as a finer estimation of fluxes during more intense episodic events.

Appendix A
Figure A1
Figure A2
Figure A3
Figure A4

Appendix B
Figure B1

Appendix C
Figure C1
Data availability. Water temperature, buoyancy flux at surface, and meteorological data are available in the datalakes – Open research data publishing platform (https://www.datalakes-eawag.ch). Water and air CO2 concentration and CO2 flux measurements are available on Zenodo (link to be added here upon acceptance).

Author contribution. PP, MEP, and DB designed the study. PP, BFC, NE, and TL collected field data and carried out data pre-processing. PP developed the model code and performed the simulation with contribution from DB. PP, MEP, and DB drafted the manuscript and all co-authors contributed to the final submitted manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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Figure 1: Conceptual scheme of the four main processes driving gas transfer velocity \( (k) \) in a large lake induced by wind and cooling events. These four processes are split into two types of \( k \): \( k\)-bubble for the bubble formation \((k_B = k_b)\) and \( k\)-no bubble for the convective mixing, wind shear and wave action term which are added \((k_{NB} = k_c + k_u + k_w)\). Below this scheme, a non-exhaustive review about conceptual approaches of \( k \)-models used in 1st Fickian law. From left to right, increase in the complexity level of \( k \)-models as well as their study site (limnological to oceanic case). All these variables are described in the section 2.4 and in Table 1.
Figure 2: Location and map of Lake Geneva with the two prevailing winds (left side) also depicted by the wind rose (top right side). The wave rose highlights the highest wavefield generated at the sampling location by the southwest wind with a larger fetch (bottom right side). Both wind and wave roses are computed with annual data from 13th June 2019 to 12th June 2020 at LéXPLORE.

Figure 3: Annual distribution of three main components used to compute $k_{sw}$ models. Orange) Wind speed at 10 m; Blue) Buoyancy flux at surface during cooling; Turquoise) Significant wave height; and these survey data observed during CO$_2$ flux measurements after quality control (+).
Figure 4: a) $k_{600}$ observed as a function of $U_{10}$ and coloured according to $H_s$ (colorbar), and the error bars produced by the uncertainty of $\Delta$CO$_2$ ($\pm$ 50 ppm) as well as the $u_{\ast}$-$k_{600}$ linear regression (solid line: see also Fig. B1); b) $k_{600}$ wind-based models (CC98, CW03 & VP13); c) $k_{600}$ process-based models (T14, S07, DM18, SD21 & SD21-fit) computed with annual data; Observed $k_{600}$ derived from CO$_2$ flux chamber measurements (+).

Figure 5: Relation $U_{10}$ vs $k_{600}$ modelled and coloured according to $H_s$ (colorbar) in a-b-c-d-e as well as coloured according to fetch distance (colorbar) in f. a) R12 integrating wind shear and convection; b) S07 integrating wind shear, convection, and wave action for fully developed waves; c) S21 integrating wind shear, convection, and wave action for not fully developed waves; d) SD21 similar to S21 adding the $k$ bubble term of DM18; e-f) SD21-fit similar to SD21 with $a_1$ and $A_b$ fitted to $k$ observed.
Figure 6: a) Cumulative $k_{600}$ modelled over an annual cycle; b) Cumulative $k_{600}$ for wind $> 5 \text{ m s}^{-1}$; c) Cumulative $k_{600}$ for wind $\leq 5 \text{ m s}^{-1}$.

Figure 7: a) Distribution of $k_{CO2}$ generated by two main processes ($k_u$ and $k_c$) in R12 and four main processes ($k_w$, $k_u$, $k_c$, and $k_b$) in SD21-fit for each season; Spring (April-May-June); Summer (July-August-September); Fall (October-November-December); Winter (January-February-March). The height of bar represents the cumulative of $k_{CO2}$ by season for both models (R12 and SD21-fit); b) Distribution of four $k$ generated by wind shear, convection, wave action and bubble enhancement ($k_w$, $k_u$, $k_c$, and $k_b$) along the annual cycle. Use of SD21-fit model where $k_u = \text{SRM}(\epsilon_u)$, $k_c = \text{SRM}(\epsilon_u + \epsilon_c + \epsilon_w) - \text{SRM}(\epsilon_u + \epsilon_w)$, $k_w = \text{SRM}(\epsilon_u + \epsilon_c + \epsilon_w) - \text{SRM}(\epsilon_u + \epsilon_c)$, and $k_b$. 

[Diagrams and graphs as shown in the image]
Figure 8: a-b) Wind fields from COSMO-1 for two episodes of Northeast and Southwest wind directions; c-d) Wave fields on Lake Geneva considering the two prevailing winds (Northeast and Southwest); e-f) Gas transfer velocity from SD21-fit; g) Boxplots of the spatial variability, at the lake scale, of $k$-values computed from CC98, R12 and SD21-fit under both meteorological conditions. Diamonds represents the spatial mean and the cross (+) the $k$-value computed from the averaged fetch distance (NE: 9.5 km and SW: 9.3 km).
Table 1: Summary of characteristics of \( k_{sc} \) models for predicting the air-water gas transfer velocity based on wind speed (CC98, CW03) and lake size (VP13), surface renewal model (T14, R12 & S07), COARSE approach (DM18) and both adapted models called SD21 and SD21-fit from a combination of S07 and DM18.

| Model   | Equation                                                                 | Method                      | Site                        | Calibrated range                       |
|---------|--------------------------------------------------------------------------|-----------------------------|-----------------------------|----------------------------------------|
| CC98   | \[ k_{600} = 2.07 + 0.215 \cdot U_{10}^{3.7} \] \[ k_{sc} = k_{600} \left( \frac{Sc}{600} \right)^{-1/2} \] | Mass Balance By gas tracer | Lake                        | Area (0.15–490 km\(^2\)) \( U_{10} < 10 \text{ m s}^{-1} \) |
| CW03   | \[ k_{600} = 0.168 + 0.228 \cdot U_{10}^{2.2} \] \[ k_{sc} = k_{600} \left( \frac{Sc}{600} \right)^{-1/2} \] | Mass Balance By gas tracer | Lake                        | Area (0.128 km\(^2\)) \( U_{10} < 6 \text{ m s}^{-1} \) |
| VP13   | \[ k_{600} = 2.51 + (148 \cdot U_{10}) + (0.39 \cdot U_{10}) \cdot \log_{10}(\text{Lake size}) \] \[ k_{sc} = k_{600} \left( \frac{Sc}{600} \right)^{-1/2} \] | Floating Chamber           | Lakes                       | Area (0.2–602 km\(^2\)) \( U_{10} < 6 \text{ m s}^{-1} \) |
| T14    | \[ k_{sc} = a_{1} \cdot (\varepsilon \cdot v)^{1/4} \cdot Sc^{-1/2} \] \[ \varepsilon = \varepsilon_{\text{Wind shear + Convection}} \] | Microstructure profiling   | Lake                        | Area (4 km\(^2\)) \( U_{10} < 10 \text{ m s}^{-1} \) |
| R12    | \[ k_{sc} = a_{1} \cdot (\varepsilon \cdot v)^{1/4} \cdot Sc^{-1/2} \] \[ \varepsilon = \varepsilon_{\text{Wind shear + Convection}} \] | -                           | -                           | Following S07                         |
| S07    | \[ k_{sc} = k_{sc-NB-S07} + k_{sc-B-W97} \] \[ k_{sc-NB} = a_{1} \cdot (\varepsilon \cdot v)^{1/4} \cdot Sc^{-1/2} \] \[ \varepsilon = \varepsilon_{\text{Wind shear + Convection + Eddy covariance}} \] | Eddy covariance            | Ocean                       | Area (>100’000 km\(^2\)) \( U_{10} < 20 \text{ m s}^{-1} \) \( \text{Wave (0–10 m)} \) |
| DM18   | \[ k_{sc} = (k_{NB} + k_{B}) \cdot (\frac{Sc}{600})^{-1/2} \] \[ k_{NB} = A_{NB} \cdot U_{atm} \] \[ k_{B} = (A_{B} / \rho_{b}) \cdot U_{atm}^{5/3} \cdot (g \cdot H_{b})^{2/3} \] | Eddy Covariance            | Ocean                       | Area (>100’000 km\(^2\)) \( U_{10} < 30 \text{ m s}^{-1} \) \( \text{Wave (1–10 m)} \) |
| SD21   | \[ k_{sc} = k_{sc-NB-S07} + k_{sc-B-DM18} \] | Floating Chamber           | Lake                        | Area (582 km\(^2\)) \( U_{10} < 16 \text{ m s}^{-1} \) \( \text{Wave (0–1.2 m)} \) |
| SD21-fit | \[ k_{sc} = k_{sc-NB-S07} + k_{sc-B-DM18} \] | -                           | -                           | -                                       |

870  **Cole and Caraco (1998), CW03 Crusius and Wanninkhof (2003), VP13 Vachon and Prairie (2013), T14 Tedford**
et al. (2014), R12 Read et al. (2012), S07 Soloviev et al. (2007), DM18 Deike and Melville (2018).

Table 2: RMSE of k models for all wind speed ($U_{10}$), $U_{10} < 5$ m s$^{-1}$ (i.e., LW) and $U_{10} \geq 5$ m s$^{-1}$ (i.e., SW).

| RMSE | CC98 | CW03 | VP13 | T14 | R12 | S07 | DM18 | SD21 | SD21-fit |
|------|------|------|------|-----|-----|-----|------|------|---------|
| All $U_{10}$ | 9.8  | 6.5  | 6.7  | 8.6 | 7.5 | 6.2 | 7.3  | 6.2  | 5.2     |
| $U_{10} < 5$ m$^{-1}$ | 3.2  | 4.2  | 4.5  | 2.9 | 3.3 | 3.3 | 3.5  | 3.3  | 3.2     |
| $U_{10} \geq 5$ m$^{-1}$ | 22.8 | 12.8 | 12.7 | 19.8| 16.6| 13  | 15.9 | 13.1 | 10.5    |

Table 3: Seasonal to annual CO$_2$ flux estimation (mmolC m$^{-2}$ d$^{-1}$) from k-models (with CE) and without chemical enhancement as well as their deviation from SD21-fit. CE was accounted for only seasons when pH > 8.4.

| Period      | $\Delta$CO$_2$ | CC98 | CW03 | VP13 | T14 | R12 | S07 | DM18 | SD21 | SD21-fit |
|-------------|----------------|------|------|------|-----|-----|-----|------|------|---------|
| Spring      | -5.1           | -4.2 | -5.9 | -10.5| -5.5| -5.4| -6.2| -5.5  | -5.6  | -6.9    |
| Spring-CE   | -5.9           | -8.7 | -11.4| -7.0 | -2.2| 7.9 | -2.4| -2.4  | -8.5  |         |
| Summer      | -145           | -9.0 | -8.3 | -23.7| -13.4| -12.8| -13.4| -12.8 | -13.2 | -16.3   |
| Summer-CE   | -17.4          | -18.9| -27.8| -20.1| -20.7| -20.3| -20.4| -22.7 |       |         |
| Fall        | 350            | 29.0 | 41.2 | 74.8 | 47.6| 42.9| 52.2| 48.4  | 52.1  | 64.5    |
| Winter      | 475            | 43.1 | 63.1 | 108.4| 65.7| 64.5| 71.1| 64.1  | 69.7  | 86.0    |
| Annual      | 157            | 14.7 | 22.5 | 37.1 | 23.6| 23.4| 25.9| 23.5  | 25.7  | 31.8    |
| Annual-CE   | 157            | 12.2 | 19.2 | 36.0 | 21.6| 21.3| 23.7| 21.2  | 23.5  | 29.8    |

| Annual gCm$^{-2}$ yr$^{-1}$ | - | 64.6 | 98.8 | 163.1 | 103.6 | 102.8 | 113.9 | 103.3 | 113.0 | 139.7 |
| Annual-CE gCm$^{-2}$ yr$^{-1}$ | - | 53.6 | 84.3 | 158.2 | 94.8 | 93.4 | 104.1 | 93.3  | 103.3 | 131.1 |

| Deviation from SD21-fit | - | -54 % | -29 % | +17 % | -26 % | -26 % | -18 % | -26 % | -19 % | -    |
| Deviation-CE from SD21-fit | - | -59 % | -36 % | +21 % | +28 % | +29 % | +21 % | +29 % | +21 % | -    |
Figure A1: Schematics of eosFD operation (eosense.com) followed by its mini-platform construction and its positioning for measurements in the field (Lake Geneva at LeXPLORE platform). The raft design also complies with recommendations to minimize artificial turbulence induced by the chamber’s walls, with 10 cm long-edges entering the water (Vachon et al., 2010).

Figure A2: a) Classical floating chamber; b) Floating chamber with 10 cm long-edges; c) Platform design used in this study: 10 cm long-edges, rounded-edges, and flat and long water wings.
Figure A3: Visualisation of 304 observed $k_{obs}$ during the five periods of flux measurements (i.e., 13th–14th June 2019, 27th–28th August 2019, 1st–5th October 2019, 18th–20th December 2019, and 20th–26th February 2020).

Figure A4: a) Raw outputs of the cosFD during one period of CO$_2$ flux measurements; $\Delta$CO$_2$ between both cavities of measures (atmosphere cavity and chamber cavity) (blue line); Standard deviation of each cavity between two automated flushing (30 minutes of interval), Chamber cavity (red line), Atmosphere cavity (yellow line); and the CO$_2$ flux (black dash line). b) Temporal evolution of $U_{10}$ and $H_s$ during the same period than CO$_2$ flux measurements. Increase in flux on 25th February corresponding to increase in wind speed and waves.
Figure B1: a) Comparison of Soloviev et al. (2007) and Deike and Melville (2018) for the first order function of friction velocity at the water side ($u_{\text{wall}}$) (blue points) and at the atmosphere side ($u_{\text{atm}}$) (green points) with their linear regression (black line); the linear function of Vachon and Prairie (2013) for a lake size of 582 km$^2$ (yellow points) as well as the linear regression from ; b) Visualisation of S07 with empirical parameterization of bubble term (Woolf, 1997) regardless of wave height in function of wind speed at 10 m; c) Visualisation of DM18 in function of wind speed, only effect of bubble term from 10 ms$^{-1}$.

Figure C1: Annual evolution of 3 main inputs of $k$-models; Wind speed at 10 m ($U_{10}$); Buoyancy flux at surface ($B_o$); Significant wave height ($H_s$).
