Response to Anonymous Referee #1

We thank you very much for your constructive comments and suggestions.
Below the reviews are reproduced in black font, our replies are interspersed in **blue** while preliminary updates of the text are in ***green***.

In this well-constructed study, Perolo and colleagues evaluated the performance of various empirical and process-based gas exchange (k) models using a combination of high- frequency k and weather measurements, intending to find the best model to accommodate high wind and high wave heights conditions often found in large lakes such as Lake Geneva. To be able to account for the major processes affecting k on such a large lake (i.e. wind shear, buoyance flux and wave motion including bubble enhancement), the authors cleverly combined and adapted existing process-based models first developed for the ocean. The chosen adapted model was proven the most accurate and flexible to a wide range of wind speeds and wave heights.

Investigating k at high wind and wave heights conditions is an important and overlooked aspect of k dynamics in the context of episodic events. The addition of waves and bubble enhancement to lakes k models is novel, to my knowledge. I particularly liked the cumulative k analysis as it clearly demonstrates the disproportionally important role of rare periods of high winds and waves. It also elegantly shows how each model responds as a whole to the distributions of wind and wave that actually occurs on a large lake. I also think that Figure 1 is very useful and accurately summarizes the main processes and the predictive models with their respective variables used.

Overall, I found the manuscript well written, well organized and easy to follow. The references are appropriate and the methods clear. I only have a few general and specific comments that could potentially improve the manuscript.

Reply: We very much appreciate this overall positive assessment.

General comments:

1. The lack of spatial integration (due to only one measurement station) is only discussed briefly in the conclusion. Wind and waves fields are different in other parts of the lake and this can have a large impact on the conclusions made regarding CO2 fluxes. I suggest the authors expand this part and move to the discussion section.

Reply: Following your comment and those of the second reviewer, we developed this aspect in the discussion, by computing k-values over the lake surface using the COSMO wind-model of Meteoswiss for two representative's dates with windspeeds > 10 m s^{-1}. Since this is mostly a model-based illustration of the spatial implications of k-models, this section was added to the discussion. Please find below the section added, after section 4.2.2.

"4.3 Wind and wave field on Lake Geneva and their impact on the spatial integration of k_{600}

SD21 and *SD21-fit* were built on the basis of a single measurement point on the lake, just as for most of the existing *k* models. 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 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° 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 $H_s > 0.4$ 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_s values > 0.4 m, leading to *k*-values as computed by *SD21-fit* as high as 68 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 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 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 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 frequency in large lakes.

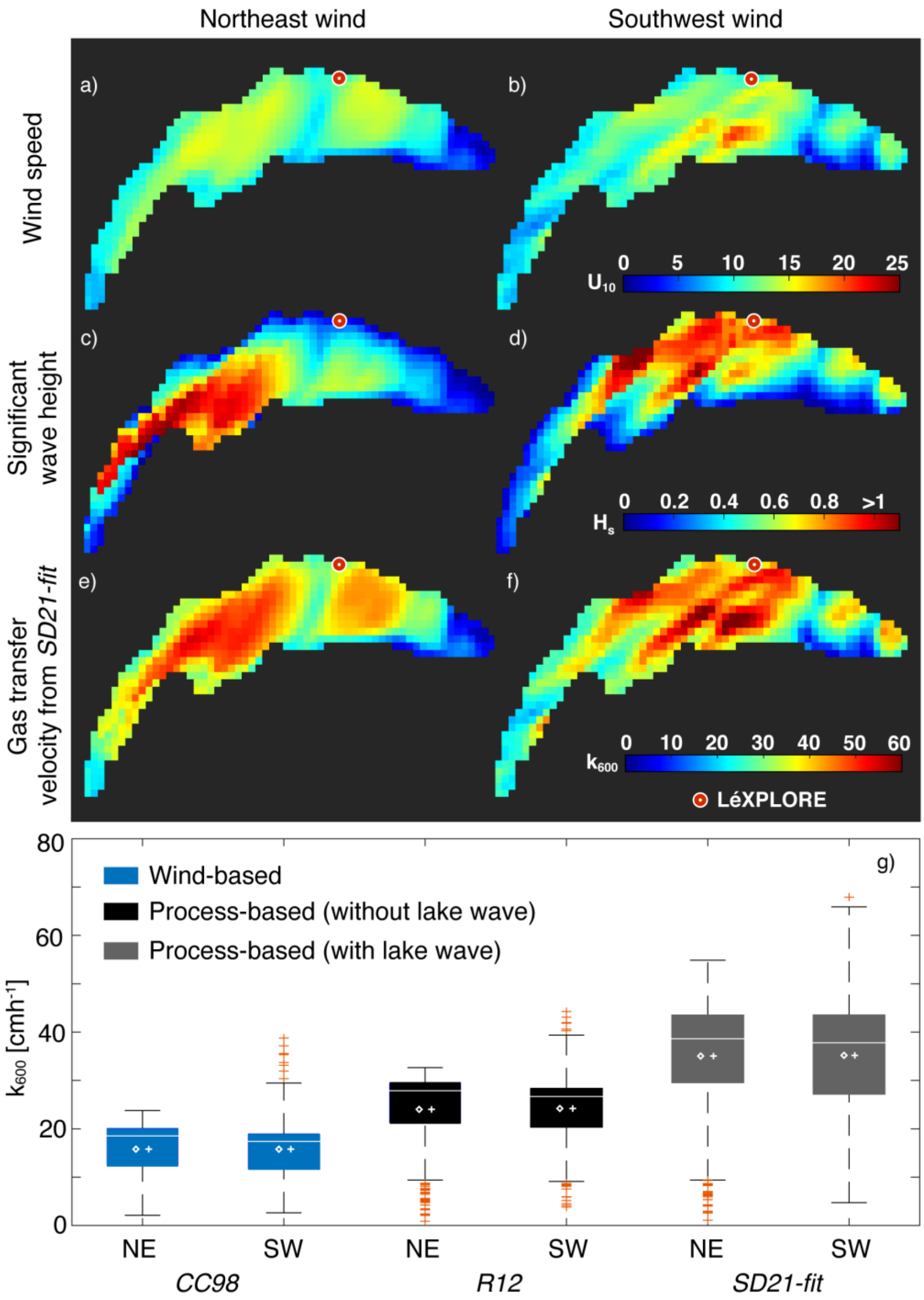

Figure 8: a-b) Wind fields from COSMO-1 in situations of Northeast and Southwest wind directions; c-d) Wave fields on Lake Geneva considering the two prevailing winds (Northeast and Southwest); e-f) Gas transfer velocity from *SD21-fit*; g) Boxplots of the spatial variability, at the lake scale, of *k*-values computed from *CC98*, *R12* and *SD21-fit* under both meteorological conditions. Diamonds represents the spatial mean and the cross (+) the *k*-value through computed from a fixed average fetch distance (NE: 9.5 km and SW: 9.3 km).

Finally, we will clarify in section 4.2.2 that our estimate of CO2 fluxes remains a coarse estimate with main goal to scale the effects of wave integration on annual CO2 fluxes. More data would be needed to provide an accurate estimate of CO2 emissions at the lake scale.

2. In undersaturated CO2 conditions (high pH), there is the possibility at CO2 fluxes are enhanced chemically (aka chemical enhancement factor). This usually happens during productive periods (summer with undersaturated pCO2), where CO2 is rapidly consumed chemically (hydration) at the very surface of water (Wanninkhof and Knox, 1996), enhancing the CO2 influx from the atmosphere, but not affecting pCO2 measurements at a deeper depth. If this is the case (at about pH > 8), it would result in an overestimation of observed k values measured from pCO2 and flux chambers, especially under calm wind conditions. In the manuscript, the chemical enhancement factor was not taken into account, and I think this should be justified. I do not think this would change the main conclusions of the paper, but it may potentially slightly affect the parameterization (SD20-fit) and the models evaluations.

Reply: We agree that the notion of chemical enhancement, which is likely to occur in Lake Geneva in the summer period, should be specified. Chemical enhancement is not relevant for the time-periods on which we tested the different k-models.

Indeed, our analysis of k-models accuracy is based on data recorded during the periods of oversaturation (December and February, i.e.: when pH data are well < 8) since summer data did not pass the quality control (CO2 flux too low or delta CO2 too low). Consequently, the parameterization of the SD20-fit (now SD21-fit) model should not be impacted by the absence of integration of the chemical enhancement factor. Besides, our parametrization mainly impacts the gas exchange velocity results for wind > 5 ms^{-1} when chemical enhancement was not expected to have a significant effect (see for instance Figure 2 in Wanninkhof and Knox, 1996).

Following this comment, we added a few sentences explaining these reasons in section 2.3 (line 161):

"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) where such a process should not affect calculations."

However, we agree chemical enhancement should be accounted for summer and annual estimates of k-values, such as on their consequences on estimated CO2 fluxes (Table 3). If this factor is to be taken into consideration at high pH, it should be incorporated into any k-models whether wind- or process-based.

As a reminder, the formulation of Wanninkhof and Knox (1996) shows that the chemical enhancement factor α depends on the stagnant boundary layer thickness z following:

$$\propto \; = T\,/((T-1)+\tanh(Q\cdot z)/(Q\cdot z)) \qquad (10)$$

While z itself depends on k as follows:

$$z = D/k \qquad (1)$$

Therefore, estimates for chemical enhancement would themselves depend on the chosen k-models, with even further consequences on estimated CO2 fluxes.

We therefore computed estimated fluxes with and without accounting for the chemical enhancement and completed Table 3 and section 4.2.2 as below:

"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 CO2 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 LéXPLORE platform from the different models at hourly timestep, 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), a constant pCO_2 in the atmosphere (400 µatm). 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 CO_2 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 CO_2 up to the lake surface, while low up-taking gas fluxes occur in spring and summer, when primary production depletes surface CO_2 below saturation. However, annual estimates of net CO_2 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 ΔpCO_2 gradient (100-200 µatm) and wave occurrence are limited. Adding chemical enhancement 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 ΔpCO_2 (475 µatm) and surface waves occurrence are higher (Table 3; Fig. 7). Therefore, while high wave events represent only 6 % of the total surface waves occurrence ($H_s > 0.4$ m) at the measuring point, an incomplete consideration and description of their contribution may lead to an annual flux underestimation of about 20–25 %."

Table 3: Seasonal to annual CO_2 flux estimation (mmolC m^{-2} d^{-1}) from *k*-models (with and without chemical enhancement (CE) during influxes and monthly ΔCO_2 average (µatm) as well as their deviation from *SD21-fit*.

Period	***ΔCO_2***	***CC98***	***CW03***	***VP13***	***T14***	***R12***	***S07***	***DM18***	***SD21***	***SD21-fit***
Spring	-51	-4.2	-5.9	-10.5	-5.5	-5.4	-6.2	-5.5	-5.6	-6.9
Spring-CE		*-5.9*	*-8.7*	*-11.1*	*-7.0*	*-7.2*	*7.9*	*-7.4*	*-7.4*	*-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*	*-20.1*	*-20.7*	*-20.3*	*-20.4*	*-22.7*
Fall	350	29.0	41.2	74.8	47.6	47.9	52.2	48.4	52.1	64.5
Winter	475	43.1	63.1	108.1	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		*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*	*93.2*	*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 %*	*-*

3. Significant wave heights (Hs) were not measured but predicted from U10 and fetch. Do the authors have any idea of uncertainties associated with the predicted Hs?

Reply: Similar comment was also raised by reviewer 2. We are unfortunately unable to provide a quantification of the uncertainties from field observations.

Therefore, we rephrased this section from the methodology concerning Hs and its definition (line 132-136):

"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).”

We instead stress that such measurements are needed in large lakes to better constrain the air-water exchanges under various surface roughness in the new section (4.2. see below)

4. The terms *SD20* and *S20* (in Fig. 5) are presented in the same way as the other published models (i.e. first letter of author name and the year of publication). In this case, does SD stands for Soloviev and *D* for Deike? and why 20? Please explain in the text and/or in the figure caption.

Reply: We added an explanation for the name given to the model on line 251 at the end of the methodology section.

“Our adapted model for lake includes a refined parametrization of the wave action term ε_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_{600} observations (a_1 = 0.33 and A_B = 3 10^{-5} m^{-2} s^2).”

The 20 there referred to the year when this article was initially written. We will transform the name to *SD21* throughout the text and figures.

Specific comments:

L42: I think it should be Cole and Caraco (1998), as it is commonly called, instead of Cole et al.

Reply: We agree. We made a reference mistake. It will be changed for Cole and Caraco.

“… starting with the Cole and Caraco (1998) seminal paper…”

L138: As this is a novel method, I think more details on the automated (forced diffusion) flux chamber should be given. How this chamber is different from the more traditional floating chamber?

Reply: Following your comment and that of the second referee, we added a few lines (after line 142) to the flux measurement methodology and created a new figure for the appendix to show the different designs.

“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 eosFD, 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 CO_2 sensor for the two measurements limits the need for intercalibration between CO_2 sensors. We tested the performance of the floating chamber by comparing the standard deviation of the CO_2 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.”

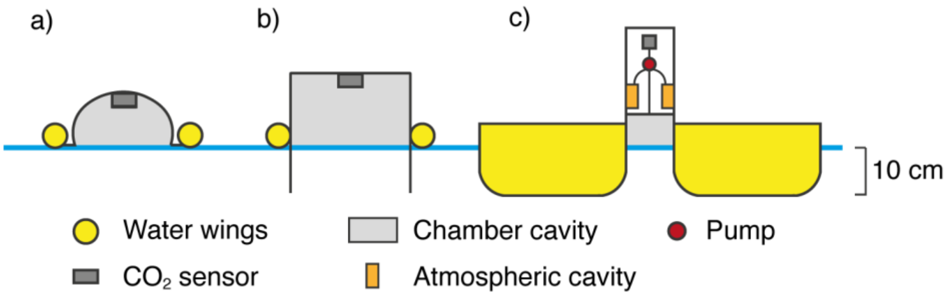

Figure A2new: a) Classic floating chamber; b) Floating chamber with 10 cm long-edges; c) Platform design used in this study: 10 cm long-edges, rounded-edges, and flat and long water wings.

L235: I don’t know what the “^” symbol means here?

Reply: We will remove this typo.

L280. Missing dot.

Reply: We will correct this typo.

L362: But specific calibration (a and A) would also be needed in process-based models like it is the case for Lake Geneva.

Reply: We agree with this remark. The process-based models can also have a recalibration with these two empirical parameters (a_1 and A_b) in different systems. However, they remain more consistent than wind-based models even without refining these parameters. Also, we had to recalibrate, as they had never been implemented in lakes with the surface wave effect included. Therefore, we will remove this sentence.

Fig. C1. The models used only negative buoyancy flux, which induces turbulence by convection. I wonder what is the effect of positive buoyancy during heating on k. Could it reduce the effect of wind shear as it suppresses turbulence?

Reply: This is an important point that we now discuss in the manuscript. We now clearly state that our model only considers buoyancy flux when this terms directly enter into the turbulent kinetic equation as production term. We acknowledge that a positive buoyancy flux should reduce the effect of wind shear turbulence production to a yet to be established extent that would depend on the near surface stratification. Assuming that typically 20% of the production term is transferred to mixing (Ivey et al. 2008), a first order approximation would be to reduce the effect of the wind shear turbulence production by a similar amount in our equation in the case of net heating of the lake. Another important aspect is the role of stratification that offset the measured CO2 values with respect to those at the interface. This subject deserves an attention beyond the scope and data availability of this work.

We added this sentence in section 2.4.2 (line 231):

“A second source of dissipation at the surface is the convection (ε_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):”

Change of this reference

Simon A.: *Turbulent mixing in the surface boundary layer of lakes.* PhD thesis no. 12,272. Swiss Fed. Inst. Technol. (ETH), Zurich, 1997.

Add references

Ivey, G.N., Winters, K.B., and Koseff, J.R.: Density stratification, turbulence, but how much mixing? Annu. Rev. Fluid Mech., https//doi.org/10.1146/annurev.fluid.39.050905.110314, 2008.

Other changes

We add in table 1 (CC98 for Calibrated range) Area (0.15-490 km^2) because CC98 had taken other lake in the literature to perform his model.

We found a typo in the caption of the figure 6 with a sign error between 5b and 5c (greater and smaller). Here is the correction:

Figure 6: a) Cumulative k_{600} *modelled over an annual cycle; b) Cumulative* k_{600} *for wind* > 5 *m* s^{-1}*; c) Cumulative* k_{600} *for wind* ≤ 5 *m* s^{-1}*.*

We also found small errors in the labels of y axes of figure 5 which will be corrected.