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METHANE EBULLITION PREDICTORS FOR RESERVOIR EMISSIONS COMBINING MODELING AND MACHINE LEARNING

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Abstract − Freshwater reservoirs have been reported as contributors to greenhouse gas emissions to the atmosphere, but their contribution is uncertain. The produced methane in such systems can be transported to the atmosphere by ebullition and diffusion. Ebullition is highly variable in space and time, as the formation of bubbles depends on physical, chemical and biological factors. Therefore, there are still gaps in the processes underlying ebullition for accurate emissions estimation. Data scarcity is one of the problems faced to comprehend the process, hence the application of lake models can assist to overcome this issue. In addition to the physical based model, machine learning methods can be applied to provide predictions while classical approaches are still not resolved. Thus, in this study simulation results of a calibrated 3D hydrodynamic-water quality model (temperature, dissolved oxygen, chlorophyll-a and water depth) were combined with observed ebullition data to bring insights about methane release from a drinking water reservoir in Paraná - Brazil, then machine learning techniques were explored to predict ebullition (MatLab toolbox). A supervised machine learning regression model confirmed that the magnitude of ebullition cannot be well predicted by considering only water quality conditions (RMSE = 0.17 and R2 = 0.34). On the other hand a classification model predicted the occurrence of ebullition events independent of its magnitude with an accuracy of 77%, indicating a potential use of the combined techniques of data analysis. This study provided preliminary results on a new approach for gas ebullition assessment, and improvements are still required.

Key-words – Methane ebullition; Delft3D; machine learning.

INTRODUCTION

Freshwater reservoirs are worldwide constructed for several purposes, including water supply, energy generation, and flood control. Nevertheless, such systems have also been reported as contributors to greenhouse gas emissions to the atmosphere, as methane and carbon dioxide. Deemer et al. (2016) for instance, estimated that reservoirs emit an amount of 606.5 $TgCO_2$ equivalent per year as methane, whereas Rosentreter et al. (2021) pointed out that methane emissions from reservoirs amount to 513.4 Tg CO_2 equivalent per year.

The amount by which reservoirs contribute to greenhouse gas emissions to the atmosphere is being debated in the literature. The sources of uncertainties for the estimates can be multiple, resulting from monitoring strategies and measurement techniques, data availability, and a lack of a

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comprehensive understanding of the processes governing the dynamics of these gases in the water bodies.

In reservoirs, methane and carbon dioxide are mainly produced from bacterial degradation of the organic matter retained. Methane formation is restricted to anoxic conditions, which commonly prevail in bottom sediments. From the sediment, the gas can be transported to the atmosphere by ebullition (bubbles) and diffusion, where bubble mediated transport is reported to account on average for 65% of methane emissions (Deemer et al., 2016).

Ebullition is highly variable in space and time, as the formation of bubbles depends on biological activity for gas production and oxidation (Valentine et al., 2004), quality and quantity of organic matter (Wik et al., 2018; Praetzel et al., 2019), sediment properties for gas storage (Liu et al., 2016) and bubble mobility (Jain and Juanes, 2009), and on external forcing that can trigger the release of bubbles (such as hydrostatic pressure reduction, Maeck et al., 2014). A number of studies have investigated ebullition with the aim to identify the main drivers, however, there are still gaps in the processes underlying ebullition and how to combine spatial and temporal variabilities for more accurate emissions estimation.

In addition, only few studies have linked water quality parameters with ebullition. Beaulieu et al. (2019) for instance, suggested that methane emissions (ebullition + diffusion) from reservoirs tend to increase with increasing nutrient loading and eutrophication. Nonetheless, the statistical models applied to represent ebullition as a function of total phosphorus, total nitrogen, and chlorophyll-a (Chla), with Chla being the best predictor with a determination coefficient (R^2) of the adjusted model of 0.3 (Del Sontro et al., 2018).

Data scarcity is one of the problems faced in order to comprehend the process and define related parameters (Soares et al., 2019). In this regard, the application of models can assist to overcome this issue, regardless of the effort for setting them up. In addition, boundary conditions to feed a reservoir model usually are easier to obtain and are widely available (e.g. meteorological conditions and inflow characteristics), than monitoring data from reservoirs, which becomes worse in tropical regions (Winton et al., 2019). Further, models can provide additional insights from other processes that can be linked with ebullition events.

In addition to the mechanistic or physical based model, statistical or data-based models (such as the machine learning methods) are being used as tools to identify patterns, for predictions, and for providing insights into complex processes that are still not easily resolved by mechanistic approaches. Faraj and Shen (2018) presented an overview of the machine learning methods used for water resources applications for water quality issues, monitoring strategies, and variable prediction. On one hand, data-based models can provide efficient and accurate results with low costs, but on the other hand they are dependent on the amount and quality of data available and the output interpretation cannot be easily linked to specific processes. Nevertheless, recently hybrid models have been developed with the possibility of integrating physical concepts to existing data-based model, such as the framework proposed by Willard et al. (2020) and the application to model lake temperature presented by Karpatne et al. (2017).

In this study, simulation results of a 3D model are assessed in combination with observed ebullition data to bring insights about the methane release from a drinking water reservoir in Paraná - Brazil. The aim of this research is to explore machine learning techniques to identify correlations, patterns and trends in observed monitoring data, complemented with continuous data provided by the reservoir hydrodynamics and water quality model.

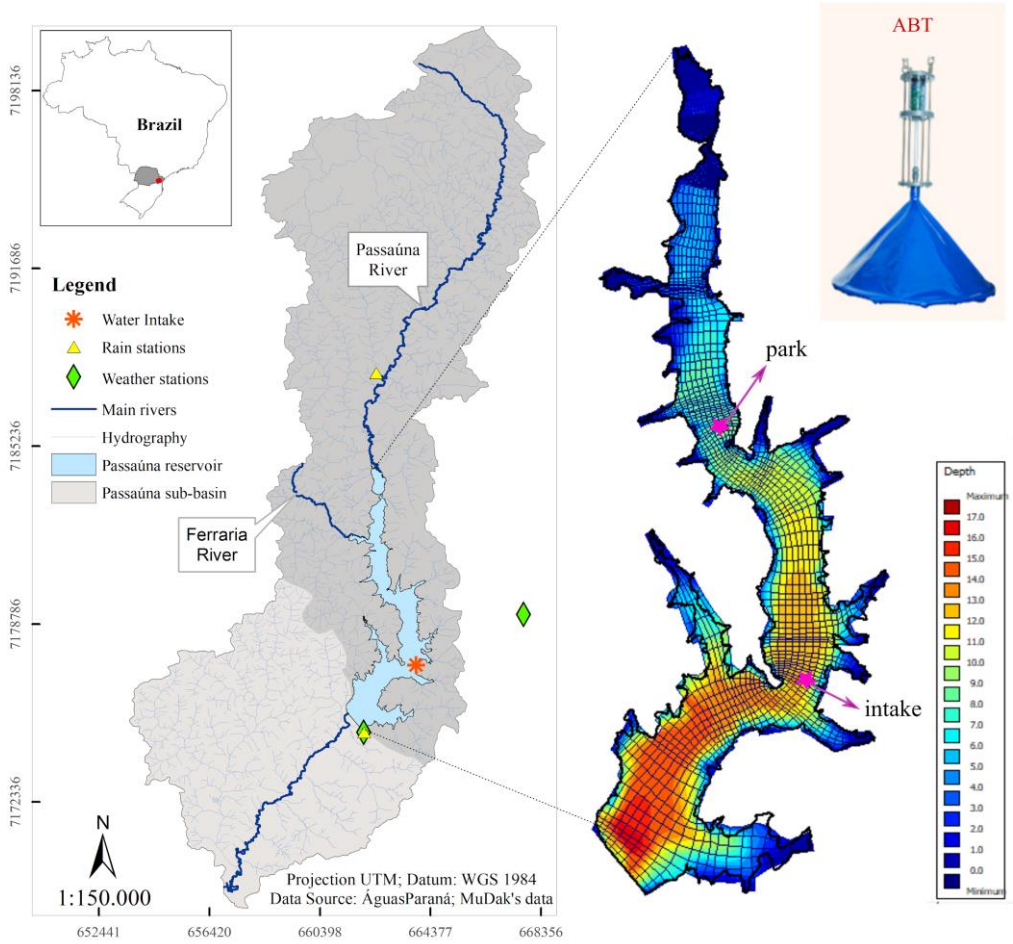
METHODS

The study site is Passaúna Reservoir, which is located in the southern region of Brazil near Curitiba-PR city (coordinates 25.5274°S and 49.3917°W). The reservoir was built in 1989 for water supply purposes and it is managed by the Companhia de Saneamento do Paraná (Sanepar) company. The reservoir has mesotrophic characteristics (IAP, 2017), polymictic mixing regime (Ishikawa et al., 2021), an average depth of 8 m with small variations of water level (on average < 1m) in regular climate conditions, and it is influenced by subtropical climate. Approximately 70% of the sub-basin contributes to runoff to the reservoir, in which Passaúna River and Ferraria River are the main water inflows. Agriculture activities have the largest share to the land-use at the basin representing 40% of the total area (Sotiri et al., 2021).

Continuous high resolution (at 1 min time interval) ebullition time series were recorded at 2 monitoring locations (Park and Intake, see Figure 1) in the reservoir during the years 2017–2019. Ebullition was shown to vary seasonally with the stratification conditions of the reservoir and to be triggered during periods of decreasing atmospheric pressure and increased wind velocity (Marcon et al., 2019). Additionally, at the intake location water temperature profiles were measured with 11 temperature loggers (Minilog-II-T, Vemco), with 1 m spacing among each other, starting 1 m from the bottom (sampling interval of 1 min, precision of ± 0.1ºC and 0.01ºC resolution). Together with the thermistor chain, two dissolved oxygen loggers (miniDOT, Precision Measurement Engineering, Inc - resolution of 0.01 mg L^{-1} and an accuracy of ± 0.3 mg L^{-1}, time resolution of 5 min) were deployed with the second and last temperature logger (from bottom). At a depth of around 1.5 m, a chlorophyll-a fluorometer (nanoFlu - TriOS Optical Sensors), measuring in 15 min temporal resolution later averaged to daily means, was deployed at the same location, measurements were calibrated with samples.

The grain size of bottom sediments in the reservoir was characterized as having a homogeneous distribution from park location towards the dam, in which silt-clay account for more than 80% of sediment fraction, whereas the distribution of Loss on Ignition (indicating the fraction of organic matter) ranged mainly between 10–20% (Sotiri, 2020). The monitoring locations Park and Intake have similar sediment conditions regarding organic matter content and grain size distribution. As the reservoir had mild water level variation during the monitored period, it favors the investigation of the effect of water quality parameters (such as dissolved oxygen, temperature, and chlorophyll-a) on gas ebullition.

Figure 1 − Location of Passaúna reservoir, the main meteorological stations of the watershed. At the right hand side, it shows the reservoir's bathymetry and the grid adopted for Delft3D modeling. At the top right corner is the automatic bubble trap (ABT) deployed for recording ebullition, details regarding the device can be found at Maeck et al. (2014).



Delft3D is a 3D model, the hydrodynamic module FLOW (Deltares, 2013) solves the Reynolds Averaged Navier Stokes equations, using the hydrostatic assumption within a finite difference grid. Results include flow velocities, temperature, and substance transport. The module is forced by meteorological and hydrological conditions, in and outflows and inflow temperature, and measured water level. FLOW simulations are used as the base for the water quality module WAQ. The coupled water quality model solves, for user defined parameters, substance transport and biochemical and biological processes. For Passaúna Reservoir a curvilinear grid was implemented (Figure 1) in a resolution of ~ 40×40 m at the horizontal, and over depth the z-layer approach was used with 20 layers of 0.83 m resolution, bathymetry was provided by Sotiri et al. (2019) through a survey. Inflow discharges and temperature were modeled and provided by Krumm et al. (2019), and meteorological data were collected from the stations shown in Figure 1. The simulation started on 01 Aug 2017, when the reservoir was fully mixed, the first 7 seven months were the spin-up period, thus the time period analyzed in this study is from 01 Mar 2018 to 28 Feb 2019.

From the simulation, daily averaged values of dissolved oxygen, Chla, and water temperature were extracted for further analysis. The deepest layer of the pointed grid cells in Figure 1 were selected. Measured gas ebullition was converted in daily time intervals as the total volume of gas measured per unit area over one day. The statistical analysis was done with MatLab R2019a, where the machine learning toolbox was used for the regression models applied for ebullition prediction (Regression Learner App) and for the classification (Classification Learner App) to define the

occurrence of ebullition events. Methods with the best results were selected based on the root-mean-square-errors (RMSE), R^2 and accuracy.

RESULTS

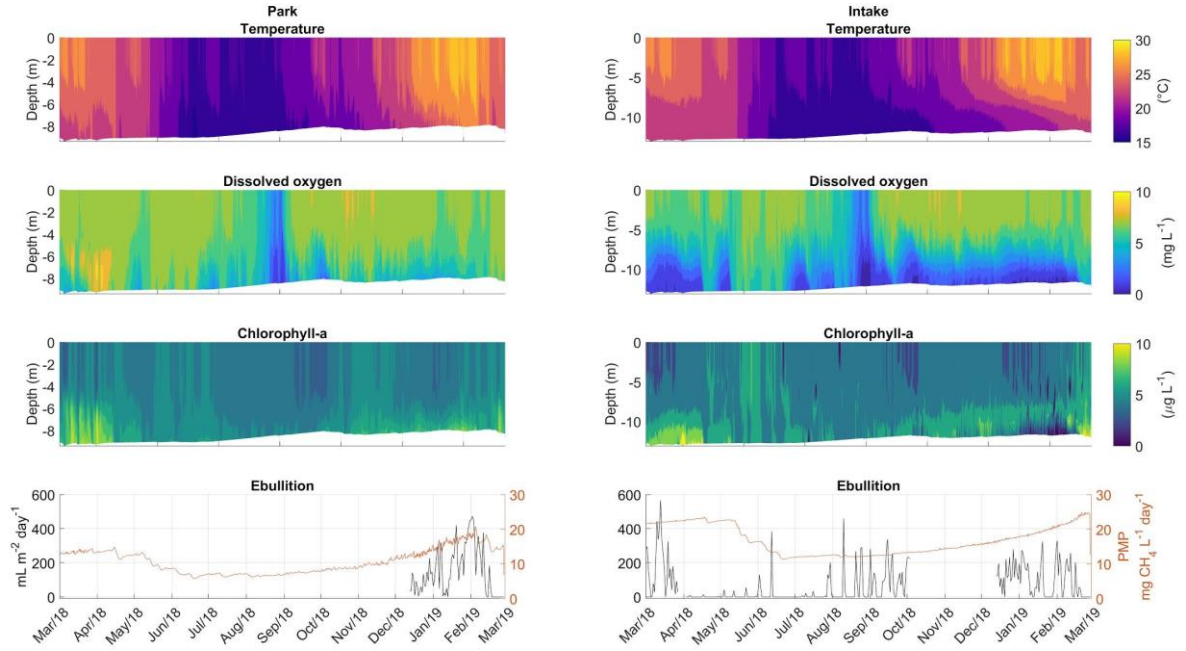
Based on the literature findings on variables influencing ebullition, Delft3D model's parameters selected for this study were water temperature, dissolved oxygen and Chla (Aben et al., 2017, Del Sontro et al., 2018, Langenegger et al., 2019 and Marcon et al., 2019).

Model results were compared to the observations, and had errors in the same range presented in other studies (e.g. Mao et al., 2015). RMSE were 0.77 °C in temperature, 1.86 mg L^{-1} in dissolved oxygen and 2.20 µg L^{-1} in chlorophyll-a. The seasonal pattern at the Intake was reproduced as observed (Ishikawa et al., 2021), where the simulated temperature started stratified, and during autumn and winter there was intermittent mixing, which alternated with slight stratification (shown in Figure 2). At Park location the model showed similar behavior, but due to the shallower depths, the bottom temperatures were higher and the upper mixed layer reached the bottom. In those cases, the vertical profile of temperature was mixed at Park while stratified at the Intake. Dissolved oxygen concentrations followed the same pattern of temperature stratification, bottom concentrations at Intake decreased to zero during stratification, while Park did not present anoxic conditions. Observed chlorophyll-a did not present great variations over the year at the surface, similar to simulation results in both locations, the larger concentrations at deeper layers are most likely to be phytoplankton transported by lateral flows.

Ebullition is well described in literature for its large temporal variability. Measured data at Intake location had fewer gaps (due to energy break) in the recorded time series, with data also available for the winter time (see bottom panel in Figure 2). In the measured period, ebullition was observed to occur for successive days and to have periods ranging from days to weeks of no event of gas release recorded. During the months of mixing and weak stratification with colder bottom water the occurrence of ebullition events was reduced with prolonged periods of no ebullition. Comparing ebullition from Park and Intake for the period with data available at both locations (Dec. 2018 - Mar. 2019), shows that both had a similar total amount of gas emitted (7.2 L at Park and 7.4 L at Intake). However, with Intake having a higher total amount of gas emitted from February–March 2019 whereas Park had more intense ebullition from December 2018–February 2019.

Temperature has manifold effects on ebullition, by influencing the gas solubility, the diffusion of substances, and the bacterial activities. Aben et al. (2017) found a significant exponential correlation between sediment temperature and methane ebullition from different aquatic systems, and Yvon-Durocher et al. (2014) attributed the seasonal variation of methane emissions to changes in methane production. Intake has a slightly higher potential of methane production compared to Park. Nevertheless, Park is a shallower location and the bottom water, and thus the sediment temperature, gets warmer earlier in the end of the colder season which may enhance methane production. On the other hand, the reduced water depth has the effect of favoring ebullition, as the minimum required partial pressure of methane to start bubble formation is smaller. The combination of both temperature and water depth might favor higher ebullition rates at Park at the beginning of the warmer season.

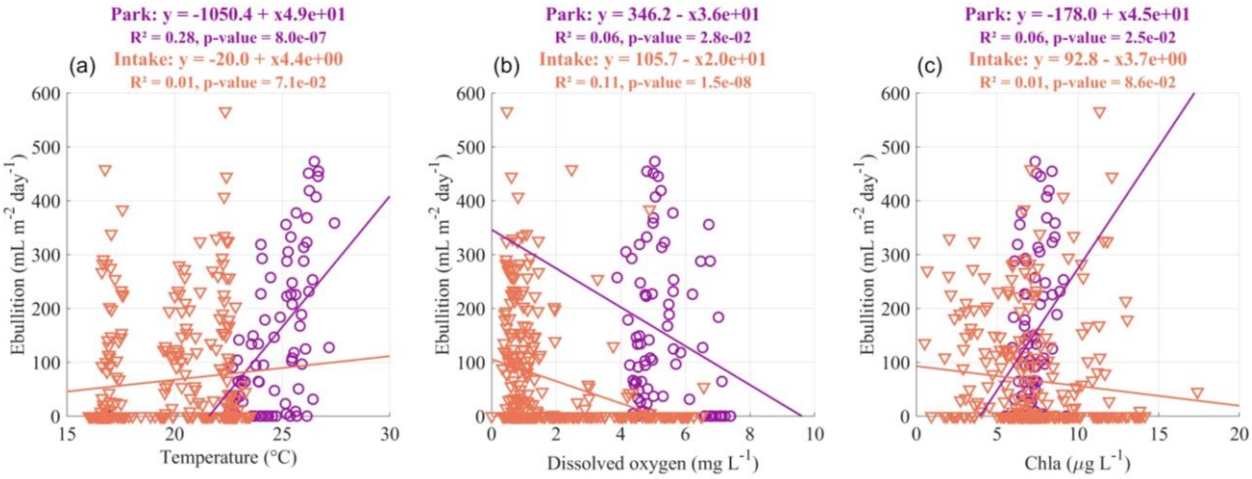
Figure 2 - Simulated time series of water temperature, dissolved oxygen and Chla at park and intake monitoring locations. Bottom panels are ebullition time series measured at each location and the potential of methane production. The potential of methane production (PMP) was obtained from Hilgert et al., (2019) of the top 21 cm layer of incubated sediments and calculated for the entire period as temperature dependent according to the equation $PMP(T) = PMP_{20^\circ}\, 10^{\theta(T-20)}$ from Wilkinson et al., (2019). Where T is the sediment temperature, assumed to be at the same temperature as the bottom water and the coefficient θ was assumed as 0.045 as proposed by the authors for incubated sediments.



Considering a longer period, one hypothesis is that the transport of methane out of sediment by diffusion plays an important role in the amount of gas accumulated in the sediment available for bubble formation. The diffusion of methane depends on the diffusivity of methane and on the concentration gradient across the sediment water interface. The concentration of methane in the bottom boundary layer can be modified by overturning events, internal motions and turbulence, and by the oxidation/production of methane. Stratified periods (e.g. more stable water column) with anoxic conditions of bottom water favors the accumulation of methane in the sediment by reducing diffusion. The dependence of ebullition on the diffusive flux at the sediment water interface was analyzed by Langenegger et al. (2019), by a modeling approach solving the transport equation. In summary, the authors showed that the accumulation of methane at the bottom water reduces methane diffusion from the sediment and ebullition is increased.

Linear regressions were analyzed between ebullition and temperature, dissolved oxygen, and Chla (Figure 3) for each location. The linear models considering each parameter have a small representation of ebullition variability for both locations separately (small R^2 showed at Figure 3). That could be attributed partially to the fact that ebullition does not respond linearly and rapidly to the parameters considered, but also because it results from a combination of physical-chemical and biological conditions with overlapping effects on bubble release from the sediment. Water temperature at Park location had the best coefficient of determination (0.28), however applying a linear model for the data set combining both locations (Park and Intake) resulted in a R^2 of 0.1, supporting that a linear regression of a single parameter is still a poor ebullition predictor.

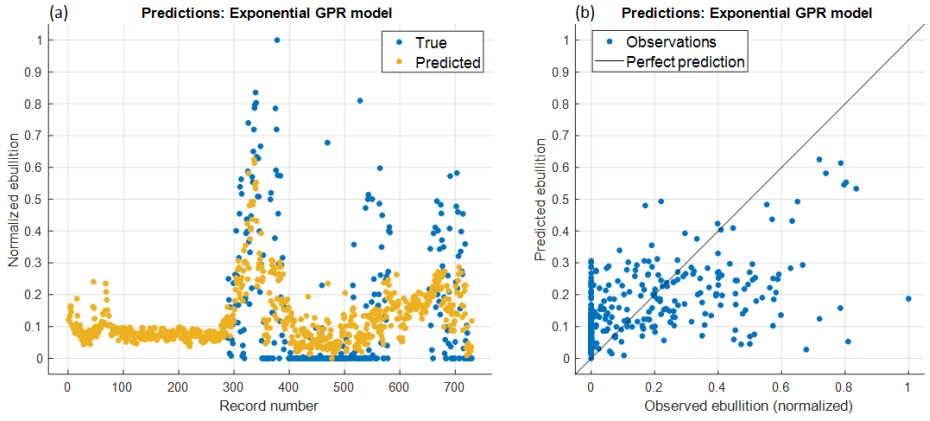
Figure 3 - Linear regressions between daily ebullition and daily averaged **(a)** temperature, **(b)** dissolved oxygen and **(c)** Chla for each location. Fitting equations, R^2 and *p-value* are in the title of each panel.



Taking into account that the water column also controls the processes occurring in the sediment, and thus, affect ebullition, it was investigated whether the combination of the different parameters would be better predictors of ebullition than considering each one separately. For this step the Regression Learner App, within the machine learning toolbox of MatLab R2019a, was applied. One of the advantages of using machine learning methods is that nonlinear relationships among the parameters are also considered.

The data set from both locations were combined and the input parameters were PMP, dissolved oxygen, temperature, Chla and water depth. A principal component analysis was performed as a previous step to only keep the components that would explain 95% of the variance, resulting in three principal components which represented 66.3, 22.2 and 7.7%. The Exponential Gaussian Process Regression model (Exponential GPR), which is a non-parametric Bayesian (based on a probability distribution) supervised model, was selected and the model prediction is shown in Figure 4.

Figure 4 - **(a)** measured and model predicted values of daily ebullition and **(b)** shows the scatter plot of predicted versus observed ebullition with 1:1 line and the model performance. Ebullition time series from Park and Intake were combined and the flux was normalized by the maximum value (566 mL m^{-2} d^{-1}) to be in the range 0–1.

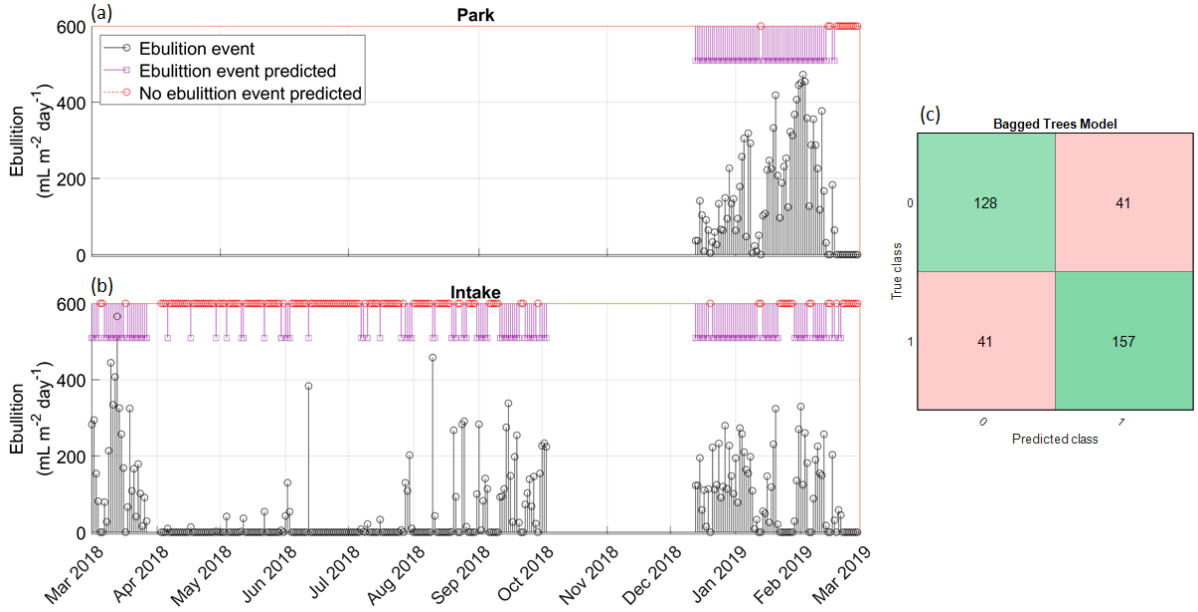


The regression model resulted in a RMSE = 0.17 and R^2 = 0.34, the four parameters combined had a better adjustment than the linear regression of single parameters (R^2 = 0.1 for temperature), however it is still reproducing less than 40% of the ebullition variability. In addition, mainly the extreme fluxes (zero fluxes and highest values) could not be reproduced by the model. As already mentioned, one of the main reasons for the model not representing well the magnitude of ebullition

can be attributed to the fact that ebullition depends on the combination of biochemical conditions for net of gas production and on physical conditions (of sediment and water) controlling gas storage in the sediment and for triggering ebullition.

In this way, similarly as with the Regression Learner App the data set was applied into the Classification Learner App to test if the occurrence of ebullition events could be predicted, where 1 represented an ebullition event independent of its magnitude. The previous principal component analysis was kept and based on the best accuracy among all models the Bagged Trees (in which the output is the average from the results of several individual trees that are constructed by the algorithm) classification, was selected, reproducing the events with an accuracy of 77.7% (Figure 5), with same error for both classes, as shown in the confusion matrix with 41 events with false positive and 41 events of false negative.

Figure 5 - Observed ebullition events and its magnitudes in black stems, purple stems are ebullition events predicted by the classification model, and red stems represent no ebullition events at **(a)** Park and **(b)** Intake. **(c)** is the confusion matrix, where 1 represents ebullition events and 0 no ebullition events, green backgrounds represent the right predictions and red backgrounds the wrong predictions.



CONCLUSIONS

A calibrated 3D hydrodynamic-water quality model provided time-series of water quality parameters for locations in the reservoir where field measurements were not conducted, allowing the investigation of the relationship between water quality parameters and observed gas ebullition. As ebullition is controlled by interacting physical, chemical, and biological effects, a linear fit of ebullition to a single water quality parameter does not explain the flux variability (R^2 in the range $0.01 - 0.28$ for separate locations and $R^2 = 0.1$ for water temperature combining locations). A supervised machine learning regression model was set up, which resulted in better ebullition prediction ($R^2 = 0.34$), however it confirmed that the magnitude of ebullition cannot be well predicted by considering only water quality conditions. On the other hand, the occurrence of ebullition events was predicted with an accuracy of 77% by a Bagged Trees classification model, indicating the potential of combining new approaches of data analysis (as machine learning techniques) with physical-based models to derive insights on the understanding of processes controlling gas ebullition from reservoirs.

This study provided preliminary results on a new approach for gas ebullition analysis. Improvements are still required and for further steps it is suggested to also consider the simulated concentration of dissolved methane at the bottom water layer, bottom currents, and measured atmospheric pressure as ebullition predictors. Additionally, machine learning techniques can be further explored to identify relationships among the different parameters and can also be applied for ebullition parametrization, and further combining regressions and classification methods.

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