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# Surface ocean CO<sub>2</sub> concentration and air-sea flux estimate by machine learning with modelled variable trends

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The global ocean is a major sink of anthropogenic carbon dioxide (CO<sub>2</sub>) emitted into the atmosphere. Machine learning has been actively used in the past decades to estimate the oceanic sink, but it is still a challenge to obtain an accurate estimate due to scarcely available CO<sub>2</sub> measurements. One of the methods to deal with data scarcity was normalizing multiple years' CO<sub>2</sub> values to a reference year to increase the spatial coverage. The practice assumed a constant CO<sub>2</sub> trend for the normalization. Here, we used three machine learning models to extract variable ocean CO<sub>2</sub> trends on a decadal scale and proposed a method to use the extracted ocean CO<sub>2</sub> trends to correct the decadal atmospheric CO<sub>2</sub> trends for data normalization. The method minimizes assumptions of using the extracted ocean CO<sub>2</sub> trends directly. Comparisons of our CO<sub>2</sub> flux estimate with machine learning products included in Global Carbon Budget 2021 indicates that using the variable trends improved the bias resulted from using a constant trend and that the trends are a critical factor for machine learning methods. Our dataset includes monthly distributions of surface ocean CO<sub>2</sub> concentration and air-sea flux in 1980–2020 with a spatial resolution of 1×1 degree.

## KEYWORDS

global ocean CO<sub>2</sub> flux, machine learning, random forest, gradient boost machine, neural network

## Introduction

The oceans play a crucial role in mitigating the increase of atmospheric CO<sub>2</sub> emitted into the atmosphere by human activities (Sabine, 2004; Khatiwala et al., 2013; McKinley et al., 2016). Using machine learning to estimate the oceanic sink has been practiced in the past decades and the results have become an important part of the Global Carbon Budget (Friedlingstein et al., 2022). Nevertheless, it is still a challenge to obtain an

accurate estimate due to scarcely available CO<sub>2</sub> measurements. Through internationally coordinated efforts, decades of *in situ* measurements have been combined to form high-quality databases, such as the Surface Ocean CO<sub>2</sub> Atlas Database (SOCAT) (Sabine et al., 2013; Pfeil et al., 2013; Bakker et al., 2016). The composite sampling map of SOCAT appears to cover most areas of the oceans. However, only a small portion of the oceans had samples in any single year and the samples were unevenly distributed in time and space. The dilemma of using multiple years' data to train a machine learning model is that while ocean CO<sub>2</sub> tends to track the increase of atmospheric CO<sub>2</sub> closely (Fay and McKinley, 2013; Bates et al., 2014), the large seasonal and spatial variabilities up to a few hundred  $\mu\text{atm}$  make it difficult to detect the trends in the order of a few  $\mu\text{atm}$  per year. Current methods for solving the problem include normalizing ocean CO<sub>2</sub> to a reference year (Takahashi et al., 2009; Sasse et al., 2013a; Sasse et al., 2013b; Nakaoka et al., 2013; Zeng et al., 2014), including a linear time-dependent term in regression (Fay and McKinley, 2013; Iida et al., 2015; Jones et al., 2015; Watson et al., 2020; Iida et al., 2021), and including atmospheric CO<sub>2</sub> as a predictor to make models learn the trend implicitly (Landschützer et al., 2016; Denvil-Sommer et al., 2019; Gregor and Gruber, 2021; Chau et al., 2022). The former two methods assume a constant trend for the whole period. This can be a good approximation when the time span is short, but the error tends to become substantial in a long period as the trend could vary greatly with time. Landschützer et al. (2016) and Gloege et al. (2021) showed that such a problem could also exist in the third method.

There are two camps of using machine learning to reconstruct ocean CO<sub>2</sub> in terms of data pooling strategy. One camp treats the global oceans as one entity (Takahashi et al., 2009; Sasse et al., 2013b; Nakaoka et al., 2013; Zeng et al., 2014; Denvil-Sommer et al., 2019; Chau et al., 2022). The other camp divides the oceans into clusters with similar biogeochemical properties. Sasse et al. (2013a) and Landschützer et al. (2013) are early pioneers in this camp. They used a Self-Organization Map (SOM) for clustering in the first step and then used different regression methods in the second step for making predictions. This method, used by Landschützer et al. (2013) and named two-step method, was also applied by Laruelle et al. (2017); Watson et al. (2020), and Gloege et al. (2021). Other clustering methods include geographical blocking (Iida et al., 2015; Watson et al., 2020), K-mean clustering (Gregor et al., 2019; Gregor and Gruber, 2021), and CO<sub>2</sub> biome clustering (McKinley et al., 2011; Fay and McKinley, 2013; Gregor et al., 2019; Watson et al., 2020).

In this study, we used three machine learning models to extract the global time-dependent ocean CO<sub>2</sub> trends. They were used to correct the decadal atmospheric CO<sub>2</sub> trends to normalize ocean CO<sub>2</sub> measurements to a reference year for modelling the

nonlinear dependence of CO<sub>2</sub> on biogeochemical predictors. Then, we reconstruct monthly CO<sub>2</sub> distributions between 1980 and 2020 with a spatial resolution of 1×1 degree. Our method is in the first camp discussed above. We compared the air-sea flux estimate with those included in the Global Carbon Budget 2021 (Friedlingstein et al., 2022). The results reveal that the ocean CO<sub>2</sub> trends are a critical factor for machine learning methods, which in turn implies the importance of having long-term observations to quantify the uptake, predict scenarios, and evolve adapting strategies.

## Method

### Model setup

Following Zeng et al. (2014) and considering the inconstancy that can be associated with long-term oceanic CO<sub>2</sub> trends, we express the nonlinear dependence of ocean CO<sub>2</sub> on time and biogeochemical variables as:

$$\text{CO}_2W = f(\text{SST}, \text{dSST}, \text{SSS}, \text{CHL}, \text{MLD}, \text{LAT}, \text{LON}) + f(\text{year}), \quad (1)$$

where SST stands for sea surface temperature, SSS for sea surface salinity, CHL for chlorophyll-a concentration, MLD for mixed layer depth, LAT for latitude, and LON for longitude. The sine and cosine converted values of LON were used to make the circular variable contingent. We replaced the month variable of Zeng et al. (2014) with the SST anomaly (dSST) against the annual mean to harmonize the seasons of the two hemispheres. The function of year represents the trends, which were a constant in Zeng et al. (2014).

We used machine learning to investigate the variable trends with varying lengths of data (Figure 1). A similar iteration method was also used by Zeng et al. (2014). For a given target year and data length, we fitted the dependence of CO<sub>2</sub>W on year by linear regression first. The first term in Eq.(1) was treated as an error in this step. Then we subtracted the trend from observations and used machine learning to model the nonlinear relationship between the residual and predictors. These two steps were repeated until the trend became stabilized. Initially, three years' data were used: the target year plus and minus one year. The data length was increased to the longest available data length gradually. The longest data length was 41 years for the target year 2000, *i.e.*, all data between 1980 and 2020 were included. The extracted trends were used as reference to model the decadal trends of atmospheric CO<sub>2</sub> by fitting its annual increase rates with the following harmonic function:

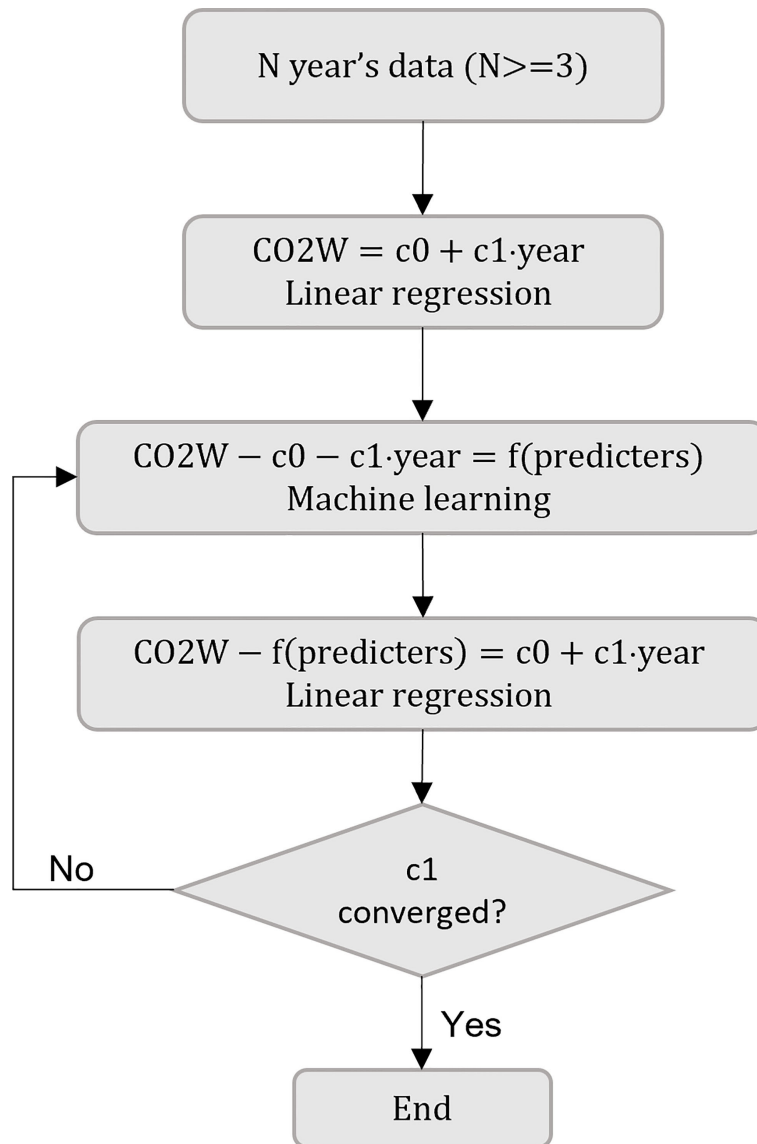


FIGURE 1  
Flow chart of the iteration method for trend extraction.

$$\begin{aligned}
 trend = & c_0 + c_1 year + c_2 \cos\left(\frac{2\pi year}{T_1}\right) \\
 & + c_3 \sin\left(\frac{2\pi year}{T_1}\right) + c_4 \cos\left(\frac{2\pi year}{T_2}\right) \\
 & + c_5 \sin\left(\frac{2\pi year}{T_2}\right). \quad (2)
 \end{aligned}$$

where  $T_1$  and  $T_2$  are time parameter in year. For training machine learning models, the atmospheric  $CO_2$  trends obtained by Eq.(2) were used to normalize the observed  $CO_2$  values to the reference year 2000 by the equation:

$$CO2W^{norm} = CO2W^{raw}(year) \pm \sum_{i=2000}^{year} trend(i), \quad (3)$$

where  $\pm$  is positive when  $i < 2000$  and negative when  $i > 2000$ . At  $i = 2000$ , the trend correction is zero. Global  $CO_2$  concentrations were constructed by adding or subtracting the trend correction of Eq.(3) to the predicted  $CO_2$  values. The process is the inverse of the normalization. Using atmospheric  $CO_2$  trends for data normalization avoided problems in using oceanic  $CO_2$  trends directly, e.g., insufficient data points in the early and later years and the difficulty of determining the best data length for trend extraction.

## Models

We deployed three machine learning models: Random Forest (RF), Gradient Boost Machine (GBM), and Feedforward Neural Network (FNN). Using multiple models has the merit of mutual overfit checking and compensating model weakness with each other.

RF was proved to be a robust method for modelling carbon flux at the global scale (Zeng et al., 2020) and was applied to global ocean CO<sub>2</sub> mapping recently (Gregor et al., 2019). RF partitions a training dataset into subsets repeatedly by random sampling and uses the subsets to construct trees. We used the python library of Ranger (Wright and Ziegler, 2017) which implements the regression algorithm using a two-stage randomization procedure to partition trees. Given a subset, the root node in a tree is recursively split into binary nodes until the number of data points in the leaf nodes becomes no larger than a specified number. In each split, the RF randomly selects a subset of predictor variables and searches them for splitting points that minimize node impurity (Ishwaran, 2015). In making a prediction, a set of predictors are passed through branches of nodes according to the splitting rule until the journey ends up in a leaf node. The mean of the target variable in the leaf node is taken as an estimate. Then the mean estimate of all leaf nodes is used as the prediction. Sensitive configuration factors for the RF include the number of trees and the number of data points in the leaf nodes (Zeng et al., 2020). The default setting includes 500 trees and 5 data points. We raised the data points to 100 based on our experiments with the ocean CO<sub>2</sub> data discussed in the data section to prevent hot spots in predicted CO<sub>2</sub> in the southern oceans where vast empty areas exist in certain months. The configuration yielded good validation results.

A decision-tree-based GBM emerged in the ocean CO<sub>2</sub> mapping recently (Gregor et al., 2019; Gregor and Gruber, 2021). Like a RF, a GBM combines weak learners into a single strong learner (Natekin and Knoll, 2013), but in an iterative fashion. It adds trees one at a time, and existing trees are not changed. We used the python library of LightGBM (Ke et al., 2017). Instead of the level-wise strategy of the RF, the GBM grows a tree leaf-wise by splitting nodes that produce the highest loss change until the number of leaf nodes becomes no larger than a specified number. The observed values of the target variable are assigned to the leaf nodes of the first tree. Then, the residuals of the previous predictions minus observations are assigned to the leaf nodes of a subsequent tree. A gradient descent procedure is used to obtain parameters that improve the accuracy of predictions. By experimenting with our ocean CO<sub>2</sub> data and using the RF as a reference, we found that LightGBM performed well with 500 trees and a maximum number of 100 terminal nodes in a tree.

FNN has been used for ocean CO<sub>2</sub> mapping since the early 2010s (e.g., Landschützer et al., 2013; Zeng et al., 2014). FNN has

a layered structure, including an input layer, one or more hidden layers, and an output layer. Neurons between adjacent layers are fully connected. A neuron in the hidden layer uses an activation function to transform the weighted sum of inputs to form an input for the neuron in the output layer, which in turn transforms the weighted sum of inputs to form a prediction. Details of the FNN method can be found in Svozil et al. (1997) and abundant other references. We used python's MLPRegressor model with one hidden layer and 64 hidden neurons, which is the same as that used by Zeng et al. (2014). Their investigations show that the setting yielded uncertainties in the level of grid mean variation of measurements. We raised the default maximum training iterations of MLPRegressor from 200 to 500. Our tests indicate that when the iterations were larger than 300, doubling or tripling the number did not make a substantial change in the flux estimate. Training the FNN took a much longer time than training the RF and GBM. As the trend extraction and validation discussed later involve many rounds of training, we had to set a fixed number of training iterations so that the training could be completed within a reasonable amount of time. The settings also yielded results well harmonized with those of the RF and GBM.

We validated the model performance by a leave-one-year-out (LOYO) method. Given N years of data, N validations were done by setting aside one year's data for validation and using the remaining N-1 years of data for training. A model's performance was evaluated by the mean bias. The validation method has an advantage over the conventional n-fold method in that the validation data of LOYO are more likely to come from unsampled domains of the training data. Another advantage is that LOYO can also be used to detect trends. If the target variable has an increasing trend, a model trained with data in early years tends to make predictions smaller than the observations in later years and vice versa.

## Data

We extracted monthly CO<sub>2</sub> fugacity (fCO<sub>2</sub>) in 1×1-degree grids from the track-gridded database of SOCAT version 2021 (Sabine et al., 2013; Pfeil et al., 2013; Bakker et al., 2016). We relaxed the criteria of Zeng et al. (2014) to include data when fCO<sub>2</sub> values are between 50 μatm and 1000 μatm and salinity is larger than 15 g kg<sup>-1</sup>. A total of 273,456 data points were extracted for 1980-2020. We confined the fCO<sub>2</sub> training data set to post-1980 due to large uncertainties in the early measuring techniques (Sasse et al., 2013). The sources of predictor variables are shown in Table 1. The monthly climatology of MODIS-AQUA and MODIS-TERRA of 2002-2019 in 0.083×0.083-degree grids (Hu et al., 2012.) were combined and re-binned into 1×1-degree grids. The values of CHL and MDL were scaled by log(1+CHL) and log(1+MDL) to reduce the skewness of

TABLE 1 Data sources.

Variable	Units	Resolutions	Source URL	DOI or Version	Reference
CO2W	$\mu\text{atm}$	Monthly, 1 $\times$ 1 degree.	<a href="https://www.socat.info/">https://www.socat.info/</a>	Version-2021	(Sabine et al., 2013; Pfeil et al., 2013; Bakker et al., 2016.)
xCO2A	ppm	Monthly, 0.05 sine latitude.	<a href="https://www.esrl.noaa.gov/gmd/ccgg/mlb/">https://www.esrl.noaa.gov/gmd/ccgg/mlb/</a>		(Conway et al., 1994 and Dlugokencky et al., 2019)
CHL	$\text{mg m}^{-3}$	Monthly climatology, 0.083 $\times$ 0.083 degrees.	<a href="https://oceancolor.gsfc.nasa.gov/cgi/l3">https://oceancolor.gsfc.nasa.gov/cgi/l3</a>		(Hu et al., 2012.)
SST	$^{\circ}\text{C}$	Monthly, 1 $\times$ 1 degree.	<a href="https://psl.noaa.gov/data/gridded/data.cobe.html">https://psl.noaa.gov/data/gridded/data.cobe.html</a> <a href="http://ds.data.jma.go.jp/tcc/library/MRCS_SV12/explanation/cobe_sst_e.htm">http://ds.data.jma.go.jp/tcc/library/MRCS_SV12/explanation/cobe_sst_e.htm</a>		(Ishii et al., 2005.)
SSS	$\text{g kg}^{-1}$	Monthly climatology, 1 $\times$ 1 degree.	<a href="https://www.nodc.noaa.gov/OC5/woa18/woa18data.html">https://www.nodc.noaa.gov/OC5/woa18/woa18data.html</a>		(Zweng et al., 2019)
MLD	m	Monthly climatology, 1 $\times$ 1 degree.	<a href="https://www.nodc.noaa.gov/OC5/woa18/woa18data.html">https://www.nodc.noaa.gov/OC5/woa18/woa18data.html</a>		
WIND	$\text{m s}^{-1}$	Monthly, 0.25 $\times$ 0.25 degrees.	<a href="https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels-monthly-means?tab=form">https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels-monthly-means?tab=form</a>	10.24381/ CDS.F17050D7	(Hersbach et al., 2020)
Ps	Pa	Monthly, 0.25 $\times$ 0.25 degrees.	Same as ERA5 WIND		

CO2W, ocean CO<sub>2</sub> fugacity ( $\mu\text{atm}$ ); xCO2A, mole fraction ratio of atmospheric CO<sub>2</sub> (ppm); CHL, chlorophyll-a concentration ( $\text{mg m}^{-3}$ ); SST, sea surface temperature ( $^{\circ}\text{C}$ ); SSS, sea surface salinity ( $\text{g kg}^{-1}$ ); MLD, mixed layer depth (m); WIND, wind speed ( $\text{m s}^{-1}$ ); Ps, surface pressure (Pa).

sample distribution. Filling missing CHL data in high latitudes with a small constant is a common practice (Gregor and Gruber, 2021; Chau et al., 2022). We filled the missing CHL in a grid with the smallest observed value in that grid. The data of SST (Ishii et al., 2005) and SSS (Zweng et al., 2019) were used without pre-processing. For flux calculation, we used the wind speed (WIND) and surface pressure (Ps) of the fifth generation ECMWF atmospheric reanalysis of the global climate (ERA5) (Hersbach et al., 2020), and the mole fraction of air CO<sub>2</sub> (xCO2A) of NOAA's Marine Boundary Layer Reference (Conway et al., 1994; Dlugokencky et al., 2021). The monthly WIND and Ps in 0.25 $\times$ 0.25 degrees were averaged to 1 $\times$ 1 degree. The surface xCO2A in sine latitude grids was interpolated to 1 $\times$ 1 degree.

## Comparison

We compared our estimates with seven machine learning products included in GCB-2021 (Table 2). We recalculated their fluxes by the same procedure to eliminate the effect of using different flux dependent data and coefficients. As each product has a different spatial coverage, we adjusted their annual fluxes using the equation

$$F_{adjusted} = F_{model} + (F_{ML3} - F'_{ML3}), \quad (4)$$

where  $F_{ML3}$  ( $\text{PgC a}^{-1}$ ) is the mean annual flux of NIES-ML3 (the ensemble mean of the three models) in the available period of a pair of products under comparison and  $F'_{ML3}$  ( $\text{PgC a}^{-1}$ ) is the mean annual flux of NIES-ML3 in the grids where both products have data. The adjustment was intended to bring fluxes

with different spatial coverages to the same coverage as the NIES-ML3.

## Results and discussion

### CO<sub>2</sub> trend

The ocean CO<sub>2</sub> trends obtained using the iteration method in Figure 1 with the longest available data are shown in Figure 2A along with the annual increase rates of global atmospheric CO<sub>2</sub> concentrations (ppm) (Friedlingstein et al., 2022). Because of data scarcity, the extracted trends fluctuate dramatically when the data length is short and converge gradually (Figure 2B). Sutton et al. (2019) pointed out that the number of years of observations needed (YON) to detect a statistically significant trend over variability ranges from 8 to 15 years at several open ocean sites. It is reasonable to assume the same YON range for open oceans. Ideally, the trend for a year should be extracted with the shortest data length possible. As it is difficult to determine the smallest stabilization length and the trend does not change much after 10 to 15 years, we presented the trend with the maximum data length. The extracted trends appear to track the decadal trends of the atmospheric CO<sub>2</sub> in 1990–2015 obtained by Eq.(2) with  $T_1 = 20$  year and  $T_2 = 40$  year.

We applied the LOYO method to the data normalized by the trends of Eq.(2). A small trend of  $0.1565 \mu\text{atm a}^{-1}$  exists in the residual of model prediction minus observations (Figure 2C). The p-value of the trend is  $9 \times 10^{-6}$ , indicating that the trend is significant. We subtracted the residual trend from the decadal air CO<sub>2</sub> trends and yielded the trends shown in magenta line in

TABLE 2 Datasets for comparison.

Dataset	Period	Reference	Regression	Trend	Clustering	Predictors
NIES-ML3	1980-2020	This study	RL, GBM, and FNN	Year-dependent rates	1 global cluster. CO <sub>2</sub> normalized to 2000 using variable annual rates	SST, dSST, SSS, CHL, MLD, LAT, LON
NIES-NN	1980-2020	Zeng et al. (2014). (doi: 10.17595/20210806.001)	FNN	Linear trend	1 global cluster. CO <sub>2</sub> normalized to 2000 using the rate of 1.54 $\mu\text{atm a}^{-1}$	SST, SSS, CHL, MLD, dSST
JMA-MLR	1990-2020	Iida et al. (2021) <a href="https://www.data.jma.go.jp/gmd/kaiyou/english/co2_flux/co2_flux_data_en.html">https://www.data.jma.go.jp/gmd/kaiyou/english/co2_flux/co2_flux_data_en.html</a>	MLR	Cluster-dependent linear trends	Manually defined clusters by regions	SST, SSS, SSH, CHL, MLD, TIME
MPI-SOMFFN	1982-2019	Landschützer et al. (2016). ( <a href="https://www.ncei.noaa.gov/data/oceans/nci/ocads/data/0160558/MPI_SOM-FFN_v2021/">https://www.ncei.noaa.gov/data/oceans/nci/ocads/data/0160558/MPI_SOM-FFN_v2021/</a> )	FNN	Implicit rates learned from air CO <sub>2</sub>	16 clusters by self-organization map	SST, SSS, MLD, CHL, CO <sub>2</sub> A
CMEMS-FFNN	1985-2019	Chau et al. (2022). ( <a href="https://resources.marine.copernicus.eu/?option=com_csw&amp;view=details&amp;product_id=MULTIOBS_GLO_BIO_CARBO_SURFACE_REP_015_008">https://resources.marine.copernicus.eu/?option=com_csw&amp;view=details&amp;product_id=MULTIOBS_GLO_BIO_CARBO_SURFACE_REP_015_008</a> )	FNN	Implicit rates learned from air CO <sub>2</sub>	Clustering by month with a window size of three months	SST, SSS, SSH, MLD, CHL, CO <sub>2</sub> A, CO <sub>2</sub> C, LAT, LON
CSIR-ML6	1982-2016	Gregor et al. (2019) ( <a href="https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0206205">https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0206205</a> )	GBM, FNN, SVR, and RF	Implicit rates learned from air CO <sub>2</sub> and time	Repetition of K-mean clustering and CO <sub>2</sub> biomes clustering	SST, dSST, SSS, MLD, CHL, dCHL, WIND, CO <sub>2</sub> A, TIME
OceanSODA-ETHZ	1982-2020	Gregor and Gruber (2021). ( <a href="https://doi.org/10.25921/m5wx-ja34">https://doi.org/10.25921/m5wx-ja34</a> )	GBM and FNN	Implicit rates learned from air CO <sub>2</sub>	Repetition of K-mean clustering	SST, SSS, CHL, MLD, WIND, CO <sub>2</sub> A
JENA	1957-2020	Rödenbeck et al. (2013). <a href="http://www.bgc-jena.mpg.de/CarboScope/Version_oc_021">http://www.bgc-jena.mpg.de/CarboScope/Version oc_021</a>	Inversion Model	Ocean inversion coupled with atmospheric model CO <sub>2</sub>	Fixed grids	Internal ocean chemistry

Machine learning models include random forest (RF), gradient boost machine (GBM), feedforward neural network (FNN), support vector machine for regression (SVR), and multiple linear regression (MLR). Predictors include atmospheric CO<sub>2</sub> (CO<sub>2</sub>A), ocean CO<sub>2</sub> climatology (CO<sub>2</sub>C), sea surface temperature (SST), sea surface temperature anomaly (dSST), sea surface salinity (SSS), sea surface height (SSH), chlorophyll-a (CHL), chlorophyll-a anomaly (dCHL), mixed layer depth (MLD), wind speed (WIND), time (TIME), latitude (LAT), and longitude (LON).

**Figure 2A.** The numerical values of the corrected trends are listed in **Table 3**. They were used for final data normalization. The corrected trends agree well with the extracted ocean CO<sub>2</sub> trends in 1996-2013, during which the data length used to extract ocean CO<sub>2</sub> trends is longer than the YON of **Sutton et al. (2019)**. The corrected trends in early 2000s are close to those obtained by **Sutton et al. (2019)** for the time series station WHOTS in the subtropical North Pacific and Stratus in the South Pacific gyre in 2004-2013. Although the corrected trends before 1997 are smaller than those used by **Takahashi et al. (2009)** and **Zeng et al.,(2014)** for data normalization, they are within the range of trends summarized by **Takahashi et al. (2009)**.

## Validation and uncertainty

The performances of the three models were evaluated by the LOYO method with the normalized CO<sub>2</sub>. The validation yields small biases (prediction minus observation) and good

correlation coefficients. The annual mean bias ranges between -4.82 and 3.79  $\mu\text{atm}$  for RF, between -4.34 and 3.92  $\mu\text{atm}$  for GBM, and between -5.29 and 4.95  $\mu\text{atm}$  for FNN (**Table 3**). Their mean biases are -0.36  $\mu\text{atm}$ , -0.24  $\mu\text{atm}$ , and -0.27  $\mu\text{atm}$ , respectively. The correlation coefficient  $R^2$  in individual years ranges between 0.50 and 0.90 for RF, between 0.49 and 0.88 for GBM, and between 0.43 and 0.89 for FNN. Their mean  $R^2$  are 0.70, 0.69, and 0.62, respectively. **Figure 3** shows the goodness of fit of the three models. The bias and  $R^2$  in the figure were calculated directly using all validation data points and therefore are equivalent to the weighted mean bias and  $R^2$  in **Table 3**.

At the CO<sub>2</sub> level in year 2000, one unit CO<sub>2</sub> change results in a flux change of 0.19 PgC a<sup>-1</sup>. We calculated the flux uncertainties approximately by multiplying this value with the biases in **Table 3**. For the RF model, the uncertainty ranges from -0.93 PgC a<sup>-1</sup> to 0.72 PgC a<sup>-1</sup> and the mean is -0.07 PgC a<sup>-1</sup>. The GBM model has a smaller uncertainty range from -0.83 PgC a<sup>-1</sup> to 0.74 PgC a<sup>-1</sup> and a mean of -0.05 PgC a<sup>-1</sup>. The FNN model has the largest uncertainty range from -1.01 PgC a<sup>-1</sup> to 0.94 PgC a<sup>-1</sup> and a mean of -0.05 PgC a<sup>-1</sup>.

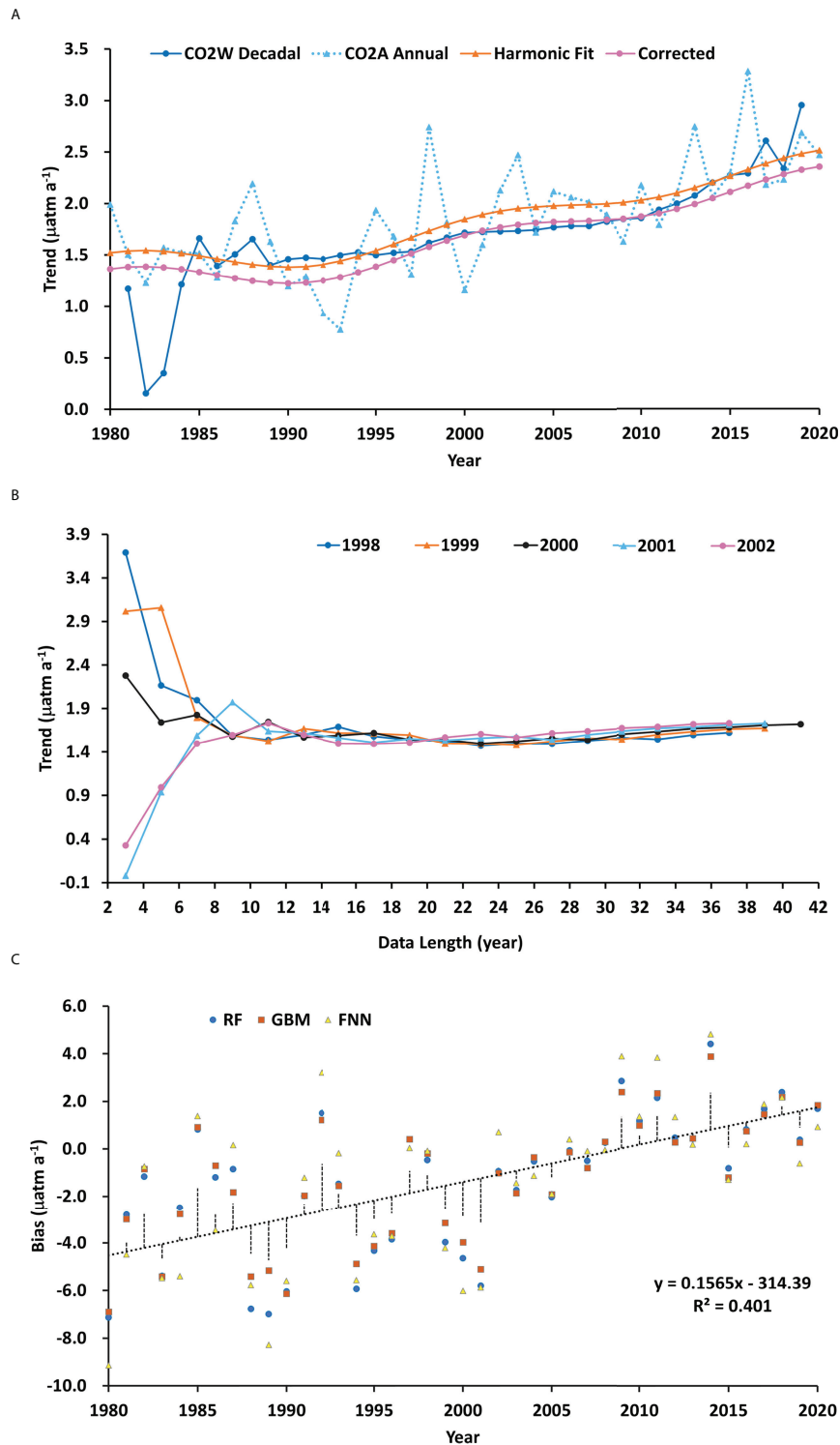


FIGURE 2

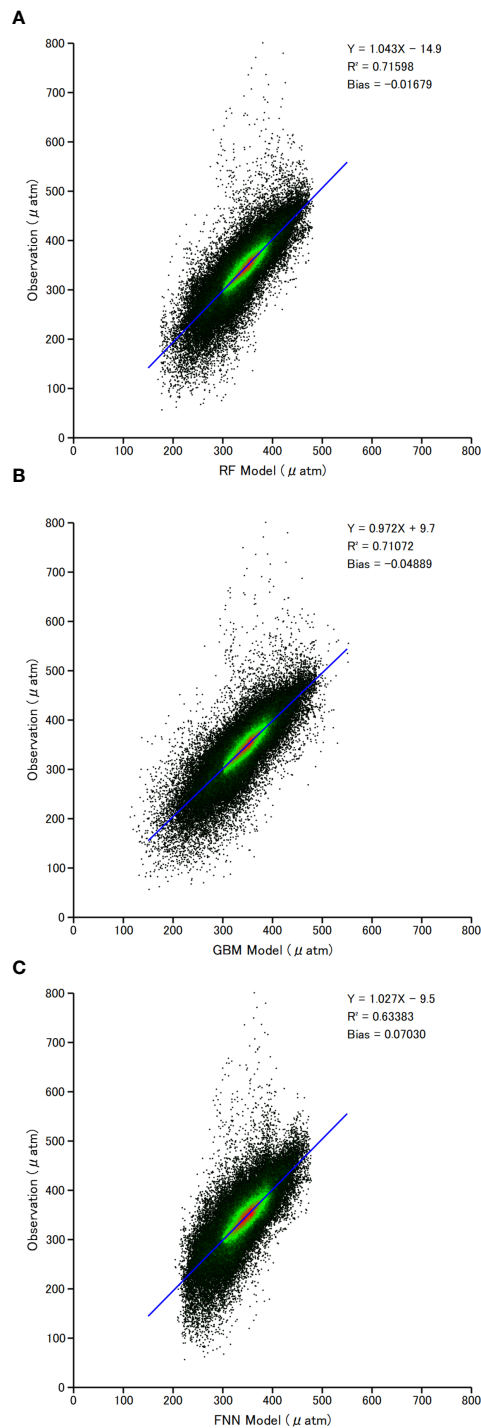
Trends extraction. (A) The trend of ocean CO<sub>2</sub> for a target year (blue) was estimated by using the iteration method with the longest data length around the year. The final trends to be used for data normalization (magenta) are the corrected function fitting trends (orange) of the annual increase rate of air CO<sub>2</sub> (cyan). (B) Examples of trend variations with data length for the target years 1998–2002. (C) The trend of CO<sub>2</sub> biases (prediction – observation) detected by LOYO with CO<sub>2</sub> data normalized by the uncorrected decadal trends of air CO<sub>2</sub>. The vertical lines show the standard residuals of the regression.

TABLE 3 Trends for data normalization and LOYO validation results.

Year	Rate ( $\mu\text{atm a}^{-1}$ )	RF			GBM			FNN			ND
		Bias ( $\mu\text{atm}$ )	STD ( $\mu\text{atm}$ )	R <sup>2</sup>	Bias ( $\mu\text{atm}$ )	STD ( $\mu\text{atm}$ )	R <sup>2</sup>	Bias ( $\mu\text{atm}$ )	STD ( $\mu\text{atm}$ )	R <sup>2</sup>	
1980	1.36	-3.31	11.39	0.90	-3.09	12.42	0.88	-5.29	13.28	0.85	339
1981	1.38	1.36	11.68	0.88	1.45	12.58	0.86	0.56	11.14	0.89	610
1982	1.39	2.32	15.58	0.55	3.43	16.26	0.49	3.63	16.43	0.48	441
1983	1.38	-2.20	16.13	0.58	-1.97	15.81	0.60	-1.20	15.47	0.62	273
1984	1.36	0.87	18.26	0.63	0.66	18.33	0.63	-1.05	20.29	0.54	504
1985	1.33	3.79	15.11	0.58	3.92	15.90	0.54	3.97	15.28	0.57	708
1986	1.30	1.67	16.80	0.70	2.29	18.79	0.62	0.67	17.76	0.66	897
1987	1.27	1.93	15.95	0.50	0.98	16.29	0.49	3.25	16.83	0.43	1587
1988	1.25	-4.08	13.27	0.82	-3.06	13.62	0.82	-2.52	15.83	0.76	1010
1989	1.23	-4.25	27.73	0.54	-2.58	28.72	0.51	-5.19	29.89	0.46	1117
1990	1.22	-3.82	12.04	0.84	-3.78	12.09	0.83	-3.20	14.31	0.78	889
1991	1.23	0.14	12.63	0.81	0.09	12.81	0.81	1.26	14.94	0.73	2011
1992	1.25	3.37	15.11	0.63	3.04	15.82	0.60	4.95	16.23	0.55	2521
1993	1.28	0.46	15.72	0.74	0.28	16.33	0.72	3.16	17.80	0.66	3398
1994	1.33	-4.09	20.75	0.73	-3.27	21.36	0.71	-3.73	22.84	0.67	3981
1995	1.39	-2.70	19.28	0.65	-2.59	18.94	0.66	-1.92	24.27	0.45	6157
1996	1.45	-2.33	20.53	0.73	-2.12	20.82	0.72	-2.52	22.46	0.67	6092
1997	1.51	1.69	22.84	0.74	1.85	23.53	0.72	0.32	26.40	0.65	4336
1998	1.58	0.86	21.87	0.56	1.09	23.01	0.52	1.18	23.96	0.47	5861
1999	1.64	-2.89	21.76	0.69	-2.09	22.78	0.66	-3.51	24.64	0.60	4081
2000	1.69	-3.79	26.38	0.60	-3.39	27.21	0.57	-5.03	29.83	0.48	4656
2001	1.74	-4.92	23.15	0.70	-4.35	22.78	0.72	-5.21	26.26	0.62	4855
2002	1.77	-0.08	18.50	0.67	-0.09	18.38	0.68	1.42	21.33	0.56	6760
2003	1.79	-1.00	18.86	0.66	-1.22	19.63	0.63	-0.73	21.20	0.57	7001
2004	1.81	0.12	16.96	0.74	0.30	17.23	0.74	-0.35	19.16	0.67	8077
2005	1.82	-1.47	16.79	0.76	-1.27	17.12	0.75	-1.74	19.10	0.69	9575
2006	1.83	0.33	18.75	0.77	0.42	18.92	0.76	0.90	21.93	0.68	12192
2007	1.83	-0.18	20.30	0.69	-0.50	20.24	0.69	0.27	23.60	0.58	12326
2008	1.84	0.36	20.72	0.73	0.51	20.85	0.73	-0.13	23.37	0.66	11526
2009	1.86	2.76	20.59	0.69	2.30	20.74	0.69	3.47	23.08	0.61	11573
2010	1.88	0.99	18.46	0.72	0.74	18.72	0.71	1.15	21.39	0.63	12822
2011	1.91	1.83	23.00	0.67	2.03	23.17	0.67	3.12	25.35	0.60	13354
2012	1.95	-0.05	23.01	0.68	-0.34	23.21	0.67	0.87	25.01	0.62	13394
2013	2.00	-0.22	21.49	0.70	-0.27	21.92	0.68	-1.06	23.61	0.63	11366
2014	2.05	3.67	24.05	0.68	3.11	23.31	0.70	3.97	26.69	0.60	13447
2015	2.11	-1.73	24.34	0.69	-2.02	24.56	0.68	-2.37	27.29	0.60	13161
2016	2.18	-0.27	17.93	0.77	-0.30	18.06	0.77	-0.63	20.48	0.70	14915
2017	2.23	0.44	19.70	0.77	0.24	19.86	0.77	0.54	22.94	0.69	14694
2018	2.29	0.92	20.11	0.70	0.82	20.23	0.70	0.93	22.95	0.61	12098
2019	2.33	-1.19	20.69	0.73	-1.27	20.46	0.73	-1.97	24.65	0.61	11417
2020	2.36	0.00	23.10	0.72	0.17	23.09	0.72	-1.32	26.82	0.62	7434
Mean	1.67	-0.36	19.06	0.70	-0.24	19.41	0.69	-0.27	21.37	0.62	

Bias, prediction minus observation; STD, standard deviation of the biases; R<sup>2</sup>, correlation coefficient between predictions and observations; ND, number of data points.



**FIGURE 3**

Model predictions vs observations of ocean CO<sub>2</sub> fugacity using normalized data with the trends in Table 3. Predictions come from 41 validations for each target year between 1980 and 2020. The colour indicates the density of data points. (A) Results of the RF model. (B) Results of the GBM model. (C) Results of the FNN model.

## Comparison

The fluxes of the products in Table 2 were recalculated by equations in the Appendix and adjusted by Eq.(4) for comparison. The offset added to the products is 0.00  $\mu\text{atm}$  for NIES-NN and JENA, 0.22  $\mu\text{atm}$  for JMA-MLR, 0.01  $\mu\text{atm}$  for MPI-SOMFFN, 0.34  $\mu\text{atm}$  for CMEMS-FFNN, 0.07  $\mu\text{atm}$  for CSIR-ML6, and 0.01  $\mu\text{atm}$  for OceanSODA-ETHZ.

The difference between NIES-ML3 and NIES-NN is small in 1991-2006 but much larger in the early and late years (Figure 4A). NIES-NN used a constant trend of 1.54  $\mu\text{atm a}^{-1}$

to normalize data to the reference year 2000. The trend is larger than those in Table 3 before 1998 and smaller after that year. This resulted in larger reconstructed  $\text{CO}_2$  in the periods. The bias caused larger flux estimates in the years further away from the reference year. In 1985 and 2019, NIES-NN flux is larger than that of NIES-ML3 by 0.536  $\text{PgC a}^{-1}$  and 1.057  $\text{PgC a}^{-1}$  respectively. The latter is close to half of the fluxes in recent years. Near the reference year of 2000, NIES-NN is smaller than NIES-ML3 in the order of 0.1  $\text{PgC a}^{-1}$ . The JMA-MLR product (Iida et al., 2021) also shows an arch-shaped flux trend like NIES-NN does. Again, the differences are larger in the early and

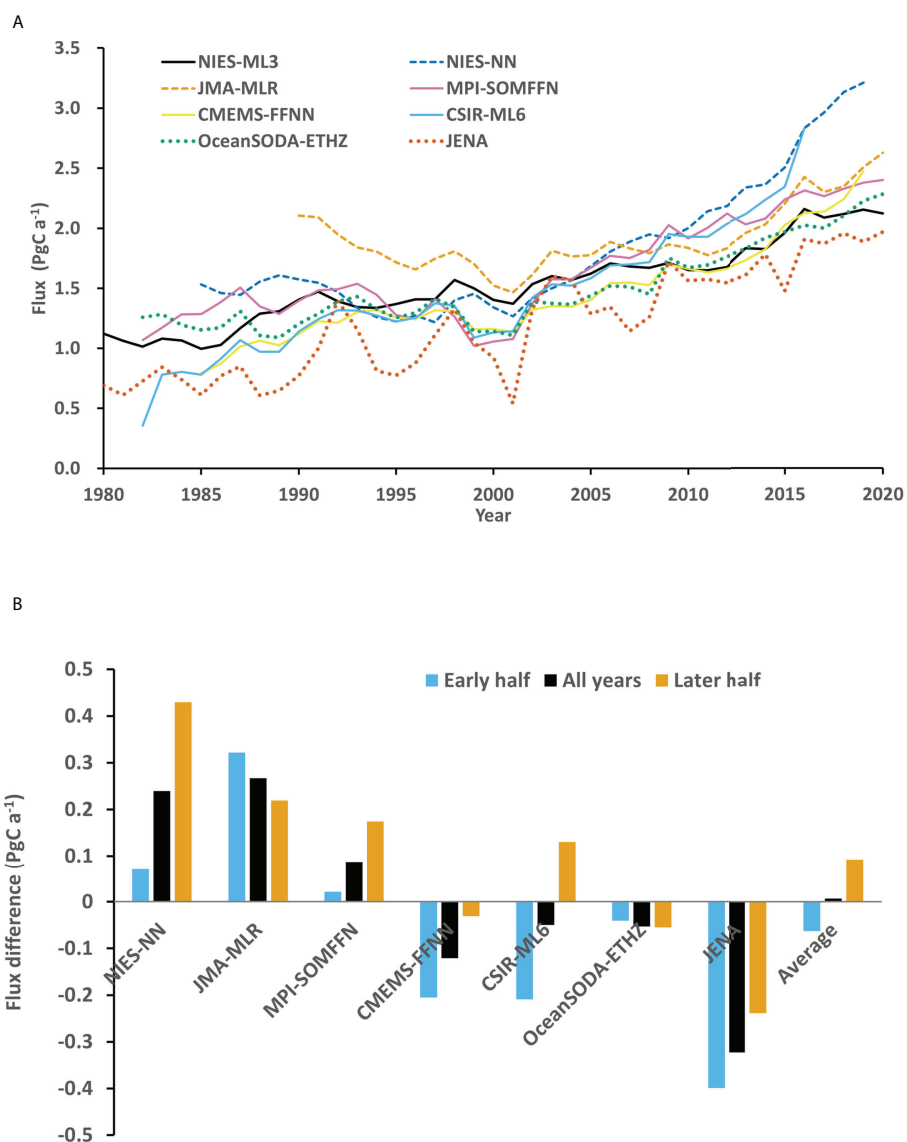


FIGURE 4 Comparisons with machine learning products included in GSB-2021. Fluxes were recalculated by the same method and adjusted to have the same spatial coverage of NIES-ML3. (A) Variations of annual fluxes with time. (B) Mean differences of third-party products minus NIES-ML3 in the whole available period, the early half years, and later half years.

late years of the comparison period, especially in the 1990s. This is expected as the regression method of JMA-MLR includes a linear term of time for each geographic box, which is equivalent to using a constant trend for data normalization. The flux estimate of JMA-MLR is larger than that of NIES-ML in all years. In 1990 to 2020, JMA-MLR flux is larger than that of NIES-ML3 by  $0.699 \text{ PgC a}^{-1}$  and  $0.506 \text{ PgC a}^{-1}$  respectively. They are about a quarter of the flux in recent years.

Instead of using explicit trends to normalize data or including a linear term of time in the regression, MPI-SOMFFN (Landschützer et al., 2016), CMEMS-FFNN (Chau et al., 2022), CSIR-ML6 (Gregor et al., 2019), and OceanSODA-ETHZ (Gregor and Gruber, 2021) used atmospheric  $\text{CO}_2$  as a predictor so that their models could learn the trends implicitly. Their flux trend patterns indicate different implicit  $\text{CO}_2$  trends. While the fluxes of MPI-SOMFFN and OceanSODA-ETHZ remain rather flat before 2000 and then increase with time, the fluxes of CMEMS-FFNN and CSIR-ML6 show a trend before the early 1990s and after 2000 and remain at a similar level in between. The JENA method (Rödenbeck et al., 2013) involves an inversion model for the ocean chemistry coupled with the atmospheric  $\text{CO}_2$  of an atmospheric transport model. It has the largest inter-annual flux variations. Note that all products except for JENA are monthly with a  $1 \times 1$ -degree spatial resolution. We calculated the monthly mean  $\text{CO}_2$  of JENA in  $2 \times 2.5$ -degree grids using its daily dataset and then filled the  $1 \times 1$ -degree grids with values in the nearest source grids. A different averaging and re-gridding method may yield a different result. The day of the year is also a predictor of CSIR-ML6.

Figure 4B reveals several patterns in the flux differences between NIES-ML3 and other products. The black, cyan, and orange bars represent the mean of a product minus NIES-ML3 in the whole available period, in the early half, and the latter-half years, respectively. NIES-ML3 agrees with CSIR-ML6 and OceanSODA-ETHZ the most in terms of the overall mean difference, which is  $-0.050 \text{ PgC a}^{-1}$  and  $-0.053 \text{ PgC a}^{-1}$ , respectively. Their p-values of two tailed t-test with a significance level of 95%, 0.615 and 0.478 respectively, indicate that the differences are insignificant. While the flux of CSIR-ML6 is smaller than that of NIES-ML3 in the early-half years and larger in the latter-half years, the long-term change of NIES-ML3 is more consistent with that of OceanSODA-ETHZ. The differences between NIES-ML3 and MPI-SOMFFN ( $0.088 \text{ PgC a}^{-1}$ , p-value=0.304), and between NIES-ML3 and CMEMS-FFNN ( $-0.121 \text{ PgC a}^{-1}$ , p-value=0.151) are moderate but insignificant. The fluxes of NIES-NN and JMA-MLR are much larger than that of NIES-ML3, by  $0.240 \text{ PgC a}^{-1}$  (p-value=0.030) and  $0.267 \text{ PgC a}^{-1}$  (p-value=0.000) respectively, especially in the latter-half years of NIES-NN and the early-half years of JMA-MLR. The largest difference was between JENA and NIES-ML3 ( $-0.322 \text{ PgC a}^{-1}$ , p-value=0.000). Overall, the difference between NIES-ML3 and other products is small, about  $0.007 \text{ PgC a}^{-1}$ .

## Conclusion

Our results point out that the ocean  $\text{CO}_2$  trends are an important factor affecting the global ocean  $\text{CO}_2$  reconstruction and flux estimate by machine learning methods. So far, explicit trend methods assumed a constant trend. They yielded much larger flux estimates than most implicit methods in the early and later years of the modelled period. Because the ocean  $\text{CO}_2$  trends tend to track the trends of air  $\text{CO}_2$  and the later increased with time, using a constant ocean  $\text{CO}_2$  trend tends to underestimate the concentration in those years. We proposed a new method to use variant trends for an explicit method that applies machine learning to trend removed data. On average, our flux estimates are significantly lower those of NIES-NN and JMA-MLR. Comparing to the implicit methods, they are smaller than that of MPI-SOMFFN but larger than those of CSIR-ML6, OceanSODA-ETHZ and CMEMS-FFNN. Even though the differences of implicit methods are less significant than those of the explicit methods, the fluxes among the implicit methods depart substantially in early and recent years. This reveals that the ocean  $\text{CO}_2$  trends obtained by the implicit methods could be largely different. All the implicit methods regrouped data by clustering. While the merit point of clustering to regroup data by their biogeochemical properties have been stressed, its demerit point of worsening the data scarcity problem was rarely discussed. Therefore, our results are expected not only to enhance the accuracy of flux estimate by machine learning but also to provide a reference to investigate the trend differences of the implicit methods.

## Data availability statement

The modelled results can be obtained from <https://www.nies.go.jp/doi/10.17595/20220311.001-e.html>. Other data and code used in the study may be provided upon request. Further inquiries can be directed to the corresponding author.

## Author contributions

JZ: Model experiment design, data processing, and draft manuscript. YI: Flux comparisons. TM: Advice on satellite data. TS: Result checking and advice on carbon budget issues. All authors contributed to the article and approved the submitted version.

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## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial

relationships that could be construed as a potential conflict of interest.

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## Appendix

According to Wanninkhof (2014), air-sea CO<sub>2</sub> fluxes can be calculated by the partial pressures of CO<sub>2</sub> in the air ( $pCO_2A$ ,  $\mu\text{atm}$ ) and seawater ( $pCO_2W$ ,  $\mu\text{atm}$ ):

$$\text{flux} = k_0 \cdot k_w \cdot (pCO_2W - pCO_2A), \quad (5)$$

where  $k_0$  ( $\text{mol L}^{-1} \text{atm}^{-1}$ ) denotes the solubility of CO<sub>2</sub> in seawater. It is a function of seawater temperature (K) and salinity ( $\text{g kg}^{-3}$ ):

$$\begin{aligned} \log(k_0) = & -58.0931 + 90.5069 \left( \frac{100}{SST} \right) + 22.2940 \cdot \log \left( \frac{SST}{100} \right) \\ & + SSS \left( 0.027766 - 0.025888 \left( \frac{SST}{100} \right) + 0.0050578 \left( \frac{SST}{100} \right)^2 \right). \end{aligned} \quad (6)$$

The gas transfer velocity  $k_w$  ( $\text{cm h}^{-1}$ ) is a function of wind speed WIND ( $\text{m s}^{-1}$ ) 10 m above the surface and the dimensionless Schmidt number:

$$\begin{aligned} k_w = & \cdot WIND^2 \cdot \left( \frac{Sc}{660} \right)^{-0.5} \\ Sc = & 2116.8 - 136.25 \cdot SST + 4.7353 \cdot SST^2 - 0.092307 \cdot SST^3 + 0.0007555 \cdot SST^4 \end{aligned} \quad (7)$$

in which  $\alpha$  is a coefficient and the unit of SST is Celsius. Wanninkhof (2014) obtained  $\alpha$  as  $0.251 (\text{cm h}^{-1}) (\text{m s}^{-1})^{-2}$  using

the high-resolution data of Cross-Calibrated Multi-Platform wind vector analysis (CCMP). The study of Fay et al. (2021) shows that  $\alpha$  should take a larger value when a monthly wind with  $1 \times 1$ -degree resolution is used. We adopted their value of  $0.271 (\text{cm h}^{-1}) (\text{m s}^{-1})^{-2}$  for ERA5 wind.

The predicted  $fCO_2$  ( $\mu\text{atm}$ ) was converted to partial pressure ( $\mu\text{atm}$ ) by the method of Weiss (1974) and Dickson et al. (2007):

$$\begin{aligned} pCO_2W = & \frac{fCO_2}{\exp \left( \frac{(b+2a) \cdot Ps}{82.05746 \cdot SST} \right)} \\ a = & 57.7 - 0.118 \cdot SST \end{aligned} \quad (8)$$

$$b = -1636.75 + 12.0408 \cdot SST - 0.0327975 \cdot SST^2 + 0.0000316528 \cdot SST^3$$

in which the unit of SST is Kelvin. The partial pressure of air CO<sub>2</sub> was calculated from  $xCO_2A$  by:

$$pCO_2A = xCO_2A \cdot (Ps - pH_2O). \quad (9)$$

The vapor pressure of seawater  $pH_2O$  (atm) was calculated by the method of Weiss & Price (1980):

$$pH_2O = \exp(24.4543 - 67.4509(100/SST) - 4.8489 \log(SST/100) - 0.000544 \cdot SSS) \quad (10)$$

with SST in Kelvin.