1 The spatiotemporal dynamics of the sources and sinks of CO<sub>2</sub> in the global coastal ocean 2

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# 13 Key Points:

- A new high-resolution monthly air-sea  $CO_2$  flux climatology implies a global coastal ocean 15 carbon sink of  $-0.20 \pm 0.02$  Pg C yr<sup>-1</sup>.
  - The zonal mean pattern of the air-sea CO<sub>2</sub> flux in coastal seas follows that of the adjacent open ocean except for river influenced regions.
- The seasonality of the coastal air-sea CO<sub>2</sub> exchange is largely driven by variations in seawater
   *pCO*<sub>2</sub> and temperature.

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## 21 Abstract

In contrast to the open ocean, the sources and sinks for atmospheric carbon dioxide (CO<sub>2</sub>) in the coastal 22 seas are poorly constrained and understood. Here, we address this knowledge gap by analyzing the 23 24 spatial and temporal variability of the coastal air-sea flux of  $CO_2$  (FCO<sub>2</sub>) using a recent high-resolution (0.25 degree) monthly climatology for coastal sea surface partial pressure in  $CO_2$  (pCO<sub>2</sub>). Coastal 25 regions are characterized by CO<sub>2</sub> sinks at temperate and high latitudes and by CO<sub>2</sub> sources at low 26 latitude and in the tropics, with annual mean CO<sub>2</sub> flux densities comparable in magnitude and pattern to 27 those of the adjacent open ocean with the exception of river dominated systems. The seasonal variations 28 in  $FCO_2$  are large, often exceeding 2 mol C m<sup>-2</sup> yr<sup>-1</sup>, a magnitude similar to the variations exhibited 29 across latitudes. The majority of these seasonal variations stems from the air-sea  $pCO_2$  difference, 30 although changes in wind speed and sea-ice cover can also be significant regionally. Globally 31 integrated, the coastal seas act currently as a CO<sub>2</sub> sink of  $-0.20 \pm 0.02$  Pg C yr<sup>-1</sup>, with a more intense 32 uptake occurring in summer because of the disproportionate influence of high latitude shelves in the 33 Northern Hemisphere. Combined with estimates of the carbon sinks in the open ocean and the Arctic, 34 this gives for the global ocean, averaged over the 1998 to 2015 period an annual net CO<sub>2</sub> uptake of -1.7 35  $\pm 0.3 \text{ Pg C yr}^{-1}$ . 36

## 37 **1 Introduction**

Globally, the ocean is currently taking up each year about  $2.4 \pm 0.5$  Petagrams (Pg) of the extra 38 (anthropogenic) carbon added to the atmosphere as a consequence of fossil fuel burning, cement 39 production and land-use change (Le Quéré et al., 2018). While this global ocean uptake is well 40 41 established on the basis of many independent methods (e.g., Gruber et al., 2009, 2019; Keeling & Manning, 2014; Landschützer et al., 2014; Manning & Keeling, 2006; Rödenbeck et al., 2015; 42 Takahashi et al., 2012), the net carbon dioxide (CO<sub>2</sub>) source/sink characteristic of the coastal ocean (a 43 term equivalent to continental shelves in the present study) is still poorly known and subject to intense 44 45 scientific debates. The discussion was launched two decades ago when Tsunogai et al. (1999) suggested, on the basis of an extrapolation of a single local study, that the coastal ocean takes up  $CO_2$ 46

from the atmosphere at a rate in excess of 1 Pg C yr<sup>-1</sup> (note that by convention, a negative flux value 47 corresponds to a CO<sub>2</sub> transfer from the atmosphere to the sea surface). Subsequent studies brought the 48 coastal ocean uptake flux considerably down, with most estimates ranging from -0.45 Pg C yr<sup>-1</sup> (Borges 49 et al., 2005) to -0.21 Pg C yr<sup>-1</sup> (Laruelle et al., 2010). The most important improvements consisted of a 50 more adequate regional aggregation of the local/regional estimates before they were upscaled to the 51 globe (Borges, 2005; Borges et al., 2005; Cai, 2011; Chen et al., 2013; Chen & Borges, 2009; Dai et al., 52 2013; Laruelle et al., 2010). But these estimates were still characterized by large uncertainties (Bauer et 53 al., 2013; Regnier et al., 2013), primarily due to the limited number of observations (typically less than 54 a hundred) available to derive local estimates of the coastal air-sea flux of CO<sub>2</sub> (FCO<sub>2</sub>). A further 55 important limitation was the relatively low granularity of the aggregation used in most upscaling 56 studies, thus neglecting potentially important regional differences in the coastal air-sea CO<sub>2</sub> fluxes. 57 Laruelle et al. (2014) overcame some of these limitations by using a fine aggregation level of 150 58 regional units, and computed globally an average coastal FCO<sub>2</sub> of -0.19 Pg C yr<sup>-1</sup>. They improved the 59 data coverage by taking advantage of the more than 3 million coastal measurements of the sea surface 60 partial pressure of  $CO_2$  (pCO<sub>2</sub>) contained in the global database of the Surface Ocean  $CO_2$  Atlas 61 (SOCAT v2.0, Pfeil et al., 2013). Further, they used various upscaling methods depending on data 62 coverage in the different regions. However, despite these improvements, the still rather poor sampling 63 in many regions made such an aggregation-based upscaling method inherently uncertain, thereby 64 limiting the constraints of these estimates, especially when considering the contribution of the coastal 65 ocean to the global ocean carbon sink (Gruber, 2015). Further, the low spatial resolution of the Laruelle 66 et al. (2014) estimates in many coastal regions did not allow them to evaluate consistently the spatial 67 variability in the coastal environments, and the  $FCO_2$  seasonal cycle could only be assessed for a 68 restricted number of areas, mostly limited to the Atlantic basin. 69

Recently, Laruelle et al. (2017) generated a continuous high spatial resolution (0.25 degree) sea surface  $pCO_2$  product for the coastal regions at monthly timescales for the period 1998-2015. They used a statistical interpolation method developed for the open ocean (Landschützer et al., 2013) and applied it for the first time in coastal regions. This method is based on neural network-inferred statistical

relationships between  $pCO_2$  and a number of environmental variables (i.e., sea surface temperature, 74 wind speed, bathymetry, ...) that are known to control the  $pCO_2$  variability in both time and space. This 75 approach has the large advantage over the previously used aggregation methods in that it estimates 76 relatively reliable  $pCO_2$  values also in data poor regions, as long as other well sampled regions exist that 77 have similar biogeochemical conditions permitting the neural network to be trained for such conditions 78 (see also Rödenbeck et al., 2015). Relative to the work by Laruelle et al. (2014), the new product by 79 Laruelle et al. (2017) also benefited from a substantial improvement of the data coverage in the 80 SOCATv4 product (now 13.6 coastal million observations, Bakker et al., 2016; Sabine et al., 2013). 81 This new high-resolution gridded monthly  $pCO_2$  product for coastal regions provides the basis for a 82 revised estimate of the global and regional coastal CO<sub>2</sub> sink/source characteristics at hitherto 83 unprecedented resolution in time and space. 84

In addition to determining the global coastal carbon sink, we are particularly interested in the seasonal 85 86 dynamics of the coastal air-sea CO<sub>2</sub> fluxes, as this provides insight into the sensitivity of these fluxes to environmental change. The few studies that attempted to identify and quantify the main processes 87 driving the seasonality of FCO<sub>2</sub> remained, however, regional (Arruda et al., 2015; Frankignoulle & 88 Borges, 2001; Nakaoka et al., 2006; Shadwick et al., 2010, 2011; Turi et al., 2014; Yasunaka et al., 89 90 2016) or were limited to a single oceanic basin (i.e., the Atlantic, Laruelle et al., 2014). In addition, with few exceptions (i.e. Yasunaka et al., 2016), most regional studies focused mainly on the seasonal 91 92 variability of  $pCO_2$ , neglecting the potentially important contribution of changes in the atmospheric  $pCO_2$ , the gas transfer velocity and the sea-ice coverage. In the present study, we analyze for the first 93 time the FCO<sub>2</sub> variability both in time and space in the global coastal ocean. We also quantify the major 94 trends associated to the contribution of the sea-ice cover, the wind speed and the  $pCO_2$  gradient 95 variability. 96

#### 97 2 Methodology

98 2.1 Air-sea gas exchange flux calculation

<sup>99</sup> The air-sea gas exchange rate of CO<sub>2</sub> at the air-water interface ( $FCO_2$ , mol C m<sup>-2</sup> yr<sup>-1</sup>) is calculated <sup>100</sup> using the diffusion-limited stagnant film model by Deacon (1977) given by:

(1)

101 
$$FCO_2 = k K_0 (1 - Ice) \Delta pCO_2$$

A positive  $FCO_2$  value corresponds to a transfer of  $CO_2$  from the sea towards the atmosphere (i.e., 102 source of CO<sub>2</sub> for the atmosphere) and a negative value corresponds to that from the atmosphere into 103 the ocean (a sink of  $CO_2$ ).  $K_0$  represents the sea surface temperature (SST) and salinity (SSS) dependent 104 solubility of CO<sub>2</sub> in water (mol C m<sup>-3</sup>  $\mu$ atm<sup>-1</sup>) and is calculated following Weiss (1974).  $\Delta pCO_2$  ( $\mu$ atm) 105 is the difference in the partial pressure of  $CO_2$  between the surface seawater ( $pCO_{2,water}$ , called hereafter 106  $pCO_2$ ) and the atmosphere ( $pCO_{2,air}$ ). Ice is a dimensionless coefficient corresponding to the fraction of 107 the air-water interface (between 0 and 1) covered by sea-ice. k is the gas exchange transfer velocity of 108  $CO_2$  (m yr<sup>-1</sup>) calculated following Wanninkhof et al. (2009): 109

110 
$$k = k_{660} \left(\frac{sc}{660}\right)^{-1/2}$$
 (2)

where  $k_{660}$  represents the gas exchange transfer velocity of CO<sub>2</sub> normalized to a temperature of 20 °C 111 (for a SSS of 35) and is a function of the wind speed measured at 10 meters above the sea surface ( $U_{10}$ ). 112 Several coastal studies proposed  $k_{660}$ -relationships based on empirical methods for the CO<sub>2</sub> exchange in 113 coastal seas (i.e., Jacobs et al., 1999; Kuss et al., 2004; Nightingale et al., 2000; Weiss et al., 2007) but 114 these formulations were usually developed and calibrated locally or regionally (Roobaert et al., 2018). 115 For our study, we use the quadratic formulation of Ho et al. (2011) with respect to wind speed ( $k_{660}$  = 116  $cU_{10}^2$  where c is a constant value) which was calibrated with an extensive collection of k estimates in 117 coastal environments. The Schmidt number (Sc, dimensionless) is calculated using the equation and 118 parameterization given in Wanninkhof (2014): 119

120 
$$Sc = A + B(SST) + C(SST)^2 + D(SST)^3 + E(SST)^4$$
 (3)

121 where A, B, C, D and E have constant values for CO<sub>2</sub> (for a SSS of 35) of 2116.8, -136.25, 4.7353, -

122 0.092307, 0.0007555 respectively, and where *SST* is given in units of degrees Celsius.

123 2.2 Data

We compute the air-sea  $CO_2$  flux (equation 1) for each 0.25 degree grid point in the coastal regions and 124 for each month over the period 1998 to 2015 using the following data products: For  $pCO_2$ , we employ 125 the monthly gridded product by Laruelle et al. (2017) at 0.25 degree resolution for coastal seas. This 126 product was derived from 13.6 million coastal  $pCO_2$  data contained in the Surface Ocean CO<sub>2</sub> Atlas 127 version 4 (SOCATv4, Bakker et al., 2016; Sabine et al., 2013) and in LDEOv2015 (Takahashi et al., 128 2016) using a two-step artificial neural network interpolation technique (SOM-FFN), which allows one 129 to produce contiguous high-resolution  $pCO_2$  maps based on incomplete datasets. The SOM-FFN 130 method generates  $pCO_2$  values based on numerical relationships between independent environmental 131 132 variables (i.e., SST, wind speed, bathymetry, ...) and  $pCO_2$  observations (see Landschützer et al., 2013, 2014, 2016 and Laruelle et al., 2017 for details regarding the method). The atmospheric partial 133 pressures of CO<sub>2</sub> (pCO<sub>2,air</sub>) are calculated from the dry air mixing ratio of CO<sub>2</sub> provided by the NOAA 134 Marine 100 % Boundary Layer reference product assuming humidity 135 136 (https://www.esrl.noaa.gov/gmd/ccgg/mbl/) and using the NCEP reanalysis total pressure at sea level (Kalnay et al., 1996). The monthly  $CO_2$  solubility in seawater ( $K_0$ ) is calculated following Weiss (1974) 137 using monthly mean 0.25° SST and SSS derived respectively from the daily NOAA OI SST V2 138 (Reynolds et al., 2007) and the daily Hadley center EN4 SSS (Good et al., 2013) datasets. To calculate 139 140 k, we use the monthly second moment of the 6 hour  $0.25^{\circ}$  global atmospheric reanalysis ERA-interim wind product (Dee et al., 2011) and the equation proposed by Wanninkhof (2014) for the Schmidt 141 number (Sc) combined with the transformed SST field. The sea-ice cover is from the monthly mean of 142 the daily 0.25° dataset of Reynolds et al. (2007) which is derived from Cavalieri et al. (1996) and 143 Grumbine (1996). We use the most recent versions of these data sets, which cover our entire analysis 144 period (1998-2015). Note that their referencing corresponds to the original release of these databases. 145 From the full time series, we then compute a monthly climatology of the coastal  $FCO_2$  and of all other 146 147 relevant parameters (SST, etc).

#### 148

## 2.3 Spatial analysis and comparison with the open ocean

Following Laruelle et al. (2013, 2014), the outer limit of the coastal region corresponds approximately 149 to the shelf break. Estuaries and inland water bodies are not considered in this study and the total 150 surface area of the coastal domain is 28 million km<sup>2</sup>.  $FCO_2$  and spatially integrated  $FCO_2$  (Tg C yr<sup>-1</sup>) 151 are also analyzed regionally using a division of the continental shelves into 45 regions. This delineation 152 is based on the Margins and CATchment Segmentation (MARCATS, Laruelle et al., 2013) which 153 describes coastal regions characterized by similar climatic and hydrological settings. These 45 154 MARCATS, represented in Figure 1, can also be grouped into 7 major classes based on hydrology and 155 climate following the classification proposed by Liu et al. (2010): (1) Eastern and (2) Western 156 Boundary Currents (EBC and WBC respectively), (3) tropical margins, (4) subpolar and (5) polar 157 margins, (6) marginal seas and (7) Indian margins. For each MARCATS, our results are compared to 158 those calculated in previous studies based on observational data (Laruelle et al., 2014) and on results 159 from an Oceanic General Circulation/Biogeochemical Model (Bourgeois et al., 2016). These two 160 studies use the same definition of the coastal region and take into account the effect of the sea-ice cover 161 in their  $FCO_2$  calculations. 162

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The air-sea CO<sub>2</sub> exchange dynamics in the coastal region is also compared to the climatological air-sea 164 CO<sub>2</sub> fluxes of the open ocean for the 1998-2015 period. Monthly FCO<sub>2</sub> values are calculated for the 165 open ocean following the same protocol as for the coastal seas (see section 2.1) except for the spatial 166 resolution, which is limited to  $1^{\circ} \times 1^{\circ}$ , identical to that of the oceanic pCO<sub>2</sub> climatology of 167 Landschützer et al. (2017) (see Landschützer et al., 2016 for the method) and for the calculation of the k 168 value. The datasets used for monthly sea-ice cover, wind speed, SST, SSS and  $pCO_{2,air}$  are the same as 169 those used for the coastal calculations. These latter ones are aggregated into the 1° x 1° cells of the open 170 ocean product by performing surface weighted averages of the 16 0.25° cells comprised within each 1° 171 x 1° cell. We use a quadratic  $k_{660}$ -formulation developed for a global scale  $FCO_2$  calculation based on 172 the <sup>14</sup>C inventory method (Sweeney et al., 2007; Takahashi et al., 2009; Wanninkhof, 1992, 2014) to 173

calculate k. Because the  ${}^{14}$ C inventory method is applied to the entirety of the global ocean, it is suitable 174 to derive k for the open ocean but not for the coastal seas where other regional process than wind driven 175 turbulence may disturb the gas exchange at the air-water interface. In order to limit uncertainties on k 176 associated to the wind product at its specific spatio-temporal resolution (Roobaert et al., 2018), we 177 rescale the coefficient c to fit to a global average gas exchange transfer velocity of 16 cm  $h^{-1}$ 178 (Wanninkhof et al., 2013) for the period of our study. The open ocean domain is delineated by a land-179 sea mask from Landschützer et al. (2017) excluding the Arctic Ocean basin and coastal regions. Cells 180 overlapping both oceanic and coastal domains are removed from the open ocean. The resulting open 181 ocean covers a total surface area of  $311 \times 10^6 \text{ km}^2$ . 182

#### 183 2.4 Seasonality analysis

To explore the drivers of the seasonal  $FCO_2$  variability in the coastal regions, we follow the strategy developed by Doney et al. (2009) and Couldrey et al. (2016) and construct, for each climatological month, spatial maps of the  $FCO_2$  anomaly ( $FCO'_2$ ). For each grid cell, the monthly  $FCO_2$  anomaly is calculated as the difference between the  $FCO_2$  for a given month and the climatological annual mean  $\overline{FCO_2}$ :

189 
$$FCO_2' = FCO_2 - \overline{FCO_2}$$
(4)

where  $FCO'_2$  and  $FCO_2$  represent the anomaly and the air-sea CO<sub>2</sub> exchange rate calculated for each month, respectively.

A positive  $FCO_2$  for a given month implies that the  $FCO_2$  is a stronger source or a weaker sink of  $CO_2$ 192 for the atmosphere during that month compared to the 1998-2015 mean  $FCO_2$  while a negative value 193 implies a stronger sink/weaker source. In our seasonal analysis, winter consists of the months of 194 January, February and March in the Northern Hemisphere and of July, August and September in the 195 Southern hemisphere. We then decompose the drivers of the seasonal  $FCO_2$  variability following the 196 procedure described by Couldrey et al. (2016) to isolate the contributions of the different factors to the 197 total FCO<sub>2</sub> variability. Using a Reynolds decomposition for turbulent flow, the monthly variability of a 198 term z which is a function of 3 forcing components (z = wxy) can be expressed as: 199

200 
$$z' = (wxy)'$$
 (5)

201 
$$z' = (w'\bar{x}\bar{y}) + (\bar{w}x'\bar{y}) + (\bar{w}\bar{x}y') + [(w'x'y' - \overline{w'x'y'}) + (w'x'\bar{y} - \overline{w'x'\bar{y}}) + (w'\bar{x}y' - \overline{w'\bar{x}y'}) + (w'\bar{x}y' - \overline{w'\bar{x}y'}) + (\bar{w}x'y' - \overline{\bar{w}x'y'})]$$
(6)

In this study we apply this decomposition to equation 1. z and z' thus represent  $FCO_2$  and its anomaly calculated for each month, respectively. The terms w and x correspond to the sea-ice cover and  $\Delta pCO_2$ drivers, respectively. The term y corresponds to the combined effects of k and  $K_0$ , which isolates the wind speed driver since the product of  $cU_{10}^2 \left(\frac{Sc}{660}\right)^{-\frac{1}{2}} K_0$  is invariant with temperature (Etcheto &

207 Merlivat, 1988; Wanninkhof, 2014). Equation 6 can then be expressed as:

$$FCO_{2}' = \left( (1 - ice)' \overline{\Delta pCO_{2}}(\overline{kK_{0}}) \right) + \left( \overline{(1 - \iotace)} \Delta pCO_{2}'(\overline{kK_{0}}) \right) + \left( \overline{(1 - \iotace)} \overline{\Delta pCO_{2}}(kK_{0})' \right) + \left( (1 - \iotace)' \Delta pCO_{2}'(kK_{0})' - \overline{(1 - \iotace)' \Delta pCO_{2}'(kK_{0})'} \right) + \left( \overline{(1 - \iotace)' \Delta pCO_{2}'(kK_{0})'} \right) \right) + \left( \overline{(1 - \iotace)' \Delta pC$$

210 
$$\left( (1 - ice)' \Delta p C O_2' \overline{(kK_0)} - \overline{(1 - ice)' \Delta p C O_2' \overline{(kK_0)}} \right) + \left( (1 - ice)' \overline{\Delta p C O_2} (kK_0)' - \overline{(1 - ice)' \overline{\Delta p C O_2} (kK_0)'} \right) + \left( \overline{(1 - ice)' \Delta p C O_2} (kK_0)' - \overline{(1 - ice)' \overline{\Delta p C O_2} (kK_0)'} \right) \right)$$
(7)

212 
$$\left(\overline{(1-\iota ce)}\Delta pCO_{2}^{\prime}(kK_{0})^{\prime}-\overline{(1-\iota ce)}\Delta pCO_{2}^{\prime}(kK_{0})^{\prime}\right)\right]$$
(7)

The first three terms on the right hand side of equation 7 represent the seasonal  $FCO_2$  anomaly from a long-term annual mean induced by the variability of the sea-ice cover  $((1 - ice)'\Delta pCO_2(\overline{kK_0}), \text{ term 1})$ , of the air-sea  $pCO_2$  gradient  $(\overline{(1 - ice)}\Delta pCO_2'(\overline{kK_0}), \text{ term 2})$  and of the wind speed  $(\overline{(1 - ice)}\Delta pCO_2(kK_0)', \text{ term 3})$ , respectively. Terms comprised between the large brackets correspond to the cross-correlation between the three drivers and are treated as a single term (term 4, sum of the four terms) in this study. The sum of all terms on the right-hand side of equation 7 is identical to the seasonal  $FCO_2$  anomaly calculated in equation 4.

In order to evaluate how much a driver contributes to the monthly  $FCO'_2$ , we calculate, for each grid cell, the slope ( $\beta$ ) of a simple linear regression between monthly  $FCO'_2$  and monthly values resulting from the different drivers (terms 1 to 4 in equation 7). For example, for the contribution to the  $FCO'_2$ induced by the seasonal variability of the wind speed,  $\beta$  is calculated as follows:

224 
$$\frac{\partial FCO_2'}{\partial \overline{(1-\iota ce)} \Delta pCO_2(kK_0)'} = \beta_{wind}$$
(8)

A value of  $\beta$  equal to 1 means that the term analyzed explains entirely the *FCO*<sub>2</sub> anomaly while a value of 0 means that *FCO*'<sub>2</sub> is insensitive to that term. When  $\beta$  is greater than 1, the particular term produces larger anomalies than *FCO*'<sub>2</sub>, which are compensated by anomalies of opposite sign and are attributed to other terms. The sum of all the  $\beta$  terms adds up to 1, when including the cross-correlation terms.

230 For both the open ocean and coastal regions, the  $FCO_2$  uncertainty stems from the combination of the uncertainties associated with the different terms of equation 1 used to calculate  $FCO_2$ : (1) the air-sea 231  $pCO_2$  gradient constrained by the water  $pCO_2$  field generated by the SOM-FFN interpolation method 232  $(\sigma_{pCO2}, \text{Landschützer et al., 2014, 2018; Laruelle et al., 2017}), (2)$  the choice of the  $k_{660}$ -formulation for 233 the gas exchange transfer velocity ( $\sigma_k$ ) and (3) the choice of the wind product ( $\sigma_{wind}$ , Roobaert et al., 234 2018). Considering that these three sources of error are independent from each other and following the 235 strategy used in Landschützer et al. (2014, 2018), we calculate the total error ( $\sigma$ ) as the square root of 236 the sum of the three squared errors: 237

238 
$$\sigma = \sqrt{\sigma_{pCO2}^2 + \sigma_k^2 + \sigma_{wind}^2}$$
(9)

We estimate the flux uncertainty associated to the wind product ( $\sigma_{wind}$ ) as the standard deviation of the 239 CO<sub>2</sub> flux obtained using three different wind products, namely the ERA-interim (Dee et al., 2011), the 240 Cross-Calibrated Multi-Platform Ocean Wind Vector 3.0 (Atlas et al., 2011) and the NCEP/NCAR 241 reanalysis 1 (Kalnay et al., 1996). We exclude the widely used NCEP/DOE AMIP-II Reanalysis 242 (Kanamitsu et al., 2002) climatology from our analysis because of its lesser constrained values at the 243 air-water interface (Roobaert et al., 2018; Winterfeldt & Weisse, 2008). Calculations are performed for 244 245 each wind product following the method described in sections 2.1 and 2.2 to produce three sets of  $FCO_2$ . For the open ocean, the fluxes are calculated by adjusting the value of c for each wind product 246

247 (see Roobaert et al., 2018, for further details).  $\sigma_k$  is estimated, for both the open ocean and the coastal regions, as the  $FCO_2$  standard deviation resulting from the use of four  $k_{660}$ -formulations applied to the 248 same wind field (ERA-interim). We apply the formulations of Ho et al. (2011), Sweeney et al. (2007), 249 Takahashi et al. (2009) and Wanninkhof (2014), all of which are suitable for global-scale applications. 250 251 There exist numerous formulations of k in coastal environments which but those are designed from and for regional studies (Roobaert et al., 2018). We thus decided to follow the same strategy and equations 252 (other than the one from Ho et al., 2011) for our uncertainty analysis in the coastal ocean as the strategy 253 applied to the open ocean for the sake of consistency. The uncertainty of the  $pCO_2$  gradient across the 254 air water interface ( $\sigma_{pCO2}$ ) is the result of the uncertainty in both the oceanic and atmospheric pCO<sub>2</sub>. 255 256 However, the uncertainty of atmospheric  $pCO_2$  is much lower than that of the oceanic  $pCO_2$ (Landschützer et al., 2018) permitting us to assume that the uncertainty associated to the  $pCO_2$  gardient 257 at the air-water interface can be entirely attributed to that of oceanic  $pCO_2$  ( $\theta_{pCO_2}$ ). The latter can be 258 decomposed into 3 sources of uncertainty:  $\theta_{map}$  (the uncertainty obtained by comparing the pCO<sub>2</sub> 259 products derived from the SOM-FFN with gridded observations from the SOCAT database),  $\theta_{grid}$  (the 260 uncertainty resulting from the gridding of the original SOCAT observations into a 1° open ocean grid 261 and a 0.25° coastal grid) and  $\theta_{obs}$  (the experimental uncertainty associated to pCO<sub>2</sub> measurements in 262 the field). For a given region (i.e., MARCATS or oceanic basin), each source of uncertainty is divided 263 by the square root of its degree of freedom, which is equal to the number of  $pCO_2$  samples (N) collected 264 in the region, except for  $\theta_{map}$ , for which N is corrected using the lag 1 autocorrelation coefficient to 265 evaluate the effective sample size (N<sub>eff</sub>), following a procedure described in Landschützer et al. (2018). 266 The calculation of the lag 1 autocorrelation coefficient involves the random selection of 1000 data 267 points within each region. In order to generate the most robust uncertainties possible, the procedure is 268 repeated 10 times and the final value of N<sub>eff</sub> is the median generated by these 10 ensemble runs. 269

271 
$$\theta_{pCO2} = \sqrt{\left(\frac{\theta_{obs}}{\sqrt{N}}\right)^2 + \left(\frac{\theta_{grid}}{\sqrt{N}}\right)^2 + \left(\frac{\theta_{map}}{\sqrt{N_{eff}}}\right)^2} \tag{10}$$

272

In our calculations,  $\theta_{map}$  is calculated as the root mean squared deviation between the pCO<sub>2</sub> field and 273 the gridded observations from SOCAT over the region of interest.  $\theta_{grid}$  is considered to be 5 µatm 274 following the estimate of Sabine et al. (2013) for the open ocean. We assume the same value for the 275 coastal ocean in spite of the different spatial resolutions (i.e., 1 degree and 0.25 degree for the open and 276 coastal oceans, respectively).  $\theta_{obs}$  is considered to be 2 µatm following Pfeil et al. (2013). Finally, the 277 uncertainty on the FCO<sub>2</sub>,  $\sigma_{pCO2}$  (in mol C m<sup>-2</sup> yr<sup>-1</sup>) resulting from the uncertainty on the pCO<sub>2</sub> field 278  $(\theta_{pCO2}, \text{ in } \mu \text{atm})$ , is obtained by applying equation 1 to the global  $\theta_{pCO2}$  field. Globally, the averaged 279 biaises between  $pCO_2$  generated by the SOM-FFN algorithm and observations are null for the open 280 ocean and coastal seas (Landschützer et al., 2014; Laruelle et al., 2017). At the scale of MARCATS, 281 282 however, a few regions with significant biaises have been identified in coastal seas (i.e., the Peruvian upwelling, MARCATS 4, the Moroccan upwelling, MARCATS 22, as well as along the western 283 Arabian Sea, MARCATS 27; Laruelle et al., 2017). However, considering their relatively small 284 cumulated surface area compared to the global coastal ocean (  $\sim 1$  %), the influence of theses biaises on 285 286 the global coastal  $FCO_2$  is negligible and no biais correction was performed.

## 287 **3 Results**

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#### 3.1 Spatial distribution

In the climatological annual mean (Figure 2a), about two-thirds of the coastal  $0.25^{\circ}$  x  $0.25^{\circ}$  grid points act as sinks for atmospheric CO<sub>2</sub>, while the remaining one-third are sources or are near-neutral. The CO<sub>2</sub> sink regions are mainly found at temperate and high latitudes (i.e., poleward of 30°) and the strongest sources occur in the tropics. Globally, the dominance of sinks over sources leads to a sizeable average uptake *FCO*<sub>2</sub> density of -0.58 mol C m<sup>-2</sup> yr<sup>-1</sup>, which amounts to a global uptake of -195 Tg C yr<sup>-1</sup> (-0.20 Pg C yr<sup>-1</sup>) over the 1998 through 2015 period. Our global mean flux density is very close to the -0.51 mol C m<sup>-2</sup> yr<sup>-1</sup> estimate of Laruelle et al. (2014) once their original value (-0.7 mol C m<sup>-2</sup> yr<sup>-1</sup>) is adjusted for the presence of sea-ice.

The global pattern of the coastal air-sea CO<sub>2</sub> fluxes becomes clearer when the fluxes are integrated by 297 MARCATS regions (Figure 3a and Table S1). As already indicated by the grid point level analysis 298 above, the vast majority of the MARCATS (31 over 45, 71 % of the total surface area of the coastal 299 seas) act as a sink for atmospheric CO<sub>2</sub>. The strongest and most consistent sink regions are found in the 300 polar and subpolar coastal seas. The South Greenland region (MARCATS 15) has the highest sink 301 strength, taking up annually -2.90 mol C m<sup>-2</sup> yr<sup>-1</sup>, closely followed by the Norwegian basin (MARCATS 302 303 16). On average, the marginal seas also act as sinks, but the spread among them is very large with three regions presently acting as sources, i.e., the Gulf of Mexico (MARCATS 9), the Red Sea (MARCATS 304 28) and the Persian Gulf (MARCATS 29). On the other hand, the Black Sea (MARCATS 21) has the 305 second largest uptake flux density of MARCATS regions. All Western Boundary Current (WBC) 306 307 regions are sinks, but with a relatively modest average flux density. The Eastern Boundary Current (EBC) regions are also moderate sinks, but with a somewhat larger spread as well as one region 308 emitting CO<sub>2</sub> to the atmosphere (Moroccan upwelling, MARCATS 22). In this region, the SOM-FFN 309 technique underestimates the observed  $pCO_2$  because of the scarcity in data to train the algorithm 310 (Laruelle et al., 2017). The resulting  $FCO_2$  yields thus only a weak source of  $CO_2$  for the atmosphere 311 while observations suggest a more intense evasion (Laruelle et al., 2014). The remaining 31 % of the 312 MARCATS that release CO<sub>2</sub> into the atmosphere are all located in the tropical coastal regions and in the 313 Indian Ocean, except for the West Arabian Sea, MARCATS 27. The strongest source is observed in the 314 Persian Gulf (MARCATS 29) with a  $FCO_2$  value of 0.85 mol C m<sup>-2</sup> yr<sup>-1</sup>. The 31 MARCATS sink 315 regions absorb altogether -219 Tg C yr<sup>-1</sup> while the 14 MARCATS source regions release only 25 Tg C 316 yr<sup>-1</sup>. The most important contribution to the global sink (Figure 3b) is provided by the polar/subpolar 317 margins (-143 Tg C yr<sup>-1</sup> over 14 MARCATS), followed by the marginal seas (-43 Tg C yr<sup>-1</sup> over 9 318 MARCATS). The WBCs and the EBCs also act as CO<sub>2</sub> sink with integrated FCO<sub>2</sub> values of -19 Tg C 319 yr<sup>-1</sup> (5 MARCATS) and -9 Tg C yr<sup>-1</sup> (6 MARCATS), respectively. The tropical margins (15 Tg C yr<sup>-1</sup>, 7 320 MARCATS) and the Indian margins (4 Tg C yr<sup>-1</sup>, 4 MARCATS) act as sources. 321

#### 322 3.2 Comparison with the open ocean

The spatial distribution of the mean  $FCO_2$  for the 1998 through 2015 period from the Landschützer et al. (2017)  $pCO_2$  product for the open ocean is overall remarkably similar to that found along the continents (Figures 2a and 2b). As discussed by Landschützer et al. (2014), the strong outgassing in the tropical regions are overcompensated by the strong uptake in the temperate regions and the high latitude North Atlantic (see also Takahashi et al., 2009; Gruber et al., 2009). Globally, the open ocean acts as a net sink of CO<sub>2</sub> for the atmosphere with a global mean  $FCO_2$  of -0.37 mol C m<sup>-2</sup> yr<sup>-1</sup>. This value is 36 % lower than that calculated here for the coastal regions (-0.58 mol C m<sup>-2</sup> yr<sup>-1</sup>).

There are a few regions, however, where the coastal flux densities diverge quite substantially from those 330 in the adjacent open ocean (Figures 2a and 2b). Most of these differences correspond to regions 331 influenced by large rivers which include, in the Atlantic basin, the mouths of the two large Argentinean 332 estuaries (the Rio de la Plata and the Bahia Blanca) which act as sources in opposition to negative  $FCO_2$ 333 values recorded in the surrounding adjacent open ocean. In contrast the Amazon plume acts as a sink (< 334 -1 mol C m<sup>-2</sup> yr<sup>-1</sup>) while the adjacent open ocean is a source. In the Indian basin, differences are 335 observed near the mouths of Zambezi and Pungwe rivers (southeast of Africa) and along part of the 336 coast of Madagascar. Conversely, in Southeast Asia, coastal seas are generally FCO2 sources while the 337 surrounding open ocean takes up CO<sub>2</sub>. In the Pacific basin, negative FCO<sub>2</sub> values are observed along 338 the coast and near the Baja California Peninsula coast while  $FCO_2$  is positive in the adjacent open 339 340 ocean. Two other regions, namely in the western Arabian Sea and in the Peruvian upwelling current (MARCATS 27 and 4, respectively), also present contrasted differences with the open ocean. But these 341 342 differences need be considered with caution due to limitations of the neural network technique of Laruelle et al. (2017) in regions where only limited data are available for calibration, and for which no 343 344 other region with similar biogeochemical conditions can be found for training. These two regions are characterized by strong upwelling that are known to create very high and variable  $pCO_2$  values (see e.g., 345 Friederich et al. (2008) for the Peruvian upwelling). Yet, the neural network estimates of Laruelle et al. 346 (2017) suggest very low  $pCO_2$  values, creating sinks in the coastal regions, whereas the adjacent open 347

ocean in both the western Arabian Sea and eastern tropical Pacific are strong sources. We thus regard the coastal flux estimates in these two upwelling regions as questionable.

To emphasize the similarity of the air-sea CO<sub>2</sub> fluxes in the open ocean and in the coastal regions, we 350 analyze the zonal mean profiles in Figure 4a. For both open and coastal ocean, Northern latitudes (> 40° 351 N) are characterized by intense sinks (< -1 mol C m<sup>-2</sup> yr<sup>-1</sup>) followed by an increase of  $FCO_2$  towards 352 values close to equilibrium with the atmosphere. With the exception of the latitudinal bands comprised 353 between 60°- 45° in both hemispheres where the coastal regions are more intense  $CO_2$  sinks than the 354 open ocean,  $FCO_2$  is higher in the open ocean than in the coastal regions at most latitudes. In the 355 tropics, the coastal and the open ocean are both sources of CO<sub>2</sub> for the atmosphere. However, the 356 latitudinal band where the open ocean releases CO<sub>2</sub> (15° N - 15° S) is more restricted compared to that 357 of the coastal ocean which expands from 30° N to 20° S. Around the equator, however, coastal seas 358 exhibit  $FCO_2$  values close to the equilibrium while the open ocean displays its strongest source (1.29) 359 mol C m<sup>-2</sup> yr<sup>-1</sup>) mainly driven by the equatorial Pacific upwelling. At more austral latitudes, both the 360 coastal and the open ocean act as CO<sub>2</sub> sinks, with a progressive increase in FCO<sub>2</sub> until 40° S. South of 361  $60^{\circ}$  S, FCO<sub>2</sub> values are close to equilibrium with the exception of a pronounced source around  $60^{\circ}$  S. 362

Although the meridional distribution of the average flux density  $FCO_2$  is relatively similar between the 363 open and coastal ocean, the integrated FCO<sub>2</sub> shows very different patterns (Figure 4b and c). This 364 results primarily from the large differences in the surface areas. Due to the wide distribution of the 365 coastal regions north of 60° N (7.1 million km<sup>2</sup> which represents 25 % of the total surface area of the 366 coastal seas) and to the presence of intense  $FCO_2$  sinks there, more than one-third of the global coastal 367 ocean uptake is provided by these regions, taking up -98 Tg C yr<sup>-1</sup> (Table 1). In contrast, only a small 368 fraction (6 %, -87 Tg C yr<sup>-1</sup>) is provided by the open ocean regions > 60° N, almost entirely due to the 369 small surface area of the open ocean in those regions (1 %). Another sink is located in the coastal seas 370 along the Pacific basin where -54 Tg C yr<sup>-1</sup> is taken up (when the seas of Japan and Okhotsk are 371 included) followed by the Atlantic basin (-27 Tg C yr<sup>-1</sup>, Gulf of Mexico included). Marginal seas (-18 372 Tg C yr<sup>-1</sup>), and the coastal in the Southern Ocean (south of 45° S, -17 Tg C yr<sup>-1</sup>) and in the Indian Ocean 373 where the latter represents a source of  $CO_2$  (10 Tg C yr<sup>-1</sup>), contribute to less than 10 % to the global 374

uptake. By combining the open ocean and coastal fluxes estimated using the two  $pCO_2$  climatologies of 375 Landschützer et al. (2017) and Laruelle et al. (2017) respectively, we can now assess, for the first time, 376 377 the true global ocean uptake flux of  $CO_2$  (Table 1). To this end, we also need to account for the uptake flux in the Arctic ocean basin which is not included in the  $pCO_2$  climatology of Landschützer et al. 378 (2017). Using an estimate of  $-0.12 \pm 0.06$  Pg C yr<sup>-1</sup> for the Arctic basin by Schuster et al. (2013) which 379 excludes the Baffin Bay and Nordic seas, the global net uptake of CO<sub>2</sub> by the entire ocean for the 1998 380 to 2015 period amounts to  $-1.7 \pm 0.3$  Pg C yr<sup>-1</sup>. At the basin scale, the combined continental 381 coastal/open ocean estimate reveals that all oceans act as a CO<sub>2</sub> sink for the atmosphere with a relatively 382 equal contribution between the Atlantic (-393 Tg C yr<sup>-1</sup>, 25 % of the global uptake), the Pacific (-386 383 Tg C yr<sup>-1</sup>, 25 %) and the Indian (-322 Tg C yr<sup>-1</sup>, 20 %) followed by the Southern Ocean (-275 Tg C yr<sup>-1</sup>, 384 18 %). 385

#### 386

## 3.3 Seasonality analysis

The global spatial distribution of the amplitude of the seasonal cycle in the air-sea CO<sub>2</sub> exchange flux 387 for coastal regions is presented in Figure 5a, expressed as the root-mean-square (RMS) for each grid 388 cell of the monthly  $FCO_2$  anomalies ( $FCO'_2$ ). Globally, the average  $RMS_{FCO'_2}$  of the seasonal cycle of all 389 grid cells taking into account their variable surface area is 0.8 mol C m<sup>-2</sup> yr<sup>-1</sup>. In general, high seasonal 390 variabilities (i.e., high  $RMS_{FCO_2}$ ) are found in the Northern Hemisphere from mid- to high-latitudes and 391 in the Southern hemisphere south of 40° S. In contrast, the equatorial and tropical regions exhibit less 392 pronounced seasonal variability except for some local areas such as the Amazon estuarine plume. The 393 seasonal amplitude in grid cells located at high latitudes (> 40° in both hemispheres) can reach values 394 (i.e., 2 mol C m<sup>-2</sup> yr<sup>-1</sup>) comparable to the difference in the mean  $FCO_2$  density observed between the 395 polar and equatorial regions (Figure 3). The spatially averaged  $RMS_{FCO_2}$  aggregated at the larger spatial 396 scale of the MARCATS regions (Figure 6) reveals that only three MARCATS have a seasonal 397 variability with a RMS<sub>FCO2</sub> exceeding 1 mol C m<sup>-2</sup> yr<sup>-1</sup>. They are all localized in marginal seas (Baltic 398 Sea, MARCATS 18, Black Sea, MARCATS 21 and the Hudson Bay, MARCATS 12) with the highest 399

seasonal variability observed in the Baltic Sea ( $RMS_{FCO_2'}$  of 3.4 mol C m<sup>-2</sup> yr<sup>-1</sup>). Among the other regions,  $RMS_{FCO_2'}$  values > 0.5 mol C m<sup>-2</sup> yr<sup>-1</sup> are mostly found in polar and subpolar coastal seas, in most of the marginal seas as well as in one or two MARCATS of the EBC (the Iberian upwelling, MARCATS 19) and the WBC (the Florida upwelling, MARCATS 10, and the Yellow sea, MARCATS 39).

The temporal distribution of the  $FCO'_2$  seasonal cycles for different latitudinal bands is shown in Figure 5b to 5f. These bands were selected in order to (1) compare the seasonal dynamics between the two hemispheres consistently and (2) evaluate the seasonal dynamics between high, mid- and low latitudes. For each latitudinal band, monthly  $FCO'_2$  are calculated from the mean of the surface weighted average  $FCO'_2$  of all grid cells pertaining to that band. Their seasonal amplitude is also presented in each panel (RMS<sub>FCO'\_2</sub>). The latter is calculated from the mean seasonal profile and consequently can be lower than the average of all grid cells for a given latitudinal band.

The seasonal cycle is more pronounced at higher latitudes and in regions with a strong seasonal signal 412 (> 40° in both hemispheres),  $RMS_{FCO'_2}$  values vary from 0.5 to 0.7 mol C m<sup>-2</sup> yr<sup>-1</sup>. The amplitude of the 413 seasonal variability at lower latitudes between 40° S and 40° N is smaller with a  $RMS_{FCO'_2}$  value of 0.25 414 mol C m<sup>-2</sup> yr<sup>-1</sup>. At low latitudes between 10° N and 10° S, the amplitude of the seasonal cycle is very 415 low compared to other latitudinal bands (not shown in Figure 5, see Figure 8 for further details). 416 Overall, FCO<sub>2</sub> seasonal cycles are very similar within the same latitudinal bands in both hemispheres. 417 Generally, the largest difference in  $FCO_2$  intensity between seasons is observed between summer and 418 winter except for the temperate region between 40° - 60° in both hemispheres where they are observed 419 in spring and fall. At high latitudes (> 60° in both hemispheres), FCO2 reaches its minimum (most 420 negative FCO2 value indicating the strongest sink/weakest source) in summer and its maximum (most 421 positive FCO'2 value indicating the strongest source/weakest sink) in winter. These two extremes are 422 slightly shifted in time between 40° - 60° in both hemispheres where they are observed during spring 423 and fall, respectively. Compared to the high latitudes, regions between 40° S and 40° N exhibit a six-424 month phase shift with the minimum FCO' occurring in winter and the maximum in summer. Globally, 425

426 coastal seas act as a  $CO_2$  sink all year long with a slightly more intense monthly uptake during spring (-427 18.9 Tg C month<sup>-1</sup>) and summer (-21.1 Tg C month<sup>-1</sup>) while winter and fall each contributes to an 428 equivalent  $CO_2$  uptake flux of -12.5 Tg C month<sup>-1</sup>. In the open ocean, the more intense  $CO_2$  sink occurs 429 in winter (-195.6 Tg C month<sup>-1</sup>) followed by fall (-133 Tg C month<sup>-1</sup>), spring (-122.5 Tg C month<sup>-1</sup>) and 430 summer (-7 Tg C month<sup>-1</sup>), respectively.

Global maps of the respective contribution of the different drivers (sea-ice cover, wind speed,  $\Delta pCO_2$ 431 and cross-correlation term,  $\beta$  in equation 8) to the total seasonal  $FCO_2$  cycle are presented in Figure 7 432 (see Figure S1 in supplement for values integrated per MARCATS unit). In order to highlight regions 433 with the highest absolute seasonal variability, the  $\beta$  terms are multiplied by the RMS of the total  $FCO_2^{\prime}$ . 434 Although there is a significant regional variability for the same latitudinal band (i.e. from one 435 MARCATS to another, Figure S1), a strong correlation between the seasonal cycle of  $FCO_2$  and that of 436  $\Delta pCO_2$  is observed for the vast majority of latitudinal zones. Flux and gradient anomalies are in phase 437  $(\beta_{\Delta pCO_2} > 0)$  and the high values of  $\beta_{\Delta pCO_2}$  demonstrate that changes in  $\Delta pCO_2$  explain the majority of 438 the seasonal changes in FCO<sub>2</sub>. The correlation between the other terms ( $\beta_{sea-ice}$ ,  $\beta_{wind}$ ,  $\beta_{cross-correlation}$ ) and 439  $FCO'_2$ , in addition to presenting more variable contributions within the same latitudinal band (Figure 440 S1), is more complex than that of  $\Delta pCO_2$  and can be either in phase with that of the  $FCO'_2$  ( $\beta > 0$ ) in 441 some regions or anti-correlated ( $\beta < 0$ ) in others. For instance, the anomaly associated with the sea-ice 442 cover can either be in phase ( $\beta_{sea-ice} > 0$ ) or anti-correlated ( $\beta_{sea-ice} < 0$ ) with FCO<sub>2</sub> depending on 443 whether  $FCO_2$  is a source or a sink of CO<sub>2</sub> for the atmosphere and hence on the  $pCO_2$  gradient.  $\beta_{sea-ice}$  is 444 positive when  $\Delta pCO_2 < 0$  and negative when  $\Delta pCO_2 > 0$ . 445

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Although the grid point and the MARCATS-level analysis reveal significant small-scale spatial variabilities in the  $\beta$  terms, the temporal evolution of the different drivers aggregated by latitudinal bands confirms that the changes in  $\Delta pCO_2$  explain most of the seasonal variations of  $FCO_2$  (Figure 8). In some regions (i.e., between 10° - 40° N),  $\Delta pCO_2$  explains even all of the  $FCO_2$  variability (superposition between black and red curves in Figure 8) while at latitudes > 40° N, the seasonal  $\Delta pCO_2$ 

variability induces larger anomalies than the total  $FCO'_2$ , which are compensated by the other drivers as 452 an dampening effect on FCO2 seasonal variability. Although the effect of the sea-ice cover can be 453 important locally in the Southern hemisphere, it only contributes slightly to the seasonal change in 454  $FCO_2$  in bands south of 60° S. In the Northern Hemisphere and especially for latitudinal bands > 60° N, 455 the seasonal  $FCO_2$  profile associated with the change of the sea-ice cover is pronounced underlining the 456 important contribution of this driver in this region. Although there are regions where  $FCO'_2$  is in phase 457 with the one generated by wind speed such as in the Barents Sea ( $\beta_{wind} > 0$ ),  $\beta_{wind}$  tends to act as a 458 compensating term with respect to other parameters for most latitudinal bands. This is particularly true 459 in the latitudinal band >  $60^{\circ}$  N, where the  $FCO'_{2}$  seasonal signal is in opposition to the one generated by 460 wind speed. The effect of wind speed on the  $FCO_2$  variability is mainly found in latitudinal band > 40° 461 in both hemispheres. In the other regions, the effect of the wind either intensifies or dampens the 462 magnitude of the FCO<sub>2</sub> seasonality depending on the month, resulting in a negligible net effect except 463 in bands  $0^{\circ}$  -  $10^{\circ}$  S where it contributes significantly to the (small) FCO<sub>2</sub> variability. Finally, the 464 contribution of the cross-correlation terms illustrates non-linear interactions between the various drivers 465 and has a negligible effect on the  $FCO_2$  variability in about half of the latitudinal bands. The cross terms 466 are significant only at  $> 60^{\circ}$  S and between  $40^{\circ}$  N -  $60^{\circ}$  N. In the latter latitudinal band, however, the 467 contribution of the cross terms to the seasonality of  $FCO_2$  remains lower than that of both  $\Delta pCO_2$  and 468 wind speed. Globally, when all of the coastal seas are integrated (bottom panel of Figure 8), the 469 seasonal pattern mostly follows that of the high latitudes with a minimum  $FCO'_2$  in summer and a 470 maximum in winter. The  $RMS_{FCO'_2}$  amplitude of the global seasonal profile amounts to a value of 0.1 471 mol C m<sup>-2</sup> yr<sup>-1</sup>. Similarly to latitudes > 40° N, the global seasonal  $\Delta pCO_2$  variability induces larger 472 anomalies than the total  $FCO'_2$ , which are partly compensated by the effect of wind. 473

## 474 3.4 Quantification of uncertainty

In coastal seas, the total uncertainty ( $\sigma$ ) varies widely from one MARCATS to another, with values ranging from 0.1 mol C m<sup>-2</sup> yr<sup>-1</sup> to 0.8 mol C m<sup>-2</sup> yr<sup>-1</sup> (Figure 3 and Table S1). The largest uncertainty is found in areas along the coast of Southwest Africa (MARCATS 24) and values of  $\sigma \ge 0.5$  mol C yr<sup>-1</sup> are

mainly encountered in subpolar margins (MARCATS 1, 5, 11 and 42), EBCs (MARCATS 4, 22, 24), in 478 marginal seas (MARCATS 21, 40, 41) and in the Norwegian Basin of polar margins. In MARCATS 12 479 (Hudson Bay), 29 (Persian Gulf), 30 (East Arabian Sea) and 31 (Bay of Bengal), most of which are 480 located in the Indian Ocean,  $\sigma$  is low ( $\leq 0.1 \text{ mol m}^{-2} \text{ yr}^{-1}$ ). However, they should be considered with 481 caution since the contribution of the pCO<sub>2</sub> uncertainty ( $\sigma_{pCO2}$ ) cannot be calculated due to the absence 482 of observational  $pCO_2$  data in those regions. Globally, for the coastal ocean, the  $FCO_2$  uncertainty 483 amounts to 0.02 Pg C yr<sup>-1</sup> (~ 10 % of the coastal seas  $FCO_2$ , Table 1). In all MARCATS, the uncertainty 484 associated to the  $pCO_2$  field is the largest and the global  $\sigma_{pCO_2}$  amounts to 0.017 Pg C yr<sup>-1</sup> for coastal 485 regions. A similar pattern is found in the open ocean, where  $\sigma_{pCO2}$  is the main source of uncertainty 486 with a global value of 0.23 Pg C yr<sup>-1</sup> to be compared with the total uncertainty of 0.26 Pg C yr<sup>-1</sup> for the 487 open ocean (Table 1). Both in coastal seas and the open ocean, the choice of the  $k_{660}$ -formulation ( $\sigma_k$ ) 488 yields a relative uncertainty of 7 %. The uncertainty associated to wind speed ( $\sigma_{wind}$ ) however reveals a 489 more complex spatial pattern. On the coastal seas,  $\sigma_{wind}$  is low when integrated globally (0,003 Pg C yr 490 <sup>1</sup>, corresponding to 4 % difference between  $FCO_2$  obtained using the three wind products) but can be as 491 large as 16 % and 12 % in marginal seas and in the Indian basin, respectively. In agreement with the 492 study of Roobaert et al. (2018), the effect associated with the choice of the wind product is more 493 important for the open ocean than for coastal seas with an uncertainty of 12 % globally ( $\sigma_{wind}$  value of 494 0.090 Pg C yr<sup>-1</sup>) with the largest uncertainties in the Pacific basin and in the Southern ocean. Because of 495 the temporal heterogeneity of the data coverage in coastal regions, the resulting  $FCO_2$  uncertainty also 496 varies along the year and not only spatially (Figure 5). In all latitudinal bands, 75 % of the monthly 497 uncertainty is below 0.25 mol m<sup>-2</sup> yr<sup>-1</sup> with the exception of the band between 40° S and 60° S as a 498 consequence of a much lower  $pCO_2$  observational data coverage. Even in this last region, the amplitude 499 of the seasonal uncertainty does not exceed that of  $FCO_2$ . 500

501

502 4 Discussion

### 503 4.1 Annual mean fluxes

The global annual mean coastal CO<sub>2</sub> sink of -195 Tg C yr<sup>-1</sup> (-0.20 Pg C yr<sup>-1</sup>) generated by our 504 calculation falls at the low end of the range of past estimates spanning from -0.45 Pg C yr<sup>-1</sup> to -0.21 Pg 505 C yr<sup>-1</sup> (Borges, 2005; Borges et al., 2005; Cai et al., 2006; Chen et al., 2013; Chen & Borges, 2009; Dai 506 et al., 2013; Laruelle et al., 2010). This confirms that previous estimates based on sparse datasets likely 507 overestimated the uptake of CO<sub>2</sub> by the coastal seas because of the inherent biases associated to the 508 extrapolation of a limited number of local estimates to large scales and an over-representation of 509 temperate and sub-polar systems, which are mostly CO<sub>2</sub> sinks. Support for this interpretation comes 510 from the fact that our estimate agrees very well with that of Laruelle et al. (2014) (-192 Tg C yr<sup>-1</sup>), who 511 upscaled 3 million local measurements using different regional integration methods depending on the 512 data density, also accounting for differences in the coastal seas delimitation and taking into 513 consideration the sea-ice cover in the FCO<sub>2</sub> calculations. Our study improves upon that of Laruelle et al. 514 (2014) thanks to our continuous spatial and temporal coverage of  $FCO_2$  estimates for all the coastal seas 515 at 0.25° resolution as opposed to Laruelle et al. (2014) who calculated 150 regional FCO2 estimates 516 using the COSCAT/MARCATS segmentation (Laruelle et al., 2013). Taken that the work of Laruelle et 517 al. (2014) is the most comprehensive, spatially resolved study to date and relies on the same spatial 518 519 definition of the coastal regions as the one used in our study, we perform a comparison at the level of each MARCATS (Figure S2a and Table S1 in the supplementary information). Furthermore, the general 520 spatial trends observed in our work and in Laruelle et al. (2014) such as the latitudinal profile of  $FCO_2$ 521 are also consistent with several earlier studies (Borges et al., 2005; Cai, 2011; Chen et al., 2013; Chen & 522 Borges, 2009; Laruelle et al., 2010). The role of high latitude shelves as efficient  $CO_2$  sinks as a 523 consequence of their low water temperatures has also been evidenced by numerous studies on the Arctic 524 525 ocean (Bates & Mathis, 2009). We do not emphasize the comparison for MARCATS 4 (Peruvian upwelling), 22 (Moroccan upwelling) and 27 (West Arabian Sea) due to the low performance of the 526 527 neural network technique in these regions (see sections 3.1 and 3.2). In general, the absolute magnitude of sources and sinks are slightly lower in our calculations than in Laruelle et al. (2014), but both studies 528 are in relatively good agreement overall. All MARCATS in polar and subpolar margins act as a CO<sub>2</sub> 529

sink for both studies. In tropical margins, EBCs and WBCs, the sign (a sink or a source) of FCO<sub>2</sub> is also 530 in agreement except for one or two MARCATS in each category (i.e., MARCATS 23, 38, 6 and 19). 531 Along the Indian margins, i.e., regions where  $FCO_2$  values calculated by Laruelle et al. (2014) have a 532 low degree of confidence, our results show FCO<sub>2</sub> values with opposite signs for all of the MARCATS 533 with the exception of the East Arabian Sea (MARCATS 30). In addition, out of the 9 MARCATS 534 located in marginal seas, only two display  $FCO_2$  with the same sign. In quantitative terms, a difference 535 of less than 1 mol C m<sup>-2</sup> yr<sup>-1</sup> is observed between the two studies in all MARCATS except for two 536 regions (MARCATS 40 and 41) contributing to merely 4 % of the total surface area of the coastal 537 538 regions. These two regions have a low degree of confidence in the study of Laruelle et al. (2014).

We also compare our results to those of Bourgeois et al. (2016) which were calculated using the 539 Oceanic General Circulation Model (OGCM) NEMO-PISCES (Nucleus for European Modelling of the 540 Ocean - Pelagic Interaction Scheme for Carbon and Ecosystem Studies). Overall, the two studies are in 541 relatively good agreement regarding the FCO<sub>2</sub> sign (33 over 45 MARCATS), but FCO<sub>2</sub> values 542 modelled by Bourgeois et al. (2016) produce more intense CO<sub>2</sub> sinks and weaker sources (Figure S2b 543 and Table S1). Out of the 45 MARCATS, only 13 present differences exceeding 1 mol C m<sup>-2</sup> yr<sup>-1</sup>, 544 corresponding to a global coastal surface area of  $\sim 30$  %. These differences are mainly located on 545 546 narrow shelves, which are poorly resolved by OGCMs (Bourgeois et al., 2016) and in descending order, are located in marginal seas, EBCs and WBCs, and subpolar and polar regions. A difference greater 547 than 2 mol C m<sup>-2</sup> yr<sup>-1</sup> is observed only in the Black Sea. 548

#### 549 $4.2 \text{ CO}_2$ flux densities

Both in the open and coastal ocean, several processes and their interactions govern the direction and intensity of the  $CO_2$  exchange with the atmosphere. These processes range from physical (i.e., solubility, mixing and upwelling waters) to biological (i.e. primary production). Moreover, the supply of carbon and nutrients by rivers from continents may also affect the dynamics of the air-sea exchange in coastal regions. Due to the comparatively high photosynthetic carbon fixation and biological activity that occur in coastal regions in comparison to the rest of the ocean, past observational (Borges et al.,

2005; Cai, 2011; Chen et al., 2013; Dai et al., 2013; Laruelle et al., 2010, 2014, 2018) and modelling 556 (Andersson & Mackenzie, 2004; Mackenzie et al., 2012) studies suggest that the FCO<sub>2</sub> dynamics 557 substantially differ between the coastal regions and the adjacent open ocean basins. In particular, past 558 studies have estimated that the global mean air-sea CO2 exchange rates for coastal seas vary between -559 0.7 mol C m<sup>-2</sup> yr<sup>-1</sup> and -1.2 mol C m<sup>-2</sup> yr<sup>-1</sup> implying that, globally, they act as a more efficient CO<sub>2</sub> sink 560 than the open ocean per unit surface area (i.e., -0.36 mol C m<sup>-2</sup> yr<sup>-1</sup> in Takahashi et al., 2009). In this 561 study, we observe a difference of 36 % between the global mean  $FCO_2$  for the coastal seas (-0.58 mol C 562  $m^{-2} yr^{-1}$ ) and that for the open ocean (-0.37 mol C  $m^{-2} yr^{-1}$ ). However, our analysis of the FCO<sub>2</sub> maps 563 (Figure 2) as well as their latitudinal profiles (Figure 4) reveal that there is little difference between 564 these two systems when compared by latitude. The  $FCO_2$  distributions for coastal and the open ocean 565 are relatively similar with CO<sub>2</sub> sinks at temperate and high latitudes and sources or values close to 566 equilibrium along the Tropics and the Equator. This general latitudinal trend for coastal regions which 567 568 has been suggested by previous studies (i.e., Borges et al., 2005; Cai, 2011; Chen et al., 2013; Laruelle et al., 2010, 2014) has now become very clear. 569

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The main difference between the coastal and the open ocean consists of the width of the latitudinal band 571 where the open ocean acts as a source (Figure 4), which is narrower (15° N - 15° S) than that of the 572 coastal seas (30° N - 20° S). Our results also reveal larger differences between the two systems locally, 573 in particular in river-dominated coastal regions. The impact of riverine inputs on the coastal CO<sub>2</sub> flux, 574 for instance, was investigated by Lefèvre et al. (2010) for the Amazon plume, which acts as a CO<sub>2</sub> sink 575 while the adjacent ocean is a source . Taken this remarkable similarity in CO<sub>2</sub> flux densities, our results 576 thus suggest that most of the difference in the global mean  $FCO_2$  between coastal and the open ocean 577 stems from the differing areal distribution of the coastal regions, which are much more abundant in the 578 high latitude of the Northern Hemisphere where intense CO<sub>2</sub> sinks take place. Indeed, the coastal seas 579 located in areas >  $60^{\circ}$  N (25 % of the total surface coastal area against only 1 % for the open ocean) 580 take up more than one-third of the global coastal uptake flux (-98 Tg C yr<sup>-1</sup>). This disproportionate 581 contribution to the global coastal CO<sub>2</sub> sink is consistent with earlier works (Cai, 2011; Laruelle et al., 582

2010, 2014). Excluding regions located above  $60^{\circ}$  N, the global mean  $FCO_2$  obtained for coastal seas is 583 -0.39 mol C m<sup>-2</sup> yr<sup>-1</sup>, remarkably close to -0.35 mol C m<sup>-2</sup> yr<sup>-1</sup> for the open ocean. This similarity 584 between coastal regions and the open ocean was also suggested by the work of Bourgeois et al. (2016) 585 in their modeling study as well as by Wanninkhof et al. (2013). In the latter, the integrated coastal FCO2 586 estimate (-0.18 Pg C yr<sup>-1</sup>) was calculated by extrapolation assuming that the FCO<sub>2</sub> of the coastal seas 587 per unit area is the same as that of the adjacent ocean. With the exception of the local discrepancies 588 mentioned above (river plume, estuary mouth, ...), the surprisingly similar latitudinal distribution of 589 flux densities between the coast and the adjacent open ocean observed in this study suggests that, in 590 591 addition to others factor explaining the spatial heterogeneity, the thermal effect is the main driver that governs the zonal latitudinal pattern of sources and sinks for the ocean as a whole. This effect translates 592 into a change in  $CO_2$  solubility via the term  $K_0$  in equation 1. Cooling/warming waters in high/low 593 latitudes induce an increase/decrease in the  $CO_2$  solubility and, thus, a lower/higher  $pCO_2$  in the surface 594 595 seawater. The dominance of this process that was suggested by Gruber (2015) when commenting on the results of Laruelle et al. (2014) can now be quantified thanks to our analysis. 596

## 597 4.3 Global Ocean CO<sub>2</sub> budget

Globally, our net ocean carbon uptake of  $-1.7 \pm 0.3$  Pg C yr<sup>-1</sup> (coastal and open ocean) represents the 598 sum of a net uptake of anthropogenic CO<sub>2</sub>, a net exchange flux of natural carbon in response to climate 599 variability and change, plus an outgassing of river-derived carbon. If we assume a pre-industrial river 600 outgassing rate of  $0.45 \pm 0.18$  Pg C yr<sup>-1</sup> (Jacobson et al., 2007) and an outgassing flux of natural CO<sub>2</sub> of 601 the order of  $0.4 \pm 0.2$  Pg C yr<sup>-1</sup> (Gruber et al., 2019), our estimates implies an anthropogenic CO<sub>2</sub> 602 uptake rate of  $-2.6 \pm 0.4$  Pg C yr<sup>-1</sup> for the 1998 through 2015 period. This uptake estimate is statistically 603 identical to that recently estimated by Gruber et al. (2019) (-2.6  $\pm$  0.3 Pg C yr<sup>-1</sup>) on the basis of ocean 604 interior changes in anthropogenic CO<sub>2</sub> between 1994 and 2007. It is also consistent with other 605 independent estimates, such as those based on an Green's function ocean inversion method (-2.4 Pg C 606 yr<sup>-1</sup>; Gruber et al., 2009, adjusted to the year 2007), those based on an inverse model (-2.6 Pg C yr<sup>-1</sup>; 607 DeVries, 2014), or those based on a compilation of models and observations (-2.0  $\pm$  0.6 Pg C yr<sup>-1</sup>, 608

Wanninkhof et al. 2013). The anthropogenic CO<sub>2</sub> uptake estimated here would reaches  $-2.9 \pm 0.5$  Pg C 609 vr<sup>-1</sup> when considering the revisited global pre-industrial river carbon flux (0.78  $\pm$  0.41 Pg C yr<sup>-1</sup>) 610 proposed by Resplandy et al. (2018). While the magnitude of global ocean carbon uptake estimates have 611 not changed much in recent years, the uncertainty of our estimate as well as that for the open ocean has 612 decreased over time. While Laruelle et al. (2014) reported an uncertainty of 0.05 Pg C yr<sup>-1</sup> for the global 613 coastal ocean, our estimate is now 0.02 Pg C yr<sup>-1</sup>. Similarly, the uncertainty estimate for the global open 614 ocean of 0.26 Pg C yr<sup>-1</sup> is halved relative to the initial estimate by Landschützer et al. (2014) of 0.53 Pg 615 C yr<sup>-1</sup>. This reduction is the result of (1) improved data coverage and (2) improved analysis methods. 616 Note that the influence of regional biases is not taken into account in the calculation of the  $FCO_2$ 617 uncertainty in this study. However, considering that the globally averaged bias is null and that most 618 regional biases are much lower than  $\theta_{obs}$  (Laruelle et al., 2017), including them into the uncertainty 619 calculation would likely only marginally increase it. 620

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Our analysis reveals that, both for the coastal regions and the open ocean, most of the uncertainty over 622 the  $FCO_2$  comes from uncertainties associated to the  $pCO_2$  products themselves. This can be attributed 623 to the existence of large areas that remain undersampled in spite of the international community effort 624 to integrate more quality controlled data in the yearly releases of databases such as SOCAT (Bakker et 625 al., 2016). Although most of the world's ocean is now relatively well monitored and simulated by the 626 SOM-FFN derived data products of Landschützer et al. (2014) or Laruelle et al. (2017), some of the 627 least sampled regions represent a challenge even for the best interpolation algorithm because their 628 training relies on too small datasets which do not allow fully capturing the biogeochemical dynamics of 629 the region. This likely explains, for example, the poor representation of the Peruvian upwelling by the 630 coastal  $pCO_2$  product of Laruelle et al. (2017), which was thus removed from our regional analysis. 631 Finally, another potential limitation of our calculations is the use of the Atmospheric Boundary Layer to 632 generate the atmospheric  $pCO_2$  field used to calculate the air-water  $pCO_2$  gradient. Recent local studies 633 634 of coastal environment have suggested that, because of their proximity with continents where anthropogenic sources of CO<sub>2</sub> are taking place, coastal region might be exposed to slightly higher 635

atmospheric  $pCO_2$  than the oceanic average. In order to quantify the potential effect of such bias, our 636 calculations for the global coastal  $CO_2$  sink were also performed with an atmospheric  $pCO_2$  increased 637 by 2 µatm in the 23° N - 66° N latitudinal band (where most of the Earth landmasses are located), by 1 638 µatm in the rest of the Northern Hemisphere and by 0.5 µatm in the Southern Hemisphere. The resulting 639 global CO<sub>2</sub> uptake from coastal regions amounts to -207 Tg C yr<sup>-1</sup>, a 6 % increase in magnitude 640 compared to our initial estimate. This relatively small difference (falling within our uncertainty range) 641 might be better accounted for in future calculations with the use of spatially resolved dry air mixing 642 ratio of CO<sub>2</sub> dataset such as NASA's Goddard Earth Observing System (GEOS) model, or 643 644 CarbonTracker.

#### 645 4.4 Seasonal variability

The analysis of the relative contributions of the different drivers to the  $FCO_2$  seasonal variability reveals 646 that, for most regions, the seasonal cycle of  $FCO_2$  is intimately linked to that of  $\Delta pCO_2$  (Figures 7a and 647 8). Although the contribution of  $\Delta pCO_2$  is the main driving force of the seasonal variability of  $FCO_2$  in 648 the different latitudinal bands ( $\beta_{\Delta pCO_2} > 0$ ), the contribution of the sea-ice cover and wind speed to the 649 seasonal FCO2 variability can be large locally, in particular at temperate and high latitudes (Figures 7 650 and 8). This observation was already reported by Yasunaka et al. (2016) for the Greenland, Barents and 651 Chuchki Seas. In the Northern Hemisphere where most of the coastal seas act as a CO<sub>2</sub> sink, the 652 positive  $\beta_{sea-ice}$  values can be explained by low summer  $pCO_2$  combined with low sea-ice cover pushing 653  $FCO_2$  toward a stronger sink while in winter, when  $pCO_2$  is high, the sea-ice cover limits this exchange 654 and induces a  $FCO_2$  going towards a weaker sink. The effect of the wind tends to counteract the other 655 two mechanisms ( $\beta_{wind} < 0$ ). In winter, the wind speed is faster compared to summer and the turbulence 656 at the air-sea interface is increased accordingly. This effect favors the CO<sub>2</sub> exchange (stronger sink) in 657 winter while in summer the wind speed slows down and thereby limits the exchange (weaker sink). The 658 opposite effect of the wind speed with respect to sea-ice and  $\Delta pCO_2$  has already been observed for the 659 Barents Sea by Nakaoka et al. (2006). 660

All in all, a very strong seasonal variability is observed at higher latitudes, a smaller one between 40° S 661 and 40° N and a very low one between 10° N and 10° S. The global  $FCO_2$  seasonal profile for all of the 662 coastal seas mostly follows that observed for high latitudes (bottom panel of Figure 8). As was the case 663 for the CO<sub>2</sub> flux density analysis, this trend reflects that the coastal seas located at high latitudes 664 influence greatly the global seasonal  $FCO_2$  profile as a result of their disproportionate contribution to 665 the global uptake. Because  $\Delta pCO_2$  is the major driver for all latitudinal bands and due to the six-months 666 shift in the phase of the seasonal profile between low and high latitudes, the amplitude of the global 667  $FCO_2$  seasonal cycle is dampened and is of the same order of magnitude as that of the equatorial 668 regions. As a result of the uneven areal surface distribution of the coastal seas at high latitudes in the 669 Northern Hemisphere and their large contribution to the global seasonal profile, the more intense CO<sub>2</sub> 670 uptake is encountered in summer globally while this is observed in winter in the open ocean 671 (Landschützer et al., 2018). 672

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#### 674 **5** Concluding remarks and directions for future research

Up until now, the dynamics of the CO<sub>2</sub> exchange at the air-sea interface in coastal regions remained poorly understood and its quantification stayed tainted with large uncertainties. With a better spatial and temporal coverage of observational data, our study provides a better constrained coastal  $FCO_2$  globally and regionally using the MARCATS segmentation. We produce, for the first time, monthly  $FCO_2$  maps for the coastal sea regions worldwide at a high spatial resolution of 0.25 degree using the recent coastal  $pCO_2$  product of Laruelle et al. (2017) derived from a two-step artificial neural network for the 1998-2015 period.

Although this study allows us to analyze the climatological  $FCO_2$  seasonality and to quantify the respective contributions of  $\Delta pCO_2$ , wind speed and sea-ice cover to the seasonal signal, the roles of processes indirectly affecting the seasonality of  $FCO_2$  through their effect on  $pCO_2$  in surface coastal waters remains largely unknown. For instance, changes in *SST* and *SSS* induce variations in  $pCO_2$  by

physico-chemical processes that impact the CO<sub>2</sub> exchange. Similarly, variations in alkalinity (ALK) and 686 dissolved inorganic carbon (DIC) governed by biogeochemical and physical processes can also control 687 the seasonality of  $pCO_2$ . In the study of Laruelle et al. (2017), the effect on  $pCO_2$  seasonality associated 688 with changes in SST was quantified and compared to non-thermal effects for different oceanic basins 689 690 and at global scale. This study emphasizes that the solubility pump is the dominant process in the seasonality of  $pCO_2$  for all latitudinal bands in spite of differences between oceanic basins, a result 691 consistent with past studies (i.e., Laruelle et al., 2014; Shadwick et al., 2010). However, their study also 692 highlights that the thermal component alone cannot explain the entirety of the  $pCO_2$  seasonal cycle and 693 694 that non-thermal processes (i.e., biological uptake, mixing...) also contribute significantly to the seasonal variation of  $pCO_2$ , especially in temperate regions. At high latitudes > 60° in both 695 hemispheres, high/low pCO<sub>2</sub> values are observed during winter/summer in this study confirming that 696 the solubility pump is not the dominant process governing the  $pCO_2$  seasonal variability in these 697 698 regions.

The use of ocean biogeochemical models allows one to further disentangle the respective effects of 699 ALK, DIC, SSS and SST on the seasonal variability of  $pCO_2$  and to identify the contributions of 700 physical, biological and mixing processes to variations in the  $pCO_2$  field. Such studies have been 701 conducted along the California current along the west coast of the United States (Turi et al., 2014) as 702 well as in four different shelf seas (South and Southeast Brazilian shelves, Uruguayan and Patagonia 703 shelves) along the southwestern Atlantic ocean (Arruda et al., 2015). Both studies concluded that SST 704 (the solubility pump) and DIC associated to water circulation and the biological pump are the main 705 drivers of seasonality but that their effects on  $pCO_2$  changes can counteract one another. In the 706 California current, the water circulation generated by the strong upwelling is the main control of the 707  $pCO_2$  seasonality, which is further influenced by the solubility and biological pumps. In the study of 708 Arruda et al. (2015) on the Patagonia shelf, the opposite is observed and the solubility and biological 709 pumps govern the seasonality of  $pCO_2$  with only a small contribution from the water circulation. These 710 711 different contributions to the  $pCO_2$  seasonal variability remain however unknown at the global scale as

in coastal seas not under upwelling influence. Although they can only be estimated through Ocean 712 Biogeochemical models, our data based coastal CO<sub>2</sub> climatology at high spatial and temporal resolution 713 provide the necessary observational constraints against which these models can be evaluated. A 714 complete attribution analysis of the  $FCO_2$  seasonality on the global coastal regions is thus a goal that 715 should be reachable in the near future. Moreover, long-term trends and the interannual variability of 716 global coastal CO<sub>2</sub> fluxes still remain poorly constrained (Laruelle et al., 2018). Coastal regions are 717 subject to large interannual variations (IAV) driven by riverine loading changes or climatic events such 718 as El Nino cycle. The analysis of the coastal  $FCO_2$  IAV, its environmental controls and how it may 719 720 impact seasonal cycle should be investigated in future research. Some attention should also be paid to the potential effects of changes in others environmental parameters (i.e., expected changes in wind 721 722 speed and pattern, decrease of the sea-ice cover or increase in atmospheric  $pCO_2$ ) on the spatiotemporal  $FCO_2$  trend. Such investigations have already been carried out for the open ocean but are still 723 724 missing in coastal seas at global scale. For instance, the increase in atmospheric  $pCO_2$  in addition to increasing surface water  $pCO_2$ , tends to enhance the amplitude of the seasonal signal in  $FCO_2$ 725 (Landschützer et al., 2018). The study of Wanninkhof & Trinanes (2017) also suggests that the 726 observed increase in wind speed does not affect the air-sea exchange homogeneously in the ocean 727 728 worldwide and depends on the patterns and the localization of this wind speed changes. One could thus speculate that similar effect will be observed in the coastal ocean but future research is still needed to 729 properly decipher to complex dynamics of  $FCO_2$  in the coastal ocean and should be included in ongoing 730 investigations of the global carbon budget. Our study provides the necessary ground work for such 731 future research. In parallel, the ongoing community effort to better represent the least monitored coastal 732 regions in SOCAT will further help reducing the uncertainties associated to the  $pCO_2$  field and a better 733 representation of the spatial patterns of atmospheric  $pCO_2$  will contribute to even better constrained 734  $FCO_2$  estimates as well. 735

736 Acknowledgments, Samples, and Data

The observation-based global monthly gridded sea-surface  $pCO_2$  products are provided for the coastal seas by Laruelle et al. (2017) and for the open ocean by Landschützer et al. (2017). They are accessible

on the Biogeosciences journal website (https://doi.org/10.5194/bg-14-4545-2017) and on the NOAA 739 (https://www.nodc.noaa.gov/ocads/oceans/SPCO2 1982 2015 ETH SOM FFN.html) website 740 respectively. The atmospheric partial pressure of CO<sub>2</sub> is provided by the NOAA Marine Boundary layer 741 and can be found on their website (https://www.esrl.noaa.gov/gmd/ccgg/mbl/). The 6 hours 0.25 degree 742 global atmospheric reanalysis ERA-Interim wind product dataset (Dee et al., 2011) can be downloaded 743 on the European Centre for Medium-Range Weather Forecasts (ECMWF) website. The NOAA High-744 resolution sea surface temperature (Reynolds et al., 2007) and sea-ice cover (Cavalieri et al., 1996; 745 Grumbine, 1996; Reynolds et al., 2007) data are provided by the NOAA/OAR/ESRL PSD, Boulder, 746 Colorado, USA, from their website at https://www.esrl.noaa.gov/psd/. The daily EN4 sea surface 747 salinity (Good et al., 2013) can be found on the Met Office Hadley center website. The authors declare 748 that they have no conflict of interest. AR is funded by a teaching assistantship grant awarded by the 749 Université Libre de Bruxelles (ULB). GL has been supported by Labex L-IPSL LP3 SP3, which is 750 funded by ANR (Grant #ANR-10-LABX-0018) and is now research associate of the F.R.S.-FNRS at the 751 ULB. PR received funding from the VERIFY project from the European Union's Horizon 2020 752 753 research and innovation program under grant agreement No. 776810. PL is supported by the Max Planck Society for the Advancement of Science. NG acknowledges support from ETH Zürich and the 754 755 Swiss National Science Foundation through the XEBUS project (200020 175787). The authors thank Adam Hastie for his technical support with the GIS-based coastal seas MARCATS delimitation used in 756 757 this study.

#### 758 **References**

- Andersson, A. J., & Mackenzie, F. T. (2004). Shallow-water oceans: a source or sink of atmospheric CO<sub>2</sub>? *Frontiers in Ecology and the Environment*, 2(7), 348–353.
- Arruda, R., Calil, P. H. R., Bianchi, A. A., Doney, S. C., Gruber, N., Lima, I., & Turi, G. (2015). Air-sea CO<sub>2</sub> fluxes and the controls on ocean surface pCO<sub>2</sub> seasonal variability in the coastal and open-ocean southwestern Atlantic Ocean: a modeling study. *Biogeosciences*, *12*, 5793–5809. https://doi.org/10.5194/bg-12-5793-2015
- Atlas, R., Hoffman, R. N., Ardizzone, J., Leidner, S. M., Jusem, J. C., Smith, D. K., & Gombos, D. (2011). A cross calibrated, multiplatform ocean surface wind velocity product for meteorological and oceanographic applications.
   *Bulletin of the American Meteorological Society*, 92(2), 157–174. https://doi.org/10.1175/2010BAMS2946.1
- Bakker, D. C. E., Pfeil, B., Landa, C. S., Metzl, N., O'Brien, K. M., Olsen, A., et al. (2016). A multi-decade record of high quality fCO<sub>2</sub> data in version 3 of the Surface Ocean CO<sub>2</sub> Atlas (SOCAT). *Search.Proquest.Com*, 8, 383–413.
   https://doi.org/doi:10.5194/essd-8-383-2016
- Bates, N. R., & Mathis, J. T. (2009). The Arctic Ocean marine carbon cycle: Evaluation of air-sea CO<sub>2</sub> exchanges, ocean
   acidification impacts and potential feedbacks. *Biogeosciences*, 6(11), 2433–2459. https://doi.org/10.5194/bg-6-2433 2009
- Bauer, J. E., Cai, W., Raymond, P. A., Bianchi, T. S., Hopkinson, C. S., & Regnier, P. A. (2013). The changing carbon cycle
   of the coastal ocean. *Nature*, 504(7478), 61. https://doi.org/10.1038/nature12857
- Borges, A. V. (2005). Do we have enough pieces of the jigsaw to integrate CO<sub>2</sub> fluxes in the coastal ocean? *Estuaries*, 28(1),
   3–27. https://doi.org/10.1007/BF02732750
- Borges, A. V., Delille, B., & Frankignoulle, M. (2005). Budgeting sinks and sources of CO<sub>2</sub> in the coastal ocean: Diversity
   of ecosystem counts. *Geophysical Research Letters*, *32*(14), 1–4. https://doi.org/10.1029/2005GL023053
- Bourgeois, T., Orr, J. C., Resplandy, L., Terhaar, J., Ethé, C., Gehlen, M., & Bopp, L. (2016). Coastal-ocean uptake of
   anthropogenic carbon. *Biogeosciences*, 13(14), 4167–4185. https://doi.org/10.5194/bg-13-4167-2016

- Cai, W. (2011). Estuarine and Coastal Ocean Carbon Paradox: CO<sub>2</sub> Sinks or Sites of Terrestrial Carbon Incineration? *Annual Review of Marine Science*, 3(1), 123–145. https://doi.org/10.1146/annurev-marine-120709-142723
- Cai, W., Dai, M., & Wang, Y. (2006). Air-sea exchange of carbon dioxide in ocean margins: A province-based synthesis.
   *Geophysical Research Letters*, 33(12), 2–5. https://doi.org/10.1029/2006GL026219
- Cavalieri, D., Parkinson, C., Gloersen, P., & Zwally, H. J. (1996). Sea ice concentrations from Nimbus-7 SMMR and DMSP
   SSM/I passive microwave data. National Snow and Ice Data Center, Boulder, Colorado. Digital media.
- Chen, C. T. A., & Borges, A. V. (2009). Reconciling opposing views on carbon cycling in the coastal ocean: Continental
   shelves as sinks and near-shore ecosystems as sources of atmospheric CO<sub>2</sub>. *Deep Sea Research Part II: Topical Studies in Oceanography*, 56(8–10), 578–590. https://doi.org/10.1016/j.dsr2.2009.01.001
- Chen, C. T. A., Huang, T. H., Chen, Y. C., Bai, Y., He, X., & Kang, Y. (2013). Air-sea exchanges of CO<sub>2</sub> in the world's coastal seas. *Biogeosciences*, 10(10), 6509–6544. https://doi.org/10.5194/bg-10-6509-2013
- Couldrey, M. P., Oliver, K. I. C., Yool, A., Halloran, P. R., & Achterberg, E. P. (2016). On which timescales do gas transfer
   velocities control North Atlantic CO<sub>2</sub> flux variability? *Global Biogeochemical Cycles*, *30*(5), 787–802.
   https://doi.org/10.1002/2015GB005267
- Dai, M., Cao, Z., Guo, X., Zhai, W., Liu, Z., Yin, Z., et al. (2013). Why are some marginal seas sources of atmospheric CO<sub>2</sub>?
   *Geophysical Research Letters*, 40(10), 2154–2158. https://doi.org/10.1002/grl.50390
- Deacon, E. L. (1977). Gas transfer to and across an air-water interface. *Tellus*, 29(4), 363–374.
   https://doi.org/10.1111/j.2153-3490.1977.tb00724.x
- Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., et al. (2011). The ERA-Interim reanalysis:
   Configuration and performance of the data assimilation system. *Quarterly Journal of the Royal Meteorological Society*, 137(656), 553–597. https://doi.org/10.1002/qj.828
- BeVries, T. (2014). The oceanic anthropogenic CO<sub>2</sub> sink: Storage, air-sea fluxes, and transports over the industrial era.
   *Global Biogeochemical Cycles*, 28(7), 631–647. https://doi.org/10.1002/2013GB004739
- Boney, S. C., Lima, I., Feely, R. A., Glover, D. M., Lindsay, K., Mahowald, N., et al. (2009). Mechanisms governing
   interannual variability in upper-ocean inorganic carbon system and air–sea CO<sub>2</sub> fluxes: Physical climate and
   atmospheric dust. *Deep Sea Research Part II: Topical Studies in Oceanography*, 56(8–10), 640–655.
   https://doi.org/10.1016/j.dsr2.2008.12.006
- Etcheto, J., & Merlivat, L. (1988). Satellite determination of the carbon dioxide exchange coefficient at the oceanatmosphere interface: A first step. *Journal of Geophysical Research: Oceans*, 93(C12), 15669.
   https://doi.org/10.1029/JC093iC12p15669
- Frankignoulle, M., & Borges, A. V. (2001). European continental shelf as a significant sink for atmospheric carbon dioxide.
   *Global Biogeochemical Cycles*, 15(3), 569–576.
- Friederich, G. E., Ledesma, J., Ulloa, O., & Chavez, F. P. (2008). Air-sea carbon dioxide fluxes in the coastal southeastern
  tropical Pacific. *Progress in Oceanography*, 79(2–4), 156–166. https://doi.org/10.1016/j.pocean.2008.10.001
- Good, S. A., Martin, M. J., & Rayner, N. A. (2013). EN4: Quality controlled ocean temperature and salinity profiles and
  monthly objective analyses with uncertainty estimates. *Journal of Geophysical Research: Oceans*, 118(12), 6704–
  6716. https://doi.org/10.1002/2013JC009067
- 818 Gruber, N. (2015). Ocean biogeochemistry: Carbon at the coastal interface. *Nature*. https://doi.org/10.1038/nature14082
- Gruber, N., Gloor, M., Mikaloff Fletcher, S. E., Doney, S. C., Dutkiewicz, S., Follows, M. J., et al. (2009). Oceanic sources,
  sinks, and transport of atmospheric CO<sub>2</sub>. *Global Biogeochemical Cycles*, 23(1), 1–21.
  https://doi.org/10.1029/2008GB003349
- Gruber, N., Clement, D., Carter, B. R., Feely, R. A., Van Heuven, S., Hoppema, M., et al. (2019). The oceanic sink for
   anthropogenic CO<sub>2</sub> from 1994 to 2007. *Science*, *363*(6432), 1193–1199. https://doi.org/10.1126/science.aau5153
- Grumbine, R. W. (1996). Automated passive microwave sea ice concentration analysis at NCEP. NOAA Tech. Note, 120, 1–
   13.
- Ho, D. T., Wanninkhof, R., Schlosser, P., Ullman, D. S., Hebert, D., & Sullivan, K. F. (2011). Toward a universal
  relationship between wind speed and gas exchange: Gas transfer velocities measured with 3He/SF6 during the
  Southern Ocean Gas Exchange Experiment. *Journal of Geophysical Research: Oceans*, *116*(7), C00F04.

829 https://doi.org/10.1029/2010JC006854

- Jacobs, C. M. J., Kohsiek, W., & Oost, W. A. (1999). Air-sea fluxes and transfer velocity of CO<sub>2</sub> over the north sea: Results
   from ASGAMAGE. *Tellus, Series B: Chemical and Physical Meteorology*, *51*(3), 629–641.
- 832 https://doi.org/10.1034/j.1600-0889.1999.t01-2-00005.x
- Jacobson, A. R., Fletcher, S. E. M., Gruber, N., Sarmiento, J. L., & Gloor, M. (2007). A joint atmosphere-ocean inversion
   for surface fluxes of carbon dioxide: 1. Methods and global-scale fluxes. *Global Biogeochemical Cycles*, 21(1).
   https://doi.org/10.1029/2005GB002556
- Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., et al. (1996). The NCEP/NCAR 40-year
   reanalysis project. *Bulletin of the American Meteorological Society*, 77(3), 437–471. https://doi.org/10.1175/1520 0477(1996)077<0437:TNYRP>2.0.CO;2
- Kanamitsu, M., Ebisuzaki, W., Woollen, J., Yang, S. K., Hnilo, J. J., Fiorino, M., & Potter, G. L. (2002). NCEP-DOE
   AMIP-II reanalysis (R-2). *Bulletin of the American Meteorological Society*, 83(11), 1631-1643+1559.
   https://doi.org/10.1175/BAMS-83-11-1631
- Keeling, R. F., & Manning, A. C. (2014). Studies of Recent Changes in Atmospheric O<sub>2</sub> Content. In *Treatise on Geochemistry (Second Edition)* (pp. 385–404). https://doi.org/http://dx.doi.org/10.1016/B978-0-08-095975-7.00420-4
- Kuss, J., Nagel, K., & Schneider, B. (2004). Evidence from the Baltic Sea for an enhanced CO<sub>2</sub> air-sea transfer velocity.
   *Tellus, Series B: Chemical and Physical Meteorology*, 56(2), 175–182. https://doi.org/10.1111/j.1600 0889.2004.00092.x
- Landschützer, P., Gruber, N., Bakker, D. C. E., Schuster, U., Nakaoka, S., Payne, M. R., et al. (2013). A neural networkbased estimate of the seasonal to inter-annual variability of the Atlantic Ocean carbon sink. *Biogeosciences*, *10*(11),
  7793–7815. https://doi.org/10.5194/bg-10-7793-2013
- Landschützer, P., Gruber, N., Bakker, D. C. E., & Schuster, U. (2014). Recent variability of the global ocean carbon sink.
   *Global Biogeochemical Cycles*, 28(9), 927–949. https://doi.org/10.1002/2014GB004853
- Landschützer, P., Gruber, N., & Bakker, D. C. E. (2016). Decadal variations and trends of the global ocean carbon sink.
   *Global Biogeochemical Cycles*, *30*(10), 1396–1417. https://doi.org/10.1002/2015GB005359
- Landschützer, P., Gruber, N., & Bakker, D. C. E. (2017). An updated observation-based global monthly gridded sea surface
   pCO<sub>2</sub> and air-sea CO<sub>2</sub> flux product from 1982 through 2015 and its monthly climatology (NCEI Accession 0160558).
   Version 2.2. NOAA National Centers for Environmental Information. Dataset.
- Landschützer, P., Gruber, N., Bakker, D. C. E., Stemmler, I., & Six, K. D. (2018). Strengthening seasonal marine CO<sub>2</sub>
   variations due to increasing atmospheric CO<sub>2</sub>. *Nature Climate Change*. https://doi.org/10.1038/s41558-017-0057-x
- Laruelle, G. G., Dürr, H. H., Slomp, C. P., & Borges, A. V. (2010). Evaluation of sinks and sources of CO<sub>2</sub> in the global
   coastal ocean using a spatially-explicit typology of estuaries and continental shelves. *Geophysical Research Letters*,
   37(15), 1–6. https://doi.org/10.1029/2010GL043691
- Laruelle, G. G., Dürr, H. H., Lauerwald, R., Hartmann, J., Slomp, C. P., Goossens, N., & Regnier, P. (2013). Global multi scale segmentation of continental and coastal waters from the watersheds to the continental margins. *Hydrology and Earth System Sciences*, 17(5), 2029–2051. https://doi.org/10.5194/hess-17-2029-2013
- Laruelle, G. G., Lauerwald, R., Pfeil, B., & Regnier, P. (2014). Regionalized global budget of the CO<sub>2</sub> exchange at the airwater interface in continental shelf seas. *Global Biogeochemical Cycles*, 28, 1199–1214.
   https://doi.org/10.1002/2014GB004832
- Laruelle, G. G., Landschützer, P., Gruber, N., Tison, J. L., Delille, B., & Regnier, P. (2017). Global high-resolution monthly
   pCO<sub>2</sub> climatology for the coastal ocean derived from neural network interpolation. *Biogeosciences*, *14*(19), 4545–
   4561. https://doi.org/10.5194/bg-14-4545-2017
- Laruelle, G. G., Cai, W.-J., Hu, X., Gruber, N., Mackenzie, F. T., & Regnier, P. (2018). Continental shelves as a variable but increasing global sink for atmospheric carbon dioxide. *Nature Communications*, 9(1), 454. https://doi.org/10.1038/s41467-017-02738-z
- Lefèvre, N., Diverrès, D., & Gallois, F. (2010). Origin of CO<sub>2</sub> undersaturation in the western tropical Atlantic. *Tellus, Series B: Chemical and Physical Meteorology*, 62(5). https://doi.org/10.1111/j.1600-0889.2010.00475.x
- 876 Liu, K. K., Atkinson, L., Quiñones, R., & Talaue-McManus, L. (2010). Carbon and Nutrient Fluxes in Continental Margins.

- 877 Springer Science & Business Media. https://doi.org/10.1007/978-3-540-92735-8
- 878 Mackenzie, F. T., De Carlo, E. H., & Lerman, A. (2012). Coupled C, N, P, and O Biogeochemical Cycling at the Land-
- Ocean Interface. In *Treatise on Estuarine and Coastal Science* (Vol. 5, pp. 317–342). https://doi.org/10.1016/B978-0 12-374711-2.00512-X
- Manning, A. C., & Keeling, R. F. (2006). Global oceanic and land biotic carbon sinks from the scripps atmospheric oxygen
   flask sampling network. *Tellus, Series B: Chemical and Physical Meteorology*, 58(2), 95–116.
   https://doi.org/10.1111/j.1600-0889.2006.00175.x
- Nakaoka, S. I., Aoki, S., Nakazawa, T., Hashida, G., Morimoto, S., Yamanouchi, T., & Yoshikawa-Inoue, H. (2006).
   Temporal and spatial variations of oceanic pCO<sub>2</sub> and air-sea CO<sub>2</sub> flux in the Greenland Sea and the Barents Sea.
   *Tellus, Series B: Chemical and Physical Meteorology*, 58(2), 148–161. https://doi.org/10.1111/j.1600-0889.2006.00178.x
- Nightingale, P., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., et al. (2000). In situ evaluation of air-sea
   gas exchange parameterizations using novel conservative and volatile tracers. *Global Biogeochemical Cycles*, 14(1),
   373–387. https://doi.org/10.1029/1999GB900091
- Pfeil, B., Olsen, A., Baker, D., Hankin, S., Koyuk, H., Kozyr, A., et al. (2013). A uniform, quality controlled Surface Ocean
   CO<sub>2</sub> Atlas (SOCAT). *Earth System Science Data*, 5(1), 125–143. https://doi.org/10.5194/essd-5-125-2013
- Le Quéré, C., Andrew, R. M., Friedlingstein, P., Sitch, S., Pongratz, J., Manning, A. C., et al. (2018). Global Carbon Budget
   2017. Earth System Science Data Discussions, 10, 405–448. https://doi.org/https://doi.org/10.5194/essd-10-405-2018
- Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F. T., Gruber, N., Janssens, I. A., et al. (2013). Anthropogenic
   perturbation of the carbon fluxes from land to ocean. *Nature Geoscience*, 6(8), 597–607.
   https://doi.org/10.1038/ngeo1830
- Resplandy, L., Keeling, R. F., Rödenbeck, C., Stephens, B. B., Khatiwala, S., Rodgers, K. B., et al. (2018). Revision of
   global carbon fluxes based on a reassessment of oceanic and riverine carbon transport. *Nature Geoscience*, 11(7), 504–
   509. https://doi.org/10.1038/s41561-018-0151-3
- Reynolds, R. W., Smith, T. M., Liu, C., Chelton, D. B., Casey, K. S., & Schlax, M. G. (2007). Daily high-resolution-blended
   analyses for sea surface temperature. *Journal of Climate*, 20(22), 5473–5496. https://doi.org/10.1175/2007JCLI1824.1
- Rödenbeck, C., Baker, D., Gruber, N., Iida, Y., Jacobson, A. R., Jones, S., et al. (2015). Data-based estimates of the ocean
   carbon sink variability First results of the Surface Ocean pCO<sub>2</sub> Mapping intercomparison (SOCOM). *Biogeosciences*,
   *12*(23), 7251–7278. https://doi.org/10.5194/bg-12-7251-2015
- Roobaert, A., Laruelle, G. G., Landschützer, P., & Regnier, P. (2018). Uncertainty in the global oceanic CO<sub>2</sub> uptake induced
   by wind forcing: Quantification and spatial analysis. *Biogeosciences*, *15*(6), 1701–1720. https://doi.org/10.5194/bg-15 1701-2018
- Sabine, C. L., Hankin, S., Koyuk, H., Baker, D., Pfeil, B., Olsen, A., et al. (2013). Surface Ocean CO<sub>2</sub> Atlas (SOCAT)
   gridded data products. *Earth System Science Data*, 5(1), 145–153. https://doi.org/10.5194/essd-5-145-2013
- Schuster, U., McKinley, G. A., Bates, N., Chevallier, F., Doney, S. C., Fay, a. R., et al. (2013). An assessment of the
  Atlantic and Arctic sea–air CO<sub>2</sub> fluxes, 1990–2009. *Biogeosciences*, 10(1), 607–627. https://doi.org/10.5194/bg-10607-2013
- Shadwick, E. H., Thomas, H., Comeau, A., Craig, S. E., Hunt, C. W., & Salisbury, J. E. (2010). Air-sea CO<sub>2</sub> fluxes on the
  Scotian Shelf: Seasonal to multi-annual variability. *Biogeosciences*, 7(11), 3851–3867. https://doi.org/10.5194/bg-73851-2010
- Shadwick, E. H., Thomas, H., Azetsu-Scott, K., Greenan, B. J. W., Head, E., & Horne, E. (2011). Seasonal variability of
   dissolved inorganic carbon and surface water pCO<sub>2</sub> in the Scotian Shelf region of the Northwestern Atlantic. *Marine Chemistry*, *124*(1–4), 23–37. https://doi.org/10.1016/j.marchem.2010.11.004
- Sweeney, C., Gloor, E., Jacobson, A. R., Key, R. M., McKinley, G. A., Sarmiento, J. L., & Wanninkhof, R. (2007).
   Constraining global air-sea gas exchange for CO<sub>2</sub> with recent bomb <sup>14</sup>C measurements. *Global Biogeochemical Cycles*, 21(2), 1–10. https://doi.org/10.1029/2006GB002784
- Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W., et al. (2009). Climatological
   mean and decadal change in surface ocean pCO<sub>2</sub>, and net sea-air CO<sub>2</sub> flux over the global oceans. *Deep Sea Research*

925 926 927	Part II: Topical Studies in Oceanography, 56(8), 554–577. https://doi.org/10.1016/j.dsr2.2008.12.009.Figure Takahashi, T., Sweeney, C., Hales, B., Chipman, D., Newberger, T., Goddard, J., et al. (2012). The Changing Carbon Cycle in the Southern Ocean. Oceanography, 25(3), 26–37. https://doi.org/10.5670/oceanog.2012.71
928	Takahashi, T., Sutherland, S. C., & Kozyr, A. (2016), Global Ocean Surface Water Partial Pressure of CO <sub>2</sub> Database:
929	Measurements Performed During 1957–2015 (Version 2015), ORNL/CDIAC-160, NDP-088(V2015), Carbon Dioxide
930	Information Analysis Center, Oak Ridge National Laboratory, US Department of Energy, Oak Ridge, Tennessee,.
931	https://doi.org/https://doi.org/10.3334/CDIAC/OTG.NDP088(V2015)
932	Tsunogai, S., Watanabe, S., & Sato, T. (1999). Is there a "continental shelf pump" for the absorption of atmospheric CO <sub>2</sub> ?
933	Tellus, 51, 701–712. https://doi.org/10.3402/tellusb.v51i3.16468
934	Turi, G., Lachkar, Z., & Gruber, N. (2014). Spatiotemporal variability and drivers of pCO <sub>2</sub> and air-sea CO <sub>2</sub> fluxes in the
935	California Current System: An eddy-resolving modeling study. Biogeosciences, 11(3), 671-690.
936	https://doi.org/10.5194/bg-11-671-2014
937	Wanninkhof, R. (1992). Relationship Between Wind Speed and Gas Exchange. Journal of Geophysical Research, 97(92),
938	7373–7382. https://doi.org/10.1029/92JC00188
939	Wanninkhof, R. (2014). Relationship between wind speed and gas exchange over the ocean revisited. Limnology and
940	Oceanography: Methods, 12, 351-362. https://doi.org/10.1029/92JC00188
941	Wanninkhof, R., & Trinanes, J. (2017). The impact of changing wind speeds on gas transfer and its effect on global air-sea
942	CO2 fluxes. Global Biogeochemical Cycles. https://doi.org/10.1002/2016GB005592
943	Wanninkhof, R., Asher, W. E., Ho, D. T., Sweeney, C., & McGillis, W. R. (2009). Advances in quantifying air-sea gas
944	exchange and environmental forcing. Annual Review of Marine Science, 1(1), 213–244.
945	https://doi.org/10.1146/annurev.marine.010908.163742
946	Wanninkhof, R., Park, GH., Takahashi, T., Sweeney, C., Feely, R., Nojiri, Y., et al. (2013). Global ocean carbon uptake:
947	Magnitude, variability and trends. <i>Biogeosciences</i> , 10(3), 1983–2000. https://doi.org/10.5194/bg-10-1983-2013
948	Weiss, A., Kuss, J., Peters, G., & Schneider, B. (2007). Evaluating transfer velocity-wind speed relationship using a long-
949	term series of direct eddy correlation $CO_2$ flux measurements. Journal of Marine Systems, 66(1–4), 130–139.
950	https://doi.org/10.1016/j.jmarsys.2006.04.011
951	Weiss, R. F. (1974). Carbon dioxide in water and seawater: the solubility of a non-ideal gas. <i>Marine Chemistry</i> , 2, 203–215.
952	https://doi.org/10.1017/CBO9781107415324.004
953	Winterfeldt, J., & Weisse, R. (2008). Surface marine wind from the NCEP/NCAR and NCEP/DOE-II reanalyses. 18th
954	Symposium on Boundary Layers.
955	Yasunaka, S., Murata, A., Watanabe, E., Chierici, M., Fransson, A., van Heuven, S., et al. (2016). Mapping of the air-sea
956	$CO_2$ flux in the Arctic Ocean and its adjacent seas: Basin-wide distribution and seasonal to interannual variability.
957	<i>Polar Science</i> , 10(3), 323–334. https://doi.org/10.1016/j.polar.2016.03.006
958	

	Coastal seas	Open ocean	Global ocean
$FCO_2 (Tg C yr^{-1})$			
Indian	10 (±5)	-331 (±190)	-322 (±190)
Atlantic	-27 (±3)	-366 (±29)	-393 (±29)
Pacific	-54 (±9)	-331 (±89)	-386 (±90)
Southern Ocean <sup>*</sup>	-17 (±3)	-258 (±123)	-275 (±123)
> 60° N	-88 <sup>**</sup> (±7)	-87 (±21)	-175 (±22)
Marginal seas	-18 (±2)	-	-18 (±2)
Total	-195 (±18)	-1375 (±255)	-1570 (±256)
$FCO_2 (Pg C yr^{-1})$			
	-0.20 (±0.02)	-1.38 (±0.26)	-1.57 (±0.26)
Arctic basin***			-0.12 (±0.06)
Total with Arctic basin			-1.69 (±0.26)

960 961 \*South of 45° S. \*\*This value exclude marginal seas. With marginal seas included, a value of -98 Tg C yr<sup>-1</sup> is obtained for all coastal regions located > 60° N. \*\*\*Schuster et al. (2013).

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Table 1. Global integrated  $FCO_2$ , generated from a 18-year climatology (1998-2015), for the different oceanic basins (Tg C yr<sup>-1</sup>). The partitioning of these basin-wide fluxes between coastal seas and the open ocean is also reported. Numbers in parentheses correspond to the  $FCO_2$  uncertainties calculated according to section 2.5. Marginal seas include the Hudson Bay, the Baltic Sea, the Mediterranean Sea, the Red Sea, the Persian Gulf, and the Black Sea. By combining the coastal/open ocean estimates, the total integrated  $FCO_2$  for the different oceanic basins as well as for the entire global ocean is evaluated (global ocean column).

N°	System name	Class	Surface area $(10^3 \text{ km}^2)$				FC	CO <sub>2</sub> (mo	l C m <sup>-2</sup>	yr <sup>-1</sup> )	Integrated FCO <sub>2</sub> (Tg C yr <sup>-1</sup> )					
			BG 2016	LA 2014	This study	BG2	BG2016		014	This study		BG2016		LA 2014	This study	
					-	mean	σ	mean	σ	mean	σ	mean σ			mean	σ
1	North East Pacific	Subpolar	397	350	394	-2.29	0.17	-1.61	0.25	-1.59	0.6	-10.94	0.82	-6.78	-7.53	2.67
2	Californian current	EBC	118	208	186	-0.34	0.10	-0.05	0.50	-0.60	0.2	-0.48	0.15	-0.14	-1.33	0.48
3	Tropical East Pacific	Tropical	152	183	162	-0.12	0.05	0.09	0.60	0.00	0.1	-0.22	0.10	0.19	0.00	0.18
4	Peruvian upwelling current	EBC	138	143	121	1.44	0.80	0.65	0.60	-0.83*	0.6	2.39	1.33	1.07	-1.20	0.88
5	Southern America	Subpolar	1126	1190	1163	-1.51	0.13	-1.31	0.50	-1.24	0.5	-20.46	1.71	-18.72	-17.38	6.53
6	Brazilian current	WBC	475	484	497	-0.33	0.08	0.10	0.25	-0.12	0.3	-1.87	0.48	0.57	-0.72	1.55
7	Tropical West Atlantic	Tropical	479	488	490	0.86	0.10	0.07	0.60	0.25	0.3	4.93	0.55	0.39	1.49	1.86
8	Caribbean Sea	Tropical	303	358	289	0.10	0.10	0.81	0.60	0.80	0.2	0.37	0.35	3.46	2.77	0.53
9	Gulf of Mexico	Marginal sea	469	532	501	-0.79	0.11	-0.33	0.50	0.24	0.3	-4.48	0.63	-2.10	1.42	1.90
10	Florida upwelling	WBC	545	591	804	-2.25	0.21	-0.38	0.25	-0.83	0.2	-14.69	1.35	-2.72	-8.00	2.04
11	Sea of Labrador	Subpolar	576	638	342	-1.27	0.18	-1.72	0.60	-1.88	0.6	-8.81	1.24	-13.17	-7.70	2.38
12	Hudson Bay	Marginal sea	998	1064	1013	0.31	0.29	-	-	-1.38	0.1 <sup>*</sup>	3.76	3.42	-	-16.81	1.04
13	Canadian Archipelago	Polar	1001	1145	1020	-0.52	0.06	-1.02	0.60	-0.40	0.1	-6.23	0.75	-13.99	-4.91	1.65
14	North Greenland	Polar	544	602	572	-0.97	0.15	-0.61	0.50	-0.93	0.2	-6.33	1.00	-4.40	-6.38	0.63
15	South Greenland	Polar	238	262	248	-3.35	0.44	-3.81	0.50	-2.90	0.3	-9.56	1.26	-11.97	-8.61	0.99
16	Norwegian Basin	Polar	141	162	142	-2.87	0.23	-1.72	0.25	-2.55	0.5	-4.86	0.40	-3.34	-4.34	0.83
17	North East Atlantic	Subpolar	1020	1073	1040	-2.16	0.12	-1.33	0.25	-1.10	0.2	-26.50	1.42	-17.17	-13.68	2.77

18	Baltic Sea	Marginal sea	324	364	336	0.30	0.07	0.51	0.50	-0.39	0.4	1.18	0.29	2.25	-1.58	1.66
19	Iberian upwelling	EBC	251	267	251	-1.13	0.12	0.04	0.25	-0.87	0.3	-3.39	0.35	0.12	-2.64	0.91
20	Mediterranean Sea	Marginal sea	423	529	474	-0.24	0.06	0.62	0.60	-0.01	0.1	-1.20	0.33	3.93	-0.07	0.51
21	Black Sea	Marginal sea	131	172	104	-0.24	0.11	-	-	-2.85	0.6	-0.38	0.17	-	-3.55	0.75
22	Moroccan upwelling	EBC	177	206	179	0.18	0.12	2.92	0.50	0.24**	0.6	0.39	0.26	7.22	0.52	1.20
23	Tropical East Atlantic	Tropical	225	259	259	0.09	0.08	-0.06	0.60	0.03	0.1	0.24	0.21	-0.17	0.09	0.29
24	South West Africa	EBC	300	298	295	0.43	0.40	-1.43	0.50	-1.06	0.8	1.54	1.45	-5.10	-3.76	2.66
25	Agulhas current	WBC	189	239	228	-1.20	0.09	-0.58	0.60	-0.39	0.4	-2.73	0.21	-1.66	-1.07	1.07
26	Tropical West Indian	Tropical	46	68	60	-0.06	0.08	1.00	0.60	0.30	0.2	-0.03	0.04	0.82	0.22	0.12
27	West Arabian Sea	Indian margins	82	92	81	0.35	0.04	1.14	0.60	-0.11**	0.4	0.34	0.04	1.26	-0.11	0.42
28	Red Sea	Marginal sea	158	174	165	0.24	0.03	0.16	0.60	0.67	0.1	0.46	0.07	0.33	1.33	0.20
29	Persian Gulf	Marginal sea	208	233	217	0.04	0.08	-	-	0.85	0.1 <sup>*</sup>	0.09	0.20	-	2.20	0.26
30	East Arabian Sea	Indian margins	298	317	317	0.21	0.12	0.67	0.60	0.61	0.1 <sup>*</sup>	0.75	0.43	2.56	2.31	0.34
31	Bay of Bengal	Indian margins	197	203	204	-0.69	0.12	-0.22	0.60	0.15	0.03	-1.64	0.28	-0.53	0.36	0.08
32	Tropical East Indian	Indian margins	727	763	755	-0.06	0.07	-0.02	0.60	0.16	0.1	-0.48	0.57	-0.17	1.46	0.74
33	Leeuwin current	EBC	81	117	112	-2.05	0.15	-0.98	0.25	-0.49	0.2	-2.01	0.15	-1.38	-0.66	0.25
34	South Australia	Subpolar	392	436	429	-1.37	0.18	-1.14	0.50	-0.28	0.3	-6.44	0.86	-5.98	-1.44	1.60
35	East Australian current	WBC	98	130	133	-1.74	0.18	-1.09	0.25	-0.74	0.2	-2.04	0.21	-1.70	-1.18	0.35
36	New Zealand	Subpolar	263	286	266	-1.23	0.16	-1.25	0.50	-0.90	0.2	-3.88	0.50	-4.27	-2.88	0.49
37	North Australia	Tropical	2278	2292	2369	-0.29	0.11	0.44	0.60	0.31	0.1	-7.87	3.11	12.12	8.72	2.68

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38	South East Asia	Tropical	2130	2160	2196	-0.29	0.07	-0.91	0.60	0.07	0.2	-7.34	1.91	-23.61	1.77	4.22
39	China Sea and Kuroshio	WBC	1132	1129	1172	-1.99	0.15	-1.41	0.60	-0.59	0.1	-27.05	1.99	-19.10	-8.30	2.03
40	Sea of Japan	Marginal sea	233	147	249	-3.07	0.17	-3.47	0.50	-1.44	0.6	-8.61	0.48	-6.11	-4.32	1.84
41	Sea of Okhotsk	Marginal sea	933	952	953	-1.66	0.07	1.31	0.60	-1.85	0.6	-18.62	0.76	1.50	-21.18	7.10
42	North West Pacific	Subpolar	1025	1000	1058	-1.85	0.14	-0.70	0.60	-1.46	0.5	-22.76	1.73	-8.42	-18.58	6.17
43	Siberian Shelves	Polar	1848	1889	1864	-0.47	0.10	-0.90	0.60	-0.50	0.1	-10.50	2.12	-20.32	-11.12	1.82
44	Barents and Kara seas	Polar	1559	1680	1609	-0.75	0.14	-1.60	0.60	-1.78	0.4	-14.18	2.59	-32.23	-34.41	7.75
45	Antarctic Shelves	Polar	2452	2936	2693	-0.90	0.14	-0.15	0.50	-0.12	0.1	-26.63	3.99	-5.38	-3.80	2.55

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\* Due to the scarcity of the data in SOCATv4 dataset for this region, the pCO2 product of Laruelle et al. (2017) underestimates the observed pCO2 and does not 971 reproduce the high FCO2 source (5.7 mol C m<sup>-2</sup> yr<sup>-1</sup>) of the region observed by Friederich et al. (2008).

972 \*\* A pCO2 underestimation is also observed for these two regions because of the scarcity in data to train the algorithm for MARCATS 22 (Laruelle et al., 2017) 973 and given that the monsoon-driven summer upwelling in coastal seas is not captured by the SOM-FFN in MARCATS 27.

974 \*\*\* The scarcity in data in MARCATS 12, 29, 30 and 31 does not allow one to calculate  $\sigma_{pCO2}$  for these regions in the total FCO<sub>2</sub> uncertainty calculation. 975

Table S1. Surface area ( $10^3$  km<sup>2</sup>), air-sea CO<sub>2</sub> exchange rate (FCO<sub>2</sub>, mol C m<sup>-2</sup> yr<sup>-1</sup>) and integrated FCO<sub>2</sub> (Tg C yr<sup>-1</sup>) calculated for 976 each MARCATS regions. BG2016 and LA2014 refer to the study of Bourgeois et al. (2016) and of Laruelle et al. (2014), 977 978 respectively.  $\sigma$  in the LA2014 study corresponds to the uncertainties associated to the data coverage in time and space when

979 they derived FCO<sub>2</sub>. For BG2016, uncertainty corresponds to the FCO<sub>2</sub> interannual variability for the 1993-2012 period. In this

980 study, FCO<sub>2</sub> uncertainties are calculated according to section 2.5.

Figure 1. Discretization of the coastal seas into 45 MARCATS (Margins and CATchment Segmentation, Laruelle et al., 2013). These MARCATS are also grouped into seven major classes following Liu et al. (2010): Eastern and Western (MARCATS 6, 10, 25, 35, 39) boundary currents (EBC and WBC respectively), polar (MARCATS 13, 14, 15, 16, 43, 44, 45) and subpolar (MARCATS 1, 5, 11, 17, 34, 36, 42) margins, tropical margins (MARCATS 3, 7, 8, 23, 26, 37, 38), Indian margins (MARCATS 27, 30, 31, 32) and marginal seas (MARCATS 9, 12, 18, 20, 21, 28, 29, 40, 41).

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**Figure 2.** Global distributions of the annually averaged mean air-sea  $CO_2$  exchange rate (*FCO*<sub>2</sub>, mol C m<sup>-2</sup> yr<sup>-1</sup>) generated from a 18-year climatology (1998-2015) (a) for the coastal seas and (b) for the open ocean. A positive *FCO*<sub>2</sub> value represents a source of  $CO_2$  for the atmosphere, and vice versa for negative *FCO*<sub>2</sub>.

Figure 3. (a) Air-sea  $CO_2$  exchange rate ( $FCO_2$ , mol C m<sup>-2</sup> yr<sup>-1</sup>) aggregated per MARCATS region as a function of latitude. Colors correspond to the seven major classes (see Figure 1, Liu et al., 2010). MARCATS discussed in section 3.1 are reported with their respective number. (b) Mean integrated  $FCO_2$  (Tg C yr<sup>-1</sup>) in the different MARCATS classes. For both panels, error bars correspond to the  $FCO_2$  uncertainties calculated as described in section 2.5.

**Figure 4.** (a) Amplitude of the spatial variability within latitudinal bands of the mean air-sea gas exchange rate of  $CO_2$  (*FCO*<sub>2</sub>, mol C m<sup>-2</sup> yr<sup>-1</sup>) for the coastal seas (in red) and the open ocean (in blue). Shaded areas correspond to the *FCO*<sub>2</sub> longitudinal variability. (b-c) Spatial variation by 1 degree latitudinal bands of the integrated *FCO*<sub>2</sub> (Tg C yr<sup>-1</sup>) and the surface area (10<sup>6</sup> km<sup>2</sup>) for the open ocean and for the coastal seas, respectively.

Figure 5. Spatial distribution of the amplitude of the seasonal  $FCO_2$  variability (mol C m<sup>-2</sup> yr<sup>-1</sup>) (a) 1005 calculated as the root-mean-square for each grid cell of the monthly  $FCO_2$  anomalies (RMS<sub>FCO'</sub>). (b-f) 1006 Seasonal  $FCO_2$  anomaly ( $FCO'_2$  in mol C m<sup>-2</sup> yr<sup>-1</sup>, black lines) in different latitudinal bands calculated as 1007 the mean of the surface weighted average  $FCO'_2$  of all grid cells pertaining to that band. Dotted lines 1008 correspond to uncertainties calculated according to section 2.5. For each panel, a positive value for a 1009 given month implies that the  $FCO_2$  is a stronger source/weaker sink of  $CO_2$  than the 18-year mean 1010 FCO<sub>2</sub> calculated for this latitudinal band. A negative value means a stronger sink/weaker source. Winter 1011 covers the months of January, February and March in the Northern Hemisphere and of July, August and 1012 September in the Southern Hemisphere. The seasonal  $FCO'_2$  profile is plotted twice for each latitudinal 1013 band in order to better visualize the temporal pattern. The root-mean-square of the monthly FCO2 1014 anomalies is also calculated for each latitudinal bands (RMS<sub>FCO'2</sub>, mol C  $m^{-2}$  yr<sup>-1</sup>) and differs from the 1015 1016  $RMS_{FCO'_2}$  in panel (a), which is calculated at the grid cell level.

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Figure 6. Spatial distribution of the amplitude of the seasonal  $FCO_2$  variability (mol C m<sup>-2</sup> yr<sup>-1</sup>) calculated for each MARCATS region as a function of their latitude. Colors represent the MARCATS classes (see Figure 1). For each MARCATS, the root-mean-square of the monthly  $FCO_2$  anomalies is 1021 calculated ( $RMS_{FCO'_2}$ , mol C m<sup>-2</sup> yr<sup>-1</sup>) and differs from the  $RMS_{FCO'_2}$  calculated at the grid cell level. 1022 MARCATS discussed in section 3.3 are reported with their respective MARCATS number. The RMS 1023 value of 3.4 mol C m<sup>-2</sup> yr<sup>-1</sup> for the Baltic Sea (MARCATS 18, latitude of 64° N) is not shown in the 1024 figure.

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Figure 7. Contributions of the different drivers ( $\beta_x$  with  $x = (a) \Delta pCO_2$ , (b) the wind speed, (c) the seaice cover and (d) the cross-correlation between the 3 terms) to the seasonal  $FCO'_2$  in mol C m<sup>-2</sup> yr<sup>-1</sup>. For each map,  $\beta$  is calculated on each grid cell using equation 8. A value close to one indicates that a term contributes strongly to  $FCO'_2$  whereas a value of 0 shows that  $FCO'_2$  is insensitive to that term. Negative values indicate that a term is anti-correlated with  $FCO'_2$ .  $\beta$  are then multiplied by the root-mean-square (RMS) of the total  $FCO'_2$  to highlight regions with high absolute seasonal variability and are thus expressed in mol C m<sup>-2</sup> yr<sup>-1</sup>.

Figure 8. Seasonal  $FCO_2$  anomaly  $(FCO'_2)$  in mol C m<sup>-2</sup> yr<sup>-1</sup>, black lines) in different latitudinal bands 1033 calculated as the mean of the surface weighted average  $FCO'_2$  of all grid cells pertaining to that band. 1034 For each panel, a positive value for a given month implies that the  $FCO_2$  is a stronger source/weaker 1035 sink of  $CO_2$  compared to the 18-year mean  $FCO_2$  calculated for this latitudinal band. A negative value 1036 means a stronger sink/weaker source. The root-mean-square of the monthly FCO2 anomalies is also 1037 calculated for each latitudinal band (RMS<sub>FCO2</sub>, mol C m<sup>-2</sup> yr<sup>-1</sup>). The FCO<sub>2</sub> seasonal variability 1038 associated to the sea-ice cover,  $\Delta pCO_2$ , wind speed and the cross-correlation between the 3 terms is 1039 represented in blue, red, green and pink, respectively. Winter covers the months of January, February 1040 1041 and March in the Northern Hemisphere and of July, August and September in the Southern Hemisphere. 1042

Figure 1.



Figure 2.



Figure 3.



Figure 4.



Figure 5.



Figure 6.



Figure 7.



Figure 8.

