Ocean Surface Carbon Dioxide Fugacity
Observed from Space

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Abstract

We have developed and validated a statistical model to estimate the fugacity (or partial pressure) of carbon dioxide (CO2) at sea surface (pCO2sea) from space-based observations of sea surface temperature (SST), chlorophyll, and salinity. More than a quarter million in situ measurements coincident with satellite data were compiled to train and validate the model. We have produced and made accessible 9 years (2002–2010) of the pCO2sea at 0.5 degree resolutions daily over the global ocean. The data were used to reveal multi-year and regional variability of pCO2sea in relation to ocean parameters. The data also identify uncertainties in the current JPL Carbon Monitoring System (CMS) model-based and bottom-up estimates over the ocean in the subtropical oligotrophic oceans where biological production is not a significant factor in pCO2sea changes.

1. Significance

The alarmingly rapid increase of global atmospheric carbon dioxide (CO2) content has been well documented (e.g., Hofmann et al. 2009), but the distributions of surface sources and sinks have not been sufficiently known. NASA’s Orbiting Carbon Observatory (OCO) is designed to give a more accurate measurement of the column-integrated CO2 content (Crisp et al. 2004) from which surface sources and sinks could be inferred. In the past, sparse atmospheric measurements were assimilated into atmospheric transport models, and, with an inverse technique, the surface sources and sinks were derived (e.g., Hein et al. 1996; Gurney et al. 2004; Patra et al. 2006; Yang et al. 2007; Engelen et al. 2009). However, the large sensitivity of flux inversion systems to regional biases and the large spread of model results were well known (Chevalier et al. 2005; Gurney et al. 2004). Significant deficiency and imbalance of the carbon cycle remain (e.g., Canadell et al. 2007; Le Quere et al. 2007; Watson et al. 2009). The ocean is an important natural sink for atmospheric CO2, and the estimation of ocean-atmosphere exchange in CO2 is critical for understanding and prediction of climate change.

Ocean surface CO2 fugacity is critical in quantifying the flux as described in Section 2. It is also the surface signature of ocean acidity, dynamics, and biogeochemistry. The monitoring and characterization of its variability are also important to ecology and economy.

2. Bulk Parametrization

The air-sea exchange in CO2 (F in Equation (1)) is largely driven by turbulence and has been estimated through bulk parameterization (e.g., Takahashi et al. 2002).

\[ F = k\alpha(\Delta p_{CO2}) \]  

(1)

where \( k \) is the CO2 gas transfer (piston) velocity, \( \alpha \) is the solubility of CO2 in seawater (Weiss 1974), and \( (\Delta p_{CO2}) \) is the difference between the partial pressure of CO2 in water (pCO2sea), and that in air near the surface (pCO2air).
In many studies, the fugacity $f_{CO_2}$ is used in place of $p_{CO_2}$ to distinguish real pressure instead of ideal pressure. For an ideal gas, $f = p$. The conversion of one to another requires knowledge of pressure, temperature, and concentration of CO2 ($x_{CO_2}$). The method is described by DOE (1994) handbook and the modification by Dickson et al. (2007), using empirical formula of Weiss (1974). The difference between $f_{CO_2}$ and $p_{CO_2}$ is generally negligibly small compared with the uncertainties of measurement accuracy. For fixed pressure at 1013 mb and $x_{CO_2}$ of 350 ppm, changing temperature from −5°C to 30°C, changes the differences between $f_{CO_2}$ and $p_{CO_2}$ by less than 0.2%. With fixed temperature at 25°C and $x_{CO_2} = 350$ ppm, changing $p$ from 1013 mb to 900 mb changes the differences between $f_{CO_2}$ and $p_{CO_2}$ by less than 0.3%. In this study, we do not distinguish between the two parameters.

The modeling of the transfer velocity in term of wind speed has been extensively investigated, and the advantage of space-based measurement of wind speed in providing the needed temporal and spatial resolution has been demonstrated (e.g., Liss and Merilvat 1986; Watson et al. 1991; Wanninkhof 1992; Nightingale et al. 2000; Boutin et al. 2002, Carr et al. 2002). Other factors, such as surfactant (Frew 1997; Tsai and Liu 2004; Lin et al. 2003; Hashizume and Liu 2004), and bubbles (e.g., Woolf 1997) have also been studied. A few studies have also suggested that wind speed is not a sufficient parameter for $k$ (e.g., Glover et al. 2002), and surface roughness and stress are better parameters. Glover et al. (2002) supported the theory of the dependence of $k$ on wave slope using the specular backscatter of radar altimeter. The nadir-looking altimeters have limited coverage, and the application of Bragg backscatter measured by the scatterometers is an obvious alternative (Bogucki et al. 2010). With the new perspective of retrieving stress and roughness directly from scatterometer backscatter (e.g., Liu et al. 2010), the improvement of $k$ estimation is being investigated under a complementary study. The $p_{CO_2air}$ is believed to change much less than $p_{CO_2sea}$. It may be estimated through a combination of in situ measurements, satellite data, and numerical models, and it is explored under a separated study. The $p_{CO_2sea}$ has been measured largely on ships; they are not sufficient to characterize spatial and temporal variability. Observations from the vantage point of space may help, and such observations are the focus of this study.

3. Traditional Methods and Deficiency

Space-based sensors do not measure the flux or the fugacity directly. Attempts have been made to establish regional and seasonal relations between $p_{CO_2sea}$ and variables that are more readily measured. For example, Stephens et al. (1995) produced a statistical relation between $p_{CO_2}$ and SST from nine cruises across the Pacific between 1984 and 1989. He concluded that the relation is sufficient to estimate $p_{CO_2sea}$ from satellite SST over the oligotrophic subtropical Pacific, but not over the eutrophic Northwest Pacific, with significant primary production. Many algorithms to related $p_{CO_2sea}$ to SST followed, in the Arabian Sea (Goyet et al., 1998), in the Greenland Sea (Hood et al., 1999), in the Sargasso Sea (Nelson et al. 2001), and in the equatorial Pacific (Cosca et al., 2003), but their applicabilities are limited by geographical region, season, and time scales, depending on the data used to develop the relation. The addition of Chl-a for input was used by Ono et al. (2004) for the subtropics and the subarctic separately, using shipboard measurements in the North Pacific. They found large errors in subarctic springtime. Sarma et al. (2006) used meridional transacts to build algorithms via dissolved inorganic carbon (DIC), using multi-variate linear regression. They computed basin-wide, monthly maps using satellite data, but they found large discrepancies in some regions of the ocean basin. Zhu et al. (2009) developed summer multiple polynomial regression with SST alone and with SST with Chl-a...
together, using South China Sea cruise measurements during July 2004. Instead of developing
the algorithm using only high frequency in-situ measurement, Padin et al. (2009) was the first
one to regress in situ measurement of pCO2sea with overpass satellite observations of SST and
Chl-a, but this was only limited to cruises in the Bay of Biscay. Salinity and alkalinity are also
related to pCO2sea in some of these studies. These studies mostly use linear regression or
multiple polynomials.

Recently, the neural network approach has been applied to estimate pCO2sea in the North
Atlantic. Lefevre et al. (2005) compared two methods: neural network and linear regression in
deriving the monthly distribution of pCO2sea from SST, time, and location, with measurements
of pCO2sea in the Atlantic subpolar gyre (50–70°N, 60–10°W) from 1995 to 1997. The neural
network approach has better accuracy with root-mean-square (RMS) error of 3–11 µatm.
Telszewski et al. (2009) derived the monthly pCO2sea in the North Atlantic (10.5°N–75.5°N) by
applying a self-organizing map neural network from SST, Chl-a, and mixed layer depth (MLD).
The neural network was trained using underway measurements of pCO2sea from 2004 to 2006
and the input data. The RMS error is 11.6 µatm. Friedrich and Oschlies (2009) estimated and
validated monthly pCO2sea in the North Atlantic (15°N–65°N), using a self-organizing neural
network based on a biogeochemical model generated pCO2sea. The monthly pCO2sea has an
RMS error of 15.9 µatm.

In almost all studies, the relationships between pCO2sea and other parameters are developed
with co-incident measurements on cruises, mostly covering a limited region and a particular
season. The correlation coefficients between climatological annual cycle of pCO2sea and
oceanic parameters change from positive to negative over various regions. A single universal
linear or polynomial regression, as derived in these studies, would not work over the global
ocean across all seasons. Multiple relations covering different regions and seasons would have
strong boundary discontinuity problems. Support vector regression, with location and time
(season) as input parameters, will address such problems, and a universal model has been
established for continuous and global coverage. The seasonal and regional limitations of the
relation between pCO2sea and the “driver” parameters is demonstrated with our data product in
Section 9.

4. Support Vector Regression

A “support vector machine” (SVM) is used to derive pCO2sea using space-based observations.
Xie et al. (2008) have demonstrated that SVMs outperform linear regression and neural network
in estimating moisture advection by reducing the bias and the standard deviation in comparison
with observations, and the results include more accurate extreme values. The method has several
major applications. The SVMs for regression are referred as SVR, which is a statistical tool to
derive the relationship between input and output. A comprehensive tutorial of SVR can be found
in Smola and Schölkopf (2004). In the past decade, SVMs have become increasingly popular due
to their broad applications. The approach is relatively easy to use, because there are only a few
parameters to adjust. The simple setting of SVR, with the data training only based on support
vectors, avoids over-fitting of the training data. By using the standard quadratic programming
algorithms, only one global optimum is achieved. Mapping inputs into high-dimensional feature
space and introducing kernal function can solve the nonlinear relationship between inputs and
outputs by turning a nonlinear regression into a linear fitting. For the regression algorithms in
this study, a large training data set is needed in order to represent global coverage with space and
time dependence. The accuracy of SVR depends on selection of the two hyper-parameters and the kernel parameter to optimize the retrieval algorithms (Xie et al. 2008). The initial values of the parameters are empirically estimated from the training data based on previous studies. Then only one parameter varies until the optimized correlation between the trained output and the target data are found.

5. In Situ Measurements

Tremendous effort has been put forward to synthesize consistent and quality-controlled pCO2sea data sets. The database archived at the Lamont–Doherty Earth Observatory (LDEO) contains a large part of the pCO2sea measurements, which have been contributed by many institutions from the U.S. and other countries. The latest version (version 2012, Takahashi et al. 2013) consists of approximately 6.7 million surface ocean pCO2 observations made from 1957 to 2012. We have continuously checked and updated data sets from the continuous measurements by many U.S. and international projects and programs over the global ocean. They include the global Volunteer Observing Ship (VOS) project, the Global CO2 Time-series and Moorings Project, the International Climate Variability Program (CLIVAR) Global Ocean Carbon and Repeat Hydrography Program, the Global Coastal Carbon DATA Project along the east/west coasts of North America and European coast, and the GGlobal Ocean Data Analysis Project (GLODAP). Data collected from cruises over the Pacific are made available from PACIFIC ocean Interior CARbon (PACIFICA). Ongoing cruise measurements of atmospheric and ocean pCO2 are conducted by the CO2 group of the Atlantic Oceanographic and Meteorological Laboratory (AOML) and the Pacific Marine Environmental Laboratory (PMEL). The LDEO database may partly overlap with the latest CARbon dioxide IN the Atlantic Ocean (CARINA) data synthesis project. CARINA includes data from 188 cruises over the Atlantic Ocean, the Arctic Ocean, and the Southern Ocean (Key et al. 2010). Data from the Pacific Ocean are being synthesized in the North Pacific Marine Science Organization (PICES) effort. These data sets are distributed through the Carbon Dioxide Information Analysis Center (CDIAC). Instead of pCO2 or fCO2, the CARINA data output is total dissolved inorganic carbon (TCO2, Pierrot et al. 2010). The conversion of TCO2 and pCO2 follows the program developed for CO2 systems by Lewis and Wallace (1998), along with alkalinity, temperature, salinity, and pressure. Recently, the Surface Ocean CO2 Atlas (SOCAT, Pfeil et al. 2013) project puts together all publicly available underway pCO2sea data from the global oceans between 1968 and 2007 with the 2nd level quality control.

We were able to compile about 250,000 quality-controlled measurements between 2002 and 2010, coincident with satellite measurements of SST and Chl-a, as shown in Fig. 1. This is a living data set, and we will continue to collect new data to fill up the data gaps.
Figure 1. Collocated pCO$_{2\text{sea}}$ measurements with satellite observations during 2002–2010. The pCO$_{2\text{sea}}$ data came from SOCAT and all other sources that were compiled through the Carbon Dioxide Information Analysis Center (CDIAC).

6. Related Space-based Data
The Advanced Microwave Scanning Radiometer for the Earth Observing System (AMSR-E), on board NASA’s Aqua satellite, was launched in May 2002 and has been collecting global SST under clear and cloudy conditions. SST, averaged to 0.25° by 0.25° grids for ascending and descending paths (Wentz and Meissner 1999), was obtained from Remote Sensing System. We have uses SST from microwave sensors in this initial study because they are not obscured by clouds.

Chl-a is derived from a combination of measurements by the Sea-viewing Wide Field-of-view Sensor (SeaWiFs) and the Moderate-Resolution Imaging Spectroradiometer (MODIS) on both Terra and Aqua. The daily Level 3 standard mapped image product has a spatial resolution of 9 km. Because clouds, aerosols, and sunlight availability affect SeaWiFS and MODIS measurements, large data gaps exist on the daily maps. Both spatial and temporal smoothing/averaging will be applied to the Chl-a data to obtain a larger data set collocated with the in-situ pCO$_{2\text{sea}}$ for training/validation.

7. Statistical Model and Validation
A statistical model has been developed to retrieve pCO$_{2\text{sea}}$ from space-based observations using SVR. The training data are constructed as follows. The target data are combined daily averages computed from in situ pCO$_{2\text{sea}}$ observations over global oceans as described in Section 5. Only data starting from 2002, with collocated satellite data have been used in developing the statistical model. The input data include satellite data described in Section 6, which are daily averages of collocated SST from AMSR-E and Chl-a from SeaWiFS. Climatological sea surface salinity (SSS) data (Boyer et al. 2005) have also been used. The day of year, longitude, and latitude will
also be included in the training as input. Time and longitude are taken in the forms of sine and cosine because of their periodicity. The input parameters and the target data (x), except for time and longitude, are normalized as: \[ x' = \frac{x - \bar{x}}{\sigma} \]

where \( \bar{x} \) and \( \sigma \) are the mean and standard deviation of \( x \).

In the current version of the model, ocean MLD was added as an input factor. Operational output from the Global Ocean Data Assimilation System (GODAS) (Behringer 2007) was used. After we reserved a set of 40,000 data groups from the total data set described in Sections 5 and 6 for validation, we randomly selected another 40,000 data groups to train the model. The validation is shown in Fig. 2. For the 40,000 data pairs at 0.5° and daily resolution the mean difference between model predictions and measurements is –0.17 μatm and the root-mean-square (RMS) difference is 16.37 μatm; the latter is 6% of the data range of approximately 270 μatm. Assuming 28 degrees of freedom, the RMS error of daily data is equivalent to 3.1 μatm for a monthly mean. In actual practice, the decorrelation time scale would be longer than a day, and RMS error for monthly mean would be between 3.1 μatm and 16.37 μatm for a 3-day decorrelation time scale.

![Figure 2. Bin-averaged pCO2sea derived from our model plotted vs. observed pCO2. 40000 randomly selected observations for 2002–2010, independent from training data of the statistical model are used. Standard deviation is superimposed on each bin average as error bars.](image)

8. Characterizing the variability

The seasonal and latitudinal variabilities along 148°E in North Pacific (the position is marked in Fig. 3e) of the 9-year mean pCO2sea of our model output (Fig. 3a) agree well with Takahashi climatology (Fig. 3b). South of 34°N, pCO2sea has high values in August and low values in
March–April, in phase with SST (Fig. 3d), but in opposite phase with Chl-a (Fig. 3c). North of this latitude, there are two lows, in April–May and in September–October. The lows correspond to high values of Chl-a. Stronger biological productivity takes up more CO2. The significance of biological processes in the distribution of CO2 is found at higher latitudes while physical-chemical process dominates in sub-tropical oceans. West of this longitude, there is no climatology data at extratropical latitudes, but there are two stations, KEO and JKEO south and north of 34°N with multi-years of measurements of pCO2sea. Their locations are marked in Fig. 3e.

Figures 4 and 5 show that our output agrees well with in situ measurements, two years at KEO and one year at JKEO, that were accessible to us. Our data show year-to-year variation. At KEO to the south (Fig. 4), the annual variations of pCO2sea agree well with SST, but they do not follow the semi-annual variations of Chl-a. At JKEO to the north, there are two cycles a year in opposite phase with Chl-a, but SST has only one cycle per year.

9. Comparison with CMS product

The current JPL CMS bottom-up flux estimate is a model-based (http://cmsflux.jpl.nasa.gov) effort to compute pCO2sea by combining the Estimating the Circulation and Climate of the Ocean Phase II (ECCO2) model (Marshall et al. 1997), which provides the time-evolving physical ocean state, and the Darwin model (Follows and Dutkiewicz 2011), which provides time-evolving ocean ecosystem variables. CMS produced a comparison of the two years of data (2009 and 2010) with coincident LDEO in situ measurement, as shown in Fig 7 of Brix et al. (2012). The scatter is very large, for the high temporal resolution data, but generally around the central line. The more obvious problem is at low observed values (below 320 ppm), where a large number of over-estimations (above 370 ppm) are found. The LDEO data are a subset of the in-situ data we collected as discussed in Section 5. A similar comparison with our data is provided in Fig. 6.

The 2 years of data over the global ocean provided by CMS are collocated with our products and in-situ measurements. Figure 6 shows the results of the comparisons in the form of a bin-averaged scatterplot of 9606 daily data pairs. The lack of sensitivity of CMS product at low values is obvious in Fig. 6a, and this is in agreement with the overestimation at low values shown by Brix et al. (2012). Our products at the same locations and times show better agreement with the in-situ data, as shown in Fig. 6b. The root-mean-square (RMS) difference with in situ measurement is 52.45 μatm for CMS product, higher than 29.82 μatm for our product. The correlation coefficient of 0.45 for CMS is lower than the 0.85 for our product.

The geographic distribution of the 2-year averages of our products (Fig. 7a) agree with Takahashi climatology (Fig 7c) better than CMS product (Fig. 7b). The overestimation (lack of sensitivity) of CMS products is found largely over the oligotrophic subtropical oceans, and this suggests that the Darwin model may be deficient where ocean biological productivity is not a significant driver of surface carbon fugacity, as demonstrated in Section 8.
Figure 3 Latitudinal-time variabiliation along 148°E in North Pacific: (a) pCO2sea derived from Takahashi et al. (2013) climatology data; (b) same as (a), but from statistical model using satellite data, averaged over the 2003–2010 period; (c) and (d) the same as (b), except for log(Chl-a) and SST, respectively; (e) Location of two stations and the 148° E longitude line.
Figure 4. Time series of pCO2sea from statistical model and observed at KEO, compared with SST (top) and log(Chl-a) (bottom).
Figure 5. Time series of pCO2sea from statistical model and observed at JKEO, compared with SST (top) and log(Chl-a) (bottom).
Figure 6. Bin-average comparison with coincident in situ measurement of $pCO_2$ for (a) the version 2 CMS product and (b) for data derived from the statistical model. A total of 9606 collocated data points for the period 2009–2010 are used. Standard deviation in each bin average is superimposed as error bars.
Figure 7. Distribution of pCO2sea (a) derived from satellite data using the statistical model, averaged for period 2009-2010. (b) same as (a), except from the version 2 of CMS products. (c) same as (b), except from Takahashi et al. (2012) climatological data.
10. Future Work

The current data will be distributed through website http://airsea.jpl.nasa.gov/seaflux. It is a living data set. The model will be changed as more data with better coverage become available. It will extend, as new satellite sensors replace the old. For example, AMSR-2 on Global Change Observation Mission 1-Water (GCOM-W) has replaced AMSR-E.

Aquarius is a satellite mission to measure sea surface salinity (SSS), which was launched in 2011 and covers Earth's surface once every 7 days. The SSS data have 1º resolution. Soil Moisture and Ocean Salinity (SMOS), launched in November 2009, is also providing global SSS measurements with 3 days revisits. Intercalibration and merging of SST products are conducted and coordinated by the Group for High Resolution Sea Surface Temperature (GHRSSST) Project. We will use these products to extend the time series, when funding support becomes available.

11. References


### 12. Acronyms and Abbreviations

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Definition</th>
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<tbody>
<tr>
<td>α</td>
<td>solubility of CO2 in seawater</td>
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<tr>
<td>ΔT</td>
<td>difference in time</td>
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<tr>
<td>AMSR-E</td>
<td>Advanced Microwave Scanning Radiometer for EOS</td>
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<tr>
<td>AOML</td>
<td>Atlantic Oceanographic and Meteorological Laboratory</td>
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<tr>
<td>CARINA</td>
<td>CARbon dioxide IN the Atlantic Ocean</td>
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CDIAC Carbon Dioxide Information Analysis Center
Chl-a chlorophyll a
CLIVAR Climate Variability Program
CMS (Jet Propulsion Laboratory) Carbon Monitoring System
CO₂ carbon dioxide
DIC dissolved inorganic carbon
ECCO2 Estimating the Circulation and Climate of the Ocean Phase II (model)
EOS Earth Observing System
f fugacity
F air-sea exchange in CO₂
GCOM-W Global Change Observation Mission 1-Water
GHRSSST Group for High Resolution Sea Surface Temperature
GLODAP GLocal Ocean Data Analysis Project
GODAS Global Ocean Data Assimilation System
JKEO Japan Agency for Marine-Earth Science and Technology (JAMSTEC) Kuroshio Extension Observatory
[The definition came from JGR at http://www.pmel.noaa.gov/people/cronin/articles/TomitaetalJGR10a.pdf]
JPL Jet Propulsion Laboratory
k CO₂ gas transfer (piston) velocity
KEO Kuroshio Extension Observatory
LDEO Lamont–Doherty Earth Observatory
MLD mixed layer depth
OCO Orbiting Carbon Observatory
p pressure
PACIFICA PACIFic ocean Interior CArbon
PICES North Pacific Marine Science Organisation [British spelling]
PMEL Pacific Marine Environmental Laboratory
<table>
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<tr>
<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>RMS</td>
<td>root mean square</td>
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<tr>
<td>SeaWiFs</td>
<td>Sea-viewing Wide Field-of-view Sensor</td>
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<tr>
<td>SMOS</td>
<td>Soil Moisture and Ocean Salinity</td>
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<tr>
<td>SOCAT</td>
<td>Surface Ocean CO$_2$ Atlas</td>
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<td>SSS</td>
<td>sea surface salinity</td>
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<td>SST</td>
<td>sea surface temperature</td>
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<tr>
<td>SVM</td>
<td>support vector machine</td>
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<td>SVM</td>
<td>support vector machine used for regression</td>
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<td>TCO2</td>
<td>total dissolved inorganic carbon</td>
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<tr>
<td>VOS</td>
<td>Volunteer Observing Ship (project)</td>
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