

## ***Interactive comment on “Comparing solubility algorithms of greenhouse gases in Earth-System modelling” by V. M. N. C. S. Vieira et al.***

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### Reviewer comments

Vieira et al. compare the effect of two solubility algorithms applied to gas fluxes and surface water trace gas inventories, using temperatures at two different depths, for the European coastal oceans. They conclude that the different solubility algorithms can lead to millions of tons differences in CO<sub>2</sub> and hundreds of tons of CH<sub>4</sub> and N<sub>2</sub>O in the top meter of the ocean in the modeled region. The modeled surface area is not mentioned so it is difficult to assess if this is the case, although my rough estimate suggests less than a million ton CO<sub>2</sub> assuming a 5 % difference in solubility.

### Author comments

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Considering that each pixel in Figures 1-3 corresponds to roughly 121Km<sup>2</sup> and that there are 41776 pixels, the modelled area is roughly 5054896 Km<sup>2</sup>. This value was inserted in the text. It took us great effort to arrive at the mass balance estimates and we provided all information to consubstantiate how we did it.

#### Reviewer comments

The basic premise that solubility algorithms differ and this has an impact on the thermodynamic driving force is not novel. Neither is the issue that the measurement depth and associated temperature difference can have an impact on calculated gas inventories.

#### Author comments

As this work demonstrated, accurate solubility estimates are fundamental for accurate Earth-System modelling of greenhouse gas in the oceans, its exchange with the atmosphere and thus its impact in climate change. There are not any previous works addressing these issues and there impacts in Earth-System Modelling.

#### Reviewer comments

Part of the confusion is the inconsistent terminology regarding solubility in this paper and the literature at large. The Henry law solubility coefficient should be in (mol/l/atm). The "dimensionless[scalar]" Henry law solubility coefficient,  $K_h$  referred to in the text is better expressed as the Ostwald coefficient(see section 8-11 Reid, R. C., J. M. Prausnitz, and B. E. Poling, 1987: The properties of gases and liquids. McGraw-Hill.).

#### Author comments

The algorithms were applied exactly as in their references, making use of the Henry's law coefficient in its several forms as their authors determined i.e, in mol/l/atm, in atm\*l/mol and in its scalar form. (see Sander 1999, Johnson 2010, Weiss 1974, Weiss and Price 1980, Sarmiento and Gruber 2013). The terminology was applied as presented in these references as well as in the tens of literature on the subject. The scalar Henry's law coefficient was already presented in Sander 1999, Johnson 2010

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and Vieira et al 2013, among other published works, all subject to rigorous reviews. It is not equal to the Ostwald coefficient but its inverse.

#### Reviewer comments

A second issue is the confusing application of the basic gas exchange bulk formula and boundary layer theory, especially the aqueous mass boundary layer. The basic bulk formula is  $F = k_w (C_w - C_o)$ , where  $C_o$  is the top of the boundary layer in equilibrium with the gas concentration in the atmosphere. It can be expressed as  $p^* K_h$  where  $p$  is the fugacity of the gas in air in question. That is, in basic form the gas solubility does not come into play for waterside concentrations when calculating fluxes.

#### Author comments

The basic gas exchange formula was mentioned in two alternative, yet equivalent, formulations depending on the units the gas concentrations were provided (see the first paragraph of the introduction). The first is associated to the double layer thin film model by Liss and Slater (1974) and later used by Johnson (2010) and Vieira et al (2013). The second was implemented as in previous works by researchers as the own reviewer Dr Wanninkhof or Drs Fairall, Frankignoulle, Grachev, Goddard, Jähne, McGillis, Smedman, Wolf, Zappa, Zhang, Zhao, to name a few, and is particularly adequate when gas concentrations are given as partial pressure. Preliminary tests demonstrated both formulations always matched. Since our data provided concentrations in partial pressure, we used the second formulation for the calculus. We clarified these issues in the text.

#### Reviewer comments

I could not follow much of the paper and it will confuse readers. For instance, from first principles the solubility does not affect the gas transfer velocity ( $k(600)$ ) as shown in figure 4 [except in case of bubbles]. The difference shown in figure 1 is the way the gas transfer velocity is calculated. It is because  $k_{600}$  is estimated through the measurement of fluxes and concentration difference in air and water (see top of page

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15928) . As an aside, k600 does not seem to be defined anywhere. The figure caption for figure 4 "Bias in observed transfer velocity" is therefor not correct. It's the bias in the calculated gas transfer [using eddy correlation]

#### Author comments

We never said solubility affects the gas transfer velocity, but only its estimates from field measurements using eddy-covariance methods. This is one of the topics addressed and we made it clear. A mistake meanwhile rectified in Figure 4 caption does not make the article confusing.

#### Reviewer comments

There is not a lot of novel science and once the discussion is streamlined it can be condensed in a couple of pages. It can then be incorporated into the companion paper "improving estimations of greenhouse gas transfer velocities"

#### Author comments

This article stands on its own and can well be published even in the absence of its companion. This article also has an introduction, methods and results. Therefore, merging both would result in a gigantic article deterring potential readers.

#### Reviewer comments

The paper is confusing and difficult to comprehend . . . The grammar and syntax needs to be improved.

#### Author comments

We revised text structure, grammar and syntax, include an explanation to k600 and change Figure 4 caption as the reviewer suggested.

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