

Interactive comment on “A comprehensive estimate for loss of atmospheric carbon tetrachloride (CCl₄) to the ocean” by J. H. Butler et al.

Anonymous Referee #2

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This is a very welcome and important study reporting a revised partial ocean lifetime for CCl₄. These authors are the only ones in the world with a sufficiently large and high quality data set of ocean CCl₄ (and CFC11), alongside the relevant expertise, for such a global study, and their new data and analyses are significant and timely.

The new results are carefully reported and analysed, and the manuscript is generally extremely well written and clear. I have no major criticisms, but find that in several places there could be more detailed explanations of uncertainties and computations, as detailed below. I also recommend some discussion of the study of Huhn et al. (Deep Sea Research, 2001) which deduced that oceanic CCl₄ depletion was faster at warmer temperatures (contrary to the present study where there was no dependence

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on temperature), and deduced a depletion rate of approximately 22% per year for temperatures above 13oC.

Pg 3 – Computations. The temperature dependence of the solubility of CFC11 is about a factor of 4 different from CCl4 from 0-30 oC (Table 2). There is an even greater absolute difference in solubility. Surely the temperature dependence of the diffusivities is also important? –ideally being the same for 2 gases. How much do all these differences compromise the use of CFC11 as a way to determine irreversible loss of CCl4 by difference?. It would be good to see some quantitative analysis of this. This would help the reader to quantitatively understand the statement later on (pg 4, line 5) that “because of the differences in physical properties of various gases, in situ consumption is more probable if the corrected saturation anomaly is less than -2%.”

Pg 5, Ln 21: “While the corrections for physical effects that we use do make the saturation anomaly more negative,” I’m intrigued by this. Hypothetically, if the data coverage was 100%, wouldn’t the physical effects cancel because there would be as many instances of water masses radiatively cooling as radiatively warming? This is probably a very naïve assumption (I’m not an oceanographer). But it would be good to see an explanation as to why the physical effects act to overall make the saturation anomaly more negative.

Pg 6. Lns 12-14 “First-order computations of the time required to mix waters between the surface and intermediate depths, however, suggest that, on average, the loss at depth cannot fully support the observed surface water deficits.”. It’s not exactly clear how the estimates of mixing time scales from surface and deeper waters support this hypothesis.

Page 7 In 20 onwards. The newly calculated ocean sink has changed by more than a factor of 2 compared to the previous assessment of 94 years. The sink is directly proportional to K_w , which is 30-40% smaller in this study than that of Wanninkhof 1992 used previously. The new analysis apparently has a very similar partial pressure

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difference (air/sea) of CCl₄ used previously to estimate the 94 year lifetime. So – why has the sink changed by a factor of 2 rather than only 30-40%? Even at higher wind speed conditions, which might dominate the overall sink, the differences in K_w are nowhere near a factor of 2. I think this deserves some detail in the explanation.

Minor corrections: Page 7 ln 10 – needs space between “bomband” Eqn (3) – Dcorrected is introduced, which should be defined (DCCl₄ – $\Delta\delta\text{I}\dot{S}\dot{S}$) Fig 8 caption – needs fixing (typos). Also, please describe the separate studies used in the legend within the caption.

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