

Interactive comment on “Global Annual Mean Atmospheric Histories, Growth Rates and Seawater Solubility Estimations of the Halogenated Compounds HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, HFC-125, HFC-23, PFC-14 and PFC-116” by Pingyang Li et al.

Anonymous Referee #1

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“GENERAL COMMENTS”

The authors synthesize the atmospheric concentration history and review the solubility of HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, HFC-125, HFC-23, PFC-14 and PFC-116. This study is valuable as a first step of evaluating the utility of these compounds as oceanic transient tracers. Some revisions seem to be needed. In particular, the following two revisions are needed.

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First, estimate of Ostwald solubility coefficients by LFERs (Method II) should be revised. According to Abraham et al. (2001), the solvation parameter method of Abraham relies on two linear free energy relationships, LFERs, one for processes within condensed phases, Eq. (i), and one for processes involving gas to condensed phase transfer, Eq. (ii).

$$\log SP = c + eE + sS + aA + bB + vV \text{ (i)}$$

$$\log SP = c + eE + sS + aA + bB + IL16 \text{ (ii)}$$

Here L16 is the solute gas-hexadecane partition coefficient at 298 K; I is a coefficient for L16; and other coefficients and descriptors are the same as those described in the manuscript. Therefore, Ostwald solubility coefficients in pure water (L_0) can be estimated by use of Eq. (ii) (processes involving gas to condensed phase transfer), while salting-out coefficients (k_s) can be estimated by use of Eq. (i) (processes within condensed phases). This point should be noted clearly in the manuscript.

In this study, as seen in Eqs. 10, 12 and 13 (Sect. 2.10.2), the relationship for water to solvent (processes within condensed phases, Eq. (i)) was used to estimate Ostwald solubility coefficient in pure water (L_0). It should be revised as mentioned above. Furthermore, Tables S2 and S4 should be revised. When Eq. (ii) is used to estimate L_0 , may the discussion in Sect. 3.3 (page 20), which includes comparison between the Method II and the Revised Method II, lead to the same result as described in the manuscript?

Second, hydrolysis of HCFC-22 should be taken into consideration in discussion of transient tracer potential (for example, Sect. 3.4) because rate constants for hydrolysis of HCFC-22 in alkaline aqueous solutions are much larger than those for hydrolysis of other HCFCs and HFCs [for example, le Noble, W. J. Am. Chem. Soc., 87, 2434-2438 (1965); Kutsuna, S. et al. Int. J. Chem. Kinet. 43, 639-647 (2011)].

May hydrolysis of HCFC-22 in seawater make a significant influence on transient tracer

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potential of HCFC-22? If hydrolysis of HCFC-22 is significant, what would be expected when HCFC-22 is used as a transient tracer?

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“SPECIFIC COMMENTS”

Page 4, lines 21-22: Combustion in thermal power station has been pointed out as a tropospheric sink of PFCs [Ravishankara, A. R. et al., Science, 259, 194-199 (1993)]. It should be cited.

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Page 4, lines 28-30, “Cutting the production and consumption of HFCs by more than 80 % over the next 30 years under the Kigali amendment of the MP”: Under the Kigali amendment, the amount of cutting the production and consumption of HFCs is based on the amount scaled by GWP of each HFC. This point had better be described.

Page 5, lines 17-18: Why were the same AGAGE calibration scales used by converting NOAA and UEA data to the AGAGE scale? Is the reason explained somewhere in the manuscript?

Page 10, line 3, “The ionic strength of seawater (I_v, in g L⁻¹): The unit of ionic strength should be checked.

Page 10, Eq. (8): DS, ES and FS seems to need definition.

Page 10, line 13, unit of the McGowan’s characteristic molar volume in Eq. 8: According to Abraham et al. (2001), unit of the McGowan’s characteristic molar volume is not cm³ mol⁻¹/100 but dm³ mol⁻¹/100. The unit should be checked.

Page 12, line 34, Figures 2-9: The method to calculate IHG and its error (gray parts in Figures 2-9) seems to need explanation.

Page 14, line 11: What does it mean by “30% (median) larger”? It seems to need more explanation.

Page 16, lines 27-29: Atmospheric lifetime of HFC-23 is much longer than that of HFC-134a. Hence, time-profiles are expected to be different between HFC-134a and

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HFC-23 after the consumption restrictions imposed by the 2016 Kigali Amendment to the Montreal Protocol. This point should be discussed.

Page 16, lines 34-35, “the growth rates in both hemispheres are really similar”: Do the growth rates mean those of HFC-125 or some target compounds? What does it mean by “really similar”?

Page 17, lines 28-29, “For compounds with shorter lifetimes. . . ”: This sentence is difficult to understand. It should be revised.

Page 18, lines 7-9: The experimental data for HFC-125 are scattered as seen in Fig. S5. This point had better be described more clearly.

Page 18, lines 37-38: The dipolarity/polarizability (S) for C2F6 was estimated to be equal to the average of the S of CF4 and C3F8. This estimate might have substantial errors. Influence of errors of S on the salting-out coefficients of C2F6 should be evaluated.

Page 19, line 32: How is uncertainty from salting-out coefficients estimated? It seems to need more explanation.

Supplement, Eq. (2): What does it mean by $i = 0$ in summation?

“TECHNICAL CORRECTIONS”

Page 5, line 21: “can used” is “can be used”.

Page 6, line 32: “fin air” is “furn air”.

Page 12, line 11: “By combining Eq. (4), (12) and (14)” may be “By combining Eqs. (4), (14) and (16)”.

Page 12, line 39 – page 13, line 1: “hemispheric” is “hemisphere”.

Page 13, line 21; page 13, line 39: “This consistent” is “This is consistent”

Page 15, line 13: “These consistent” is “These are consistent”

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Page 16, line 17: “0.038 ± 0.007” is “0.138 ± 0.007”.

Page 16, line 17: “0.033 ± 0.008” is “0.133 ± 0.008”.

Page 16, line 34: “This could attributed” is “This could be attributed”.

Page 17, line 9: “This consistent” is “This was consistent”.

Page 17, line 13: “countries, Moreover,” is “countries. Moreover,”.

Page 18, line 9: “is chose” is “is chosen”.

Page 18, line 32: “298.15-338.15 K” is “273.15-313.15 K”.

Page 19, line 31: “9.0695e-05” is “ 9.0695×10^{-5} ”.

Page 19, line 31: “9.1858e-05” is “ 9.1858×10^{-5} ”.

Page 21, line 38: “can be also be” is “can also be”.

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