



Supplement of

Modelling marine emissions and atmospheric distributions of halocarbons and dimethyl sulfide: the influence of prescribed water concentration vs. prescribed emissions

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1 S-Table 1: Overview of the aircraft campaigns used in this study.

Campaign	Date	Altitude range [km]	Location	PI(s)/ Reference
TRACE-A	1992/09 - 1992/10	0 - 12	Atlantic Ocean	D. Blake/Blake et al. (2003) ¹
STRAT	1996/02 - 1996/12	14 - 21	East Pacific	E. Atlas/Schauffler et al. (1998) ²
PEM-TROPICS-A	1996/08 - 1996/10	0 - 11	Tropical Pacific Ocean	E. Atlas/D. Blake
POLARIS	1997/09	15 - 21	East Pacific	E. Atlas/ ²
PEM-TROPICS-B	1999/03 - 1999/04	0 - 12	Tropical Pacific Ocean	E. Atlas/D. Blake
ACCENT	1999/09	15 - 16	Central America	E. Atlas/ ²
TRACE-P	2001/02 - 2001/04	0 - 12	West Pacific	E. Atlas/D. Blake
PRE-AVE	2004/01 - 2004/02	8 - 19	Central America	E. Atlas/ ²
INTEX	2004/07 - 2006/05	0 - 11	Pacific/USA/Atlantic	D. Blake ¹
AVE	2006/06	15 - 19	Central America	E. Atlas/ ²
CR-AVE	2006/01 - 2006/02	2 - 19	Central America	E. Atlas/Ashfold et al. (2012) ²
CARIBIC	2006/10 - 2009/10	9 - 14	C. America, S.E. Asia, E. Pacific	D. Oram/Wisher et al. (2014)
TC4	2007/07 - 2007/08	0 - 18	Central America	E. Atlas, D.R. Blake/ Ashfold et al. (2012) ²
ARCTAS	2008/04 - 2008/07	0 - 11	Canada	D. Blake
HIPPO-1	2009/01	0 - 14	East Pacific	E. Atlas/Wofsy et al. (2012) ³
HIPPO-2	2009/11	0 - 14	Pacific Ocean	E. Atlas/Wofsy et al. (2012) ³
HIPPO-3	2010/03 - 2010/04	0 - 14	Pacific Ocean	E. Atlas/Wofsy et al. (2012) ³
HIPPO-4	2011/06 - 2011/07	0 - 14	Pacific Ocean	E. Atlas/Wofsy et al. (2012) ³
HIPPO-5	2011/08 - 2011/09	0 - 14	West Pacific	E. Atlas/Wofsy et al. (2012) ³
POST-ATTREX	2011/11	13 - 19	East Pacific	E. Atlas ²
SHIVA	2011/11- 2011/12	0 - 13	South China Sea	A. Engel/ Sala et al. (2014)
ESMVAL	2012/09	0 - 15	Africa	A. Engel
TACTS	2012/08 - 2012/09	0 - 15	North Africa	A. Engel

2 ¹ Halocat database

3 ² <https://espoarchive.nasa.gov/archive/browse>

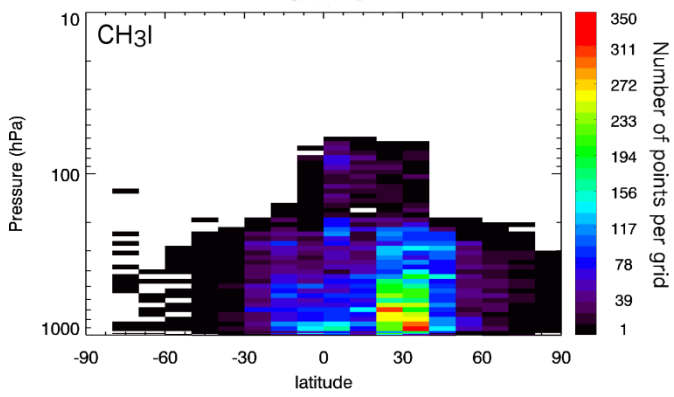
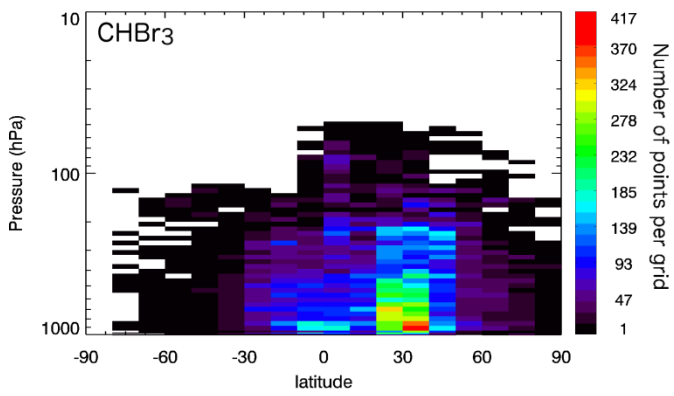
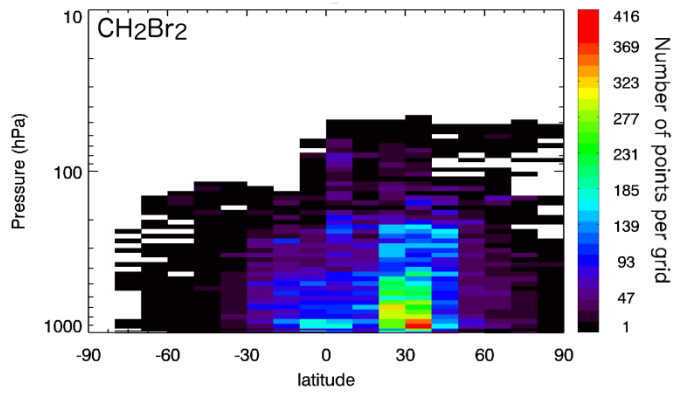
4 ³ <https://www.eol.ucar.edu>

1 S-Table 2: Global averages for the year 2012 as a comparison for the resolution of grid T42 and T106 using the
 2 prescribed emission and concentration climatologies described in section and the k_w -parameterization
 3 according to Nightingale et al. (2000). k_w = water side transfer velocity of air-sea gas exchange, vmr= volume
 4 mixing ratio.

	T42 PWC	T42 PE	T106 PWC	T106 PE
wind 10 m [m s^{-1}]	6.31		6.31	
$k_w \text{ CH}_2\text{Br}_2$ [m s^{-1}]	2.5×10^{-5}		2.3×10^{-5}	
$k_w \text{ CHBr}_3$ [m s^{-1}]	2.3×10^{-5}		2.2×10^{-5}	
$k_w \text{ CH}_3\text{I}$ [m s^{-1}]	2.6×10^{-5}		2.5×10^{-5}	
$k_w \text{ DMS}$ [m s^{-1}]	2.5×10^{-5}		2.4×10^{-5}	
surface vmr CH_2Br_2 [mol mol^{-1}]	1.1×10^{-12}	1.56×10^{-12}	1.0×10^{-12}	1.5×10^{-12}
surface vmr CHBr_3 [mol mol^{-1}]	8.9×10^{-13}	9.38×10^{-13}	8.8×10^{-13}	9.4×10^{-13}
surface vmr CH_3I [mol mol^{-1}]	5.4×10^{-13}	6.25×10^{-13}	5.0×10^{-13}	6.0×10^{-13}
surface vmr DMS [mol mol^{-1}]	1.4×10^{-10}	2.13×10^{-10}	1.3×10^{-10}	2.0×10^{-10}
flux CH_2Br_2 [$\text{mol m}^{-2} \text{ s}^{-1}$]	2.2×10^{-14}		2.2×10^{-14}	
flux CHBr_3 [$\text{mol m}^{-2} \text{ s}^{-1}$]	5.9×10^{-14}		5.9×10^{-14}	
flux CH_3I [$\text{mol m}^{-2} \text{ s}^{-1}$]	9.2×10^{-14}		8.8×10^{-14}	
flux DMS [$\text{mol m}^{-2} \text{ s}^{-1}$]	4.5×10^{-11}		4.3×10^{-11}	

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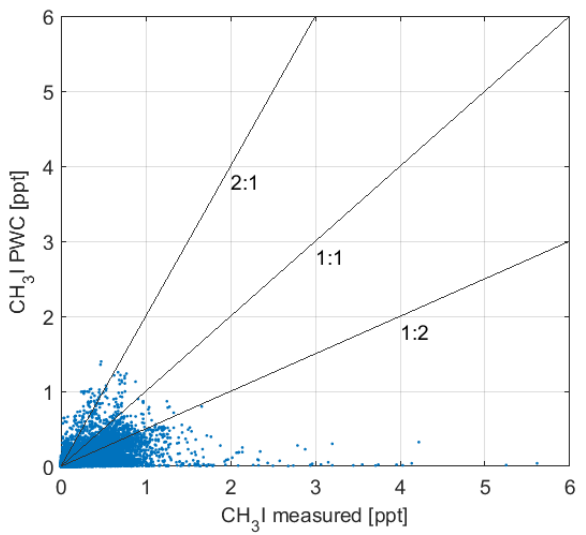
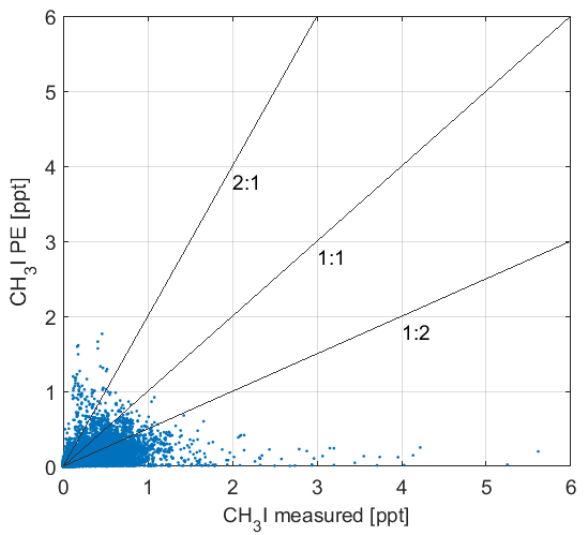
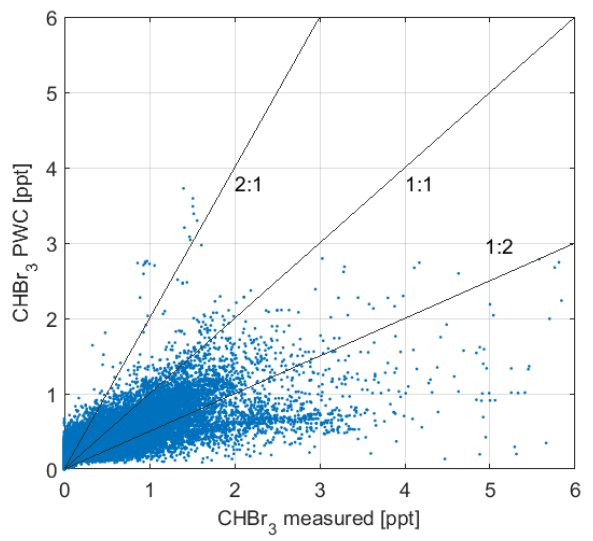
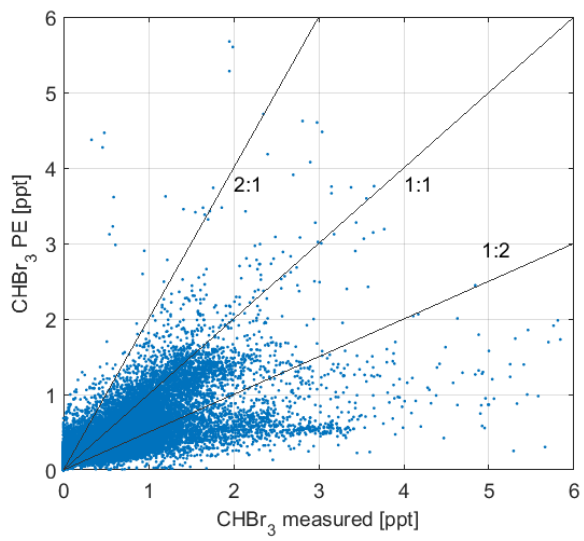
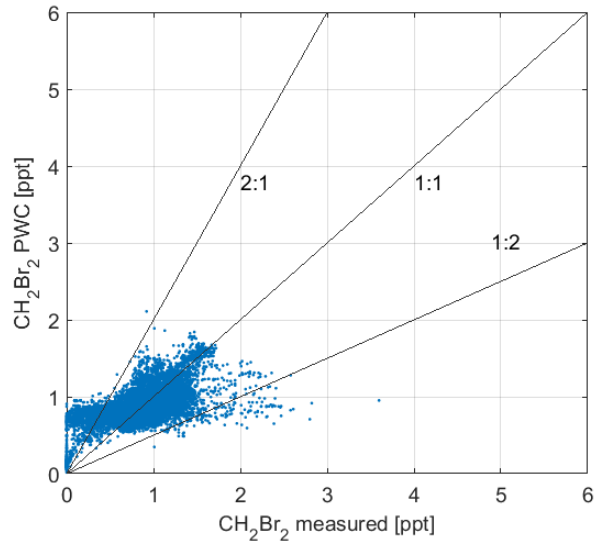
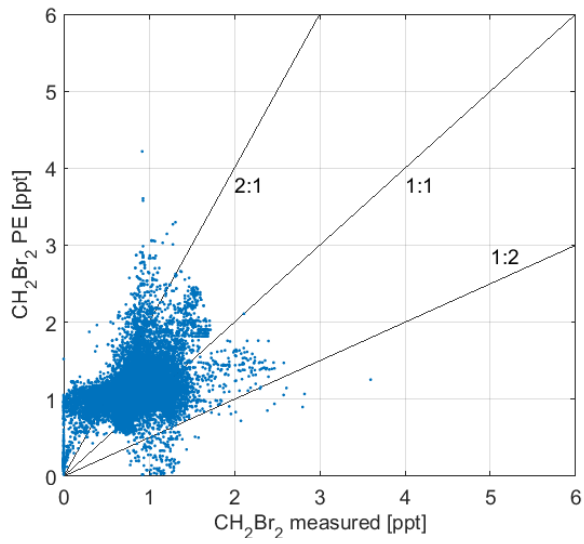
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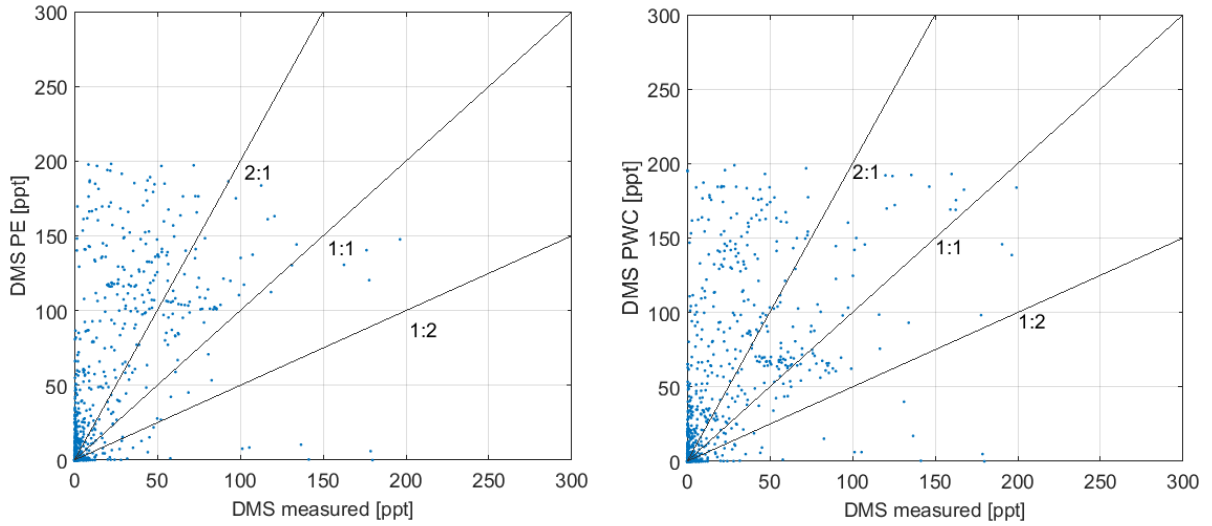
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S-Figure 1: Numbers of measurement per 10° latitude bin for Figure 7.

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1 S-Fig. 2: Scatterplots for direct comparison between model output of simulations 1 (PWC)
 2 and 2 (PE) and observations. The model was subsampled at time and location of observations.
 3 For the halocarbons, observations from 23 aircraft campaigns as illustrated in fig. 3a were
 4 used to create this scatterplot. For DMS, observations from ship and aircraft campaigns as
 5 described in figure 3b were taken into account.

6

7 Equations to compute error metrics

8

9 Error metrics are computed according to Yu et al. (2006) with N=number of data pairs
 10 observation and model output, M=model output and O=observation.

11 Mean bias

$$B_{mb} = \frac{1}{N} \sum (M_i - O_i) = \bar{M} - \bar{O}$$

12 Mean absolute gross error

$$E_{MAGE} = \frac{1}{N} \sum |M_i - O_i|$$

13 RMSE

$$E_{RMSE} = \left[\frac{1}{N} \sum (M_i - O_i)^2 \right]^{\frac{1}{2}}$$

14 Fractional bias

$$B_{fb} = \frac{1}{N} \sum \frac{(M_i - O_i)}{(M_i - O_i)/2}$$

1 Fractional absolute error

$$E_{fae} = \frac{1}{N} \sum \frac{|M_i - O_i|}{(M_i - O_i)/2}$$

2 Normalized mean bias factor

3 $B_{nmbf} = \frac{\bar{M}}{\bar{O}} - 1$ for $\bar{M} \geq \bar{O}$

4

5 $B_{nmbf} = 1 - \frac{\bar{O}}{\bar{M}}$ for $\bar{O} \geq \bar{M}$

6

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