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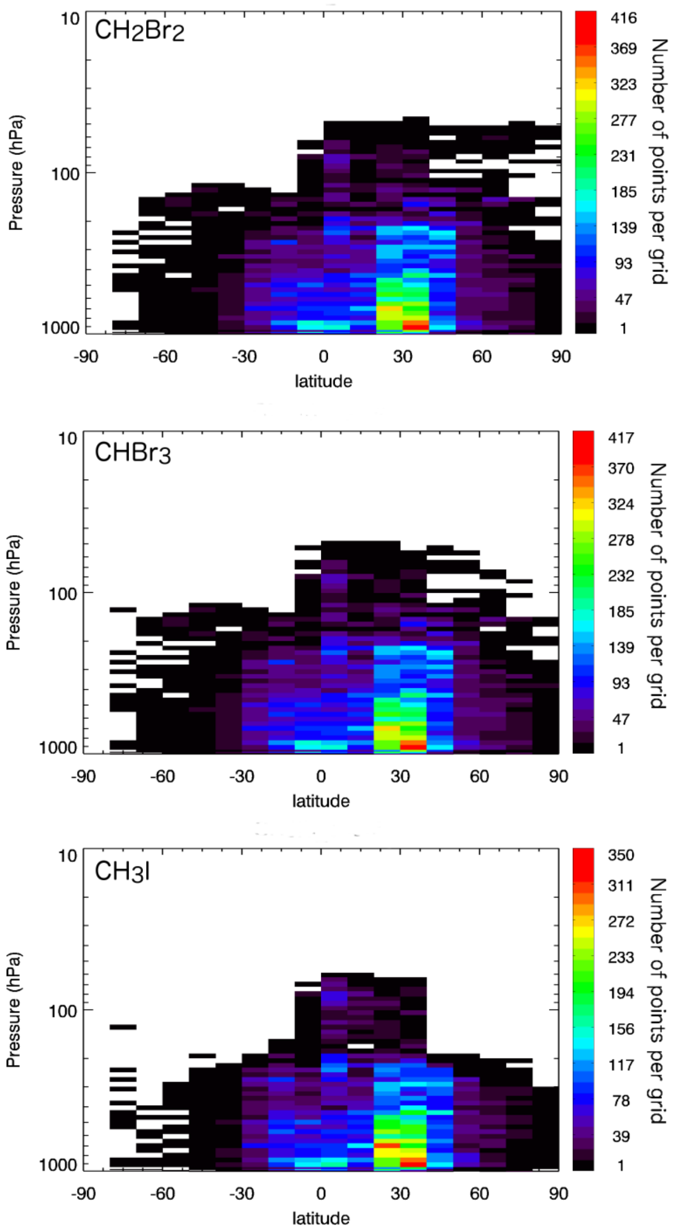
S-Table 1: Overview of the aircraft campaigns used in this study.

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	Eeat 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

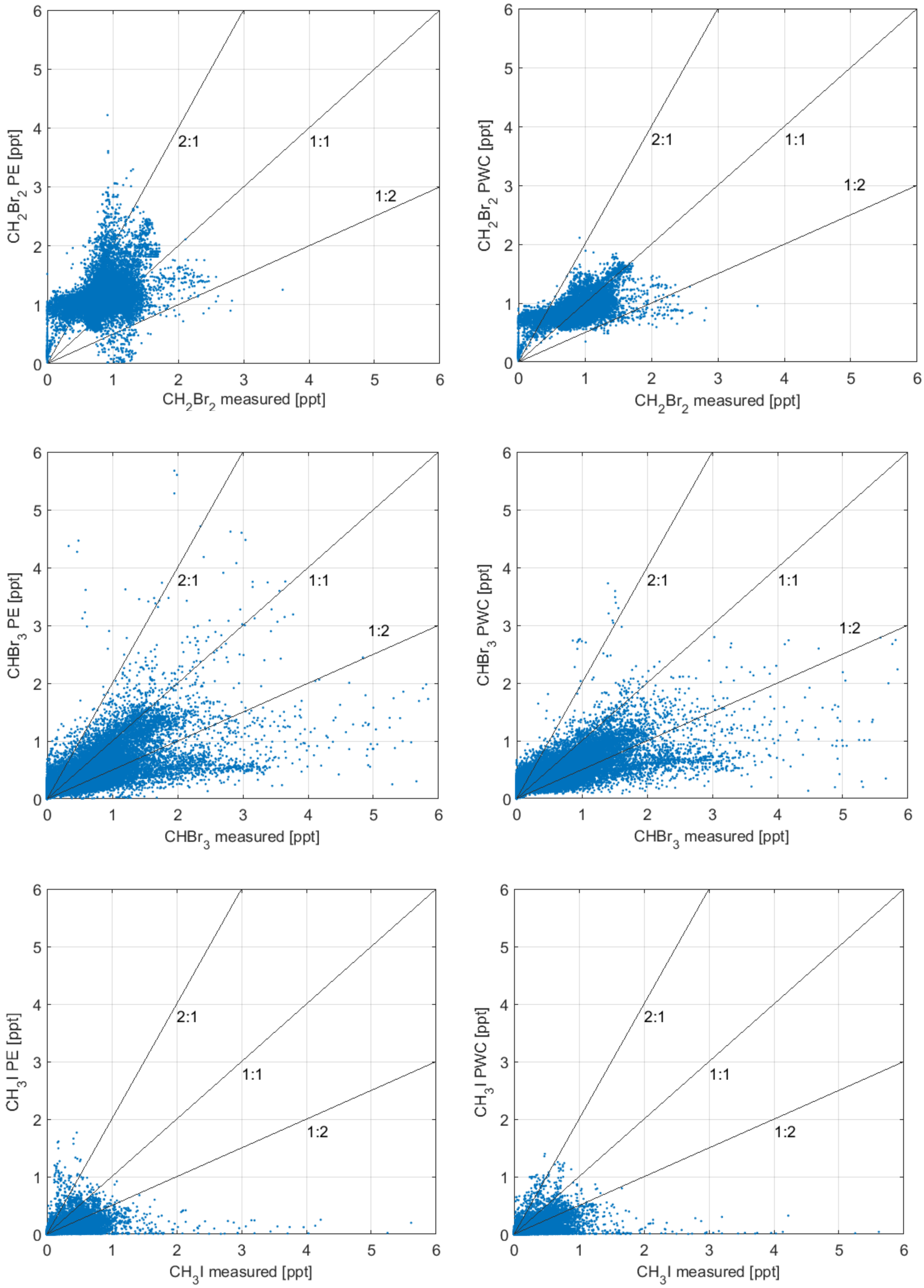
[1] *Halocat database*
[2] *https://espoarchive.nasa.gov/archive/browse*
[3] https://www.eol.ucar.edu

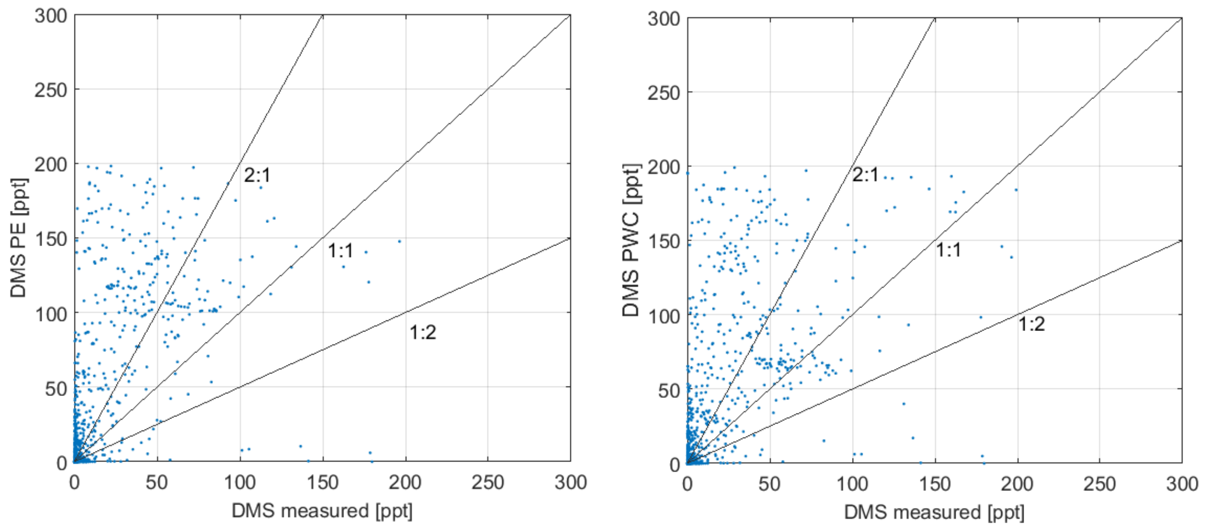
S-Table 2: Global averages for the year 2012 as a comparison for the resolution of grid T42 and T106 using the prescribed emission and concentration climatologies described in section and the k_w-parameterization according to Nightingale et al. (2000). k_w = water side transfer velocity of air-sea gas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

	T42 PWC	T42 PE	T106 PWC	T106 PE
wind 10 m [m s^{-1}]	6.31		6.31	
k_w CH_2Br_2 [m s^{-1}]	2.5 $x10^{-5}$		2.3 $x10^{-5}$	
k_w $CHBr_3$ [m s^{-1}]	2.3 $x10^{-5}$		2.2 $x10^{-5}$	
k_w CH_3I [m s^{-1}]	2.6 $x10^{-5}$		2.5 $x10^{-5}$	
k_w DMS [m s^{-1}]	2.5 $x10^{-5}$		2.4 $x10^{-5}$	
surface vmr CH_2Br_2 [mol mol^{-1}]	1.1 $x10^{-12}$	1.56 $x10^{-12}$	1.0 $x10^{-12}$	1.5 $x10^{-12}$
surface vmr $CHBr_3$ [mol mol^{-1}]	8.9 $x10^{-13}$	9.38 $x10^{-13}$	8.8 $x10^{-13}$	9.4 $x10^{-13}$
surface vmr CH_3I [mol mol^{-1}]	5.4 $x10^{-13}$	6.25 $x10^{-13}$	5.0 $x10^{-13}$	6.0 $x10^{-13}$
surface vmr DMS [mol mol^{-1}]	1.4 $x10^{-10}$	2.13 $x10^{-10}$	1.3 $x10^{-10}$	2.0 $x10^{-10}$
flux CH_2Br_2 [mol m^{-2} s^{-1}]	2.2 $x10^{-14}$		2.2 $x10^{-14}$	
flux $CHBr_3$ [mol m^{-2} s^{-1}]	5.9 $x10^{-14}$		5.9 $x10^{-14}$	
flux CH_3I [mol m^{-2} s^{-1}]	9.2 $x10^{-14}$		8.8 $x10^{-14}$	
flux DMS [mol m^{-2} s^{-1}]	4.5 $x10^{-11}$		4.3 $x10^{-11}$	



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


CH2Br2 PE [ppt]
CH2Br2 measured [ppt]
CH2Br2 PWC [ppt]
CH2Br2 measured [ppt]
CHBr3 PE [ppt]
CHBr3 measured [ppt]
CHBr3 PWC [ppt]
CHBr3 measured [ppt]
CH3I PE [ppt]
CH3I measured [ppt]
CH3I PWC [ppt]
CH3I measured [ppt]
2:1
1:1
1:2



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

Equations to compute error metrics

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

Mean bias

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

Mean absolute gross error

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

RMSE

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

Fractional bias

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

Fractional absolute error

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

Normalized mean bias factor

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

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

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