Simulations and Inverse Modeling of Global Methyl Chloride

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Introduction

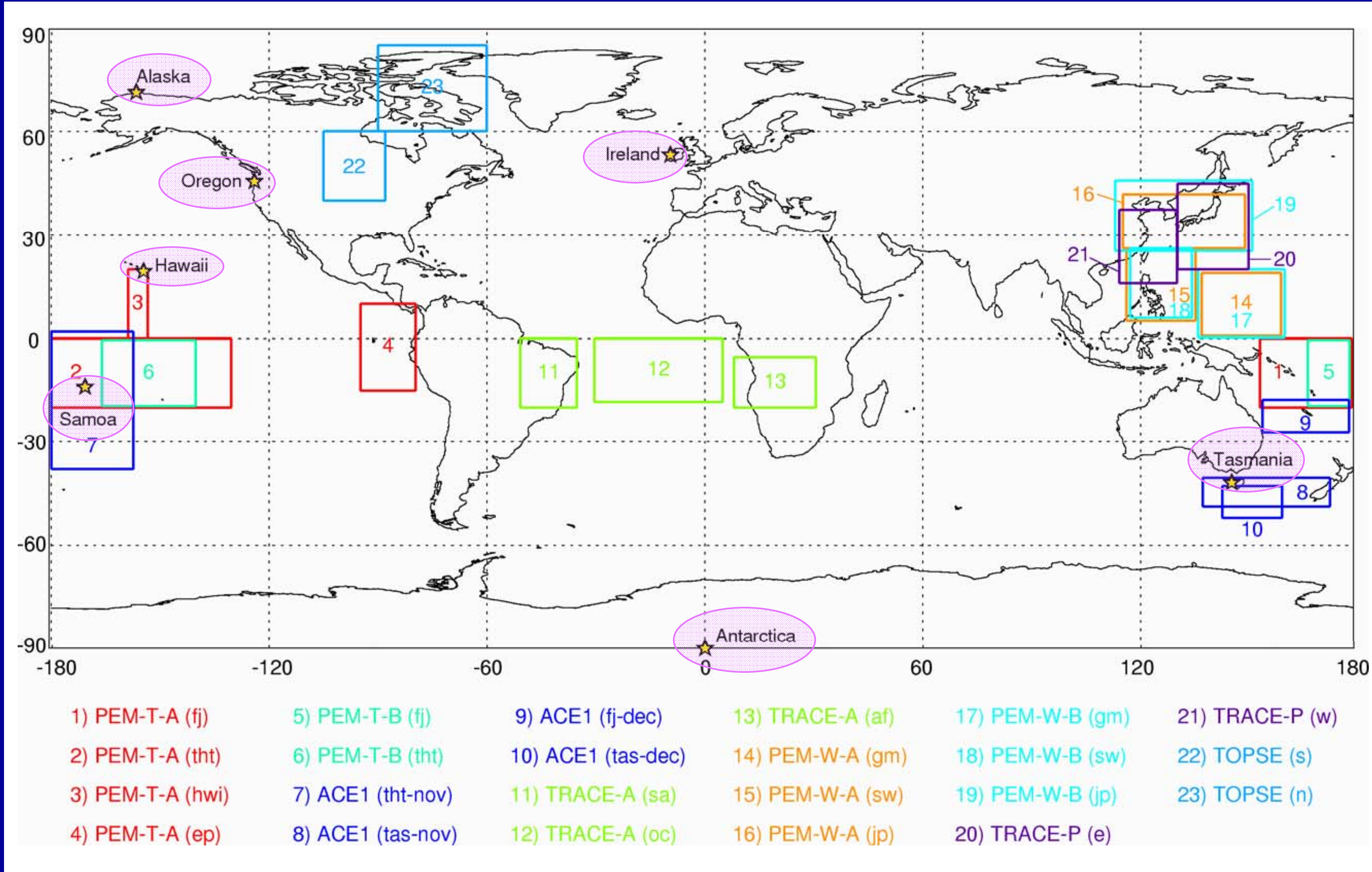
Methyl chloride (CH_3Cl)

- One of the most abundant chlorine-containing gases in the atmosphere
- A major contributor to stratospheric chlorine, which plays an important role in the processes of stratospheric ozone depletion
- Known sources are about a half of known sinks.

Objectives

- Global simulations of atmospheric CH_3Cl are conducted using a global 3-D chemical transport model.
- Inversion is applied in order to constrain the source distributions and seasonal variations using surface and aircraft observations.

Surface measurement sites and aircraft observation regions



Emissions/sinks

unit: Gg yr^{-1}

	A priori	References	
Sources (total)	**(4397)**		
Ocean	507	650 (40-950) 470 - 500	Khalil *et al*. [1999] Moore [2000]
Biomass burning	610	910 (650-1120)	Lobert *et al*. [1999]
Incineration/industrial	162	162 (21-207)	McCulloch *et al*. [1999]
Salt marshes	170	170 (65-440)	Rhew *et al*. [1999]
Wetlands	48	48	Varner *et al*. [1999]
Biogenic	2900	2330-2430	Lee-Taylor *et al*. [2001]
Sinks (total)	**(4397)**		
OH reaction	3992	3500 (2800-4600)	Koppmann *et al*. [1993]
Ocean	149	150 90-150	Khalil *et al*. [1999] Moore [2000]
Soil	256	256	Khalil and Rasmussen [1999], Keene *et al*. [1999]

Forward modeling

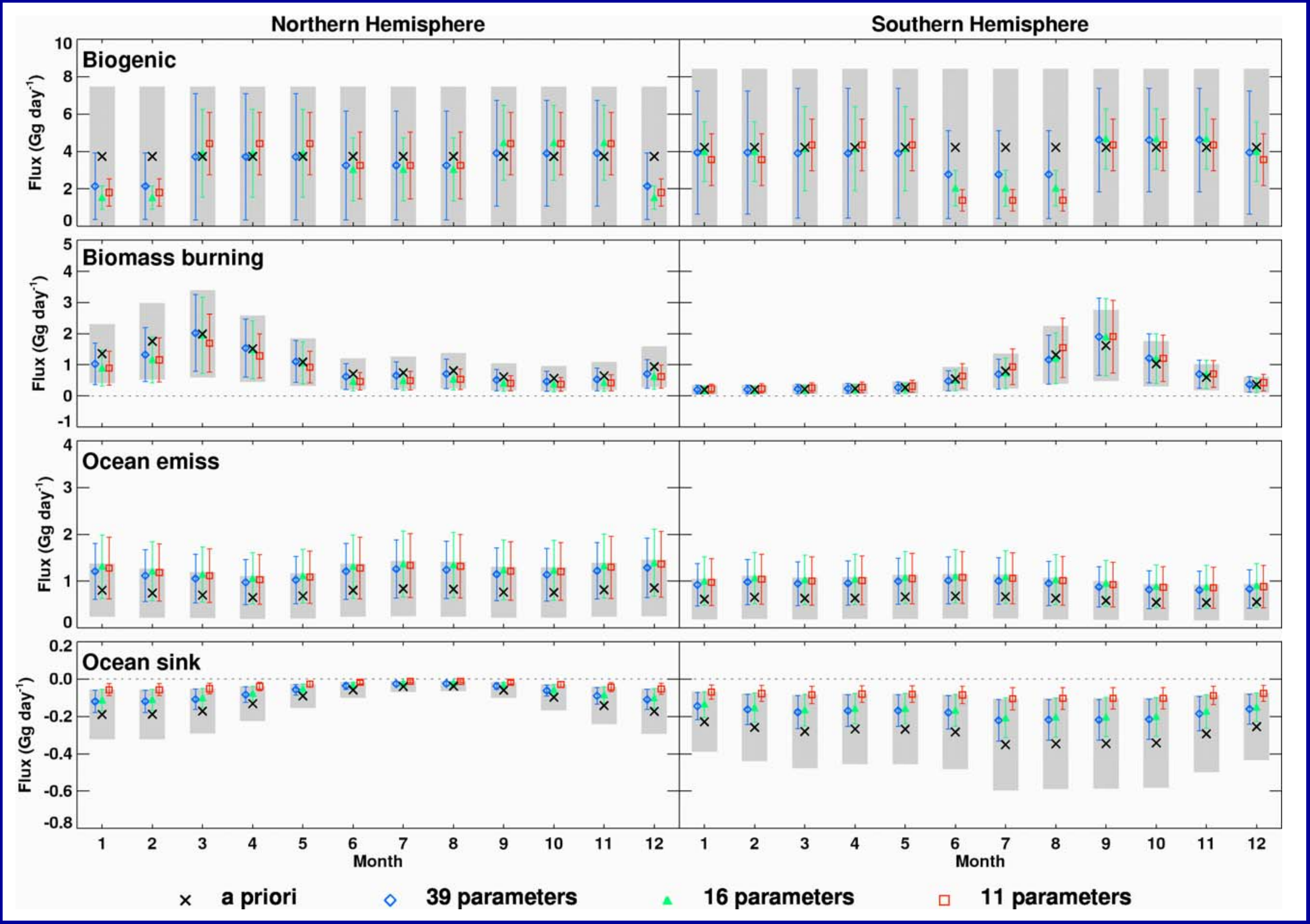
- The GEOS-CHEM global 3-D chemical transport model (v.5.02)
- Resolution: 4 º latitude × 5 º longitude and 26 vertical levels
- CH_3Cl from 6 sources and 2 sinks for various geographical regions in 4 seasons are simulated as individual tracers.
- Biomass/biofuel burning emissions are calculated using CH_3Cl/CO molar emission ratio of 5.7×10^{-4} [Lobert *et al*., 1999] based on 7-year mean GEOS-CHEM biomass burning CO emissions.
- A pseudo-biogenic source of 2,900 Gg yr^{-1} is specified with a flat emission rate from vegetated areas between 30°N and 30°S.
- Oceanic flux is calculated using a NOAA-CMDL empirical relationship between CH_3Cl saturation and sea surface temperature [Khalil *et al*.,1999].
- The OH field is taken from the work by Martin *et al*. [2003].

Inverse modeling

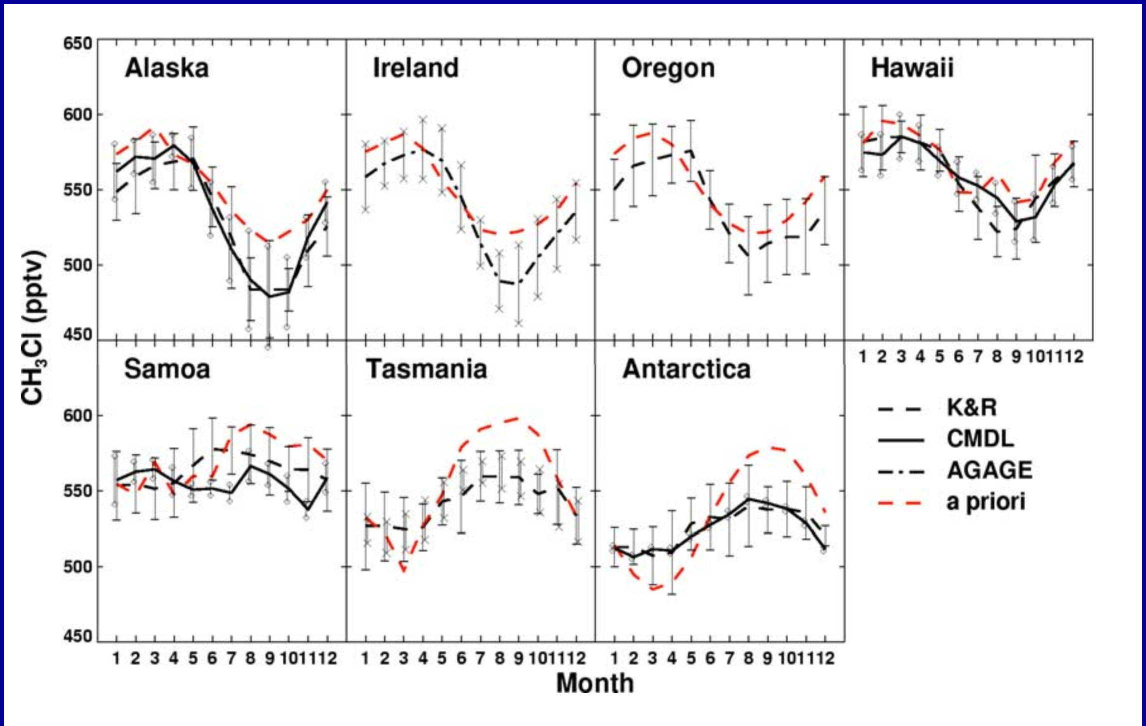
- Inverse modeling is applied using the Bayesian least-squares formulation [Rodgers, 2000].
- The pseudo-biogenic source is simulated for 4 seasons and 6 geographical regions (24 parameters), and biomass burning source for 4 seasons and 2 hemispheres (8 parameters), 39 parameters in total.
- To test a sensitivity to the number of state vector, highly correlated parameters are combined together by inspecting the model resolution matrix (16 and 11 parameter cases are simulated).

Seasonal parameters		Aseasonal parameters	
Biogenic	North America		Ocean emission
	South America		Incineration/industrial
	North Africa		Salt marshes
	South Africa		Wetlands
	Asia		Ocean sink
	Oceania		
Bio burn	Northern hemisphere	Soil sink	Northern hemisphere
	Southern hemisphere		Southern hemisphere

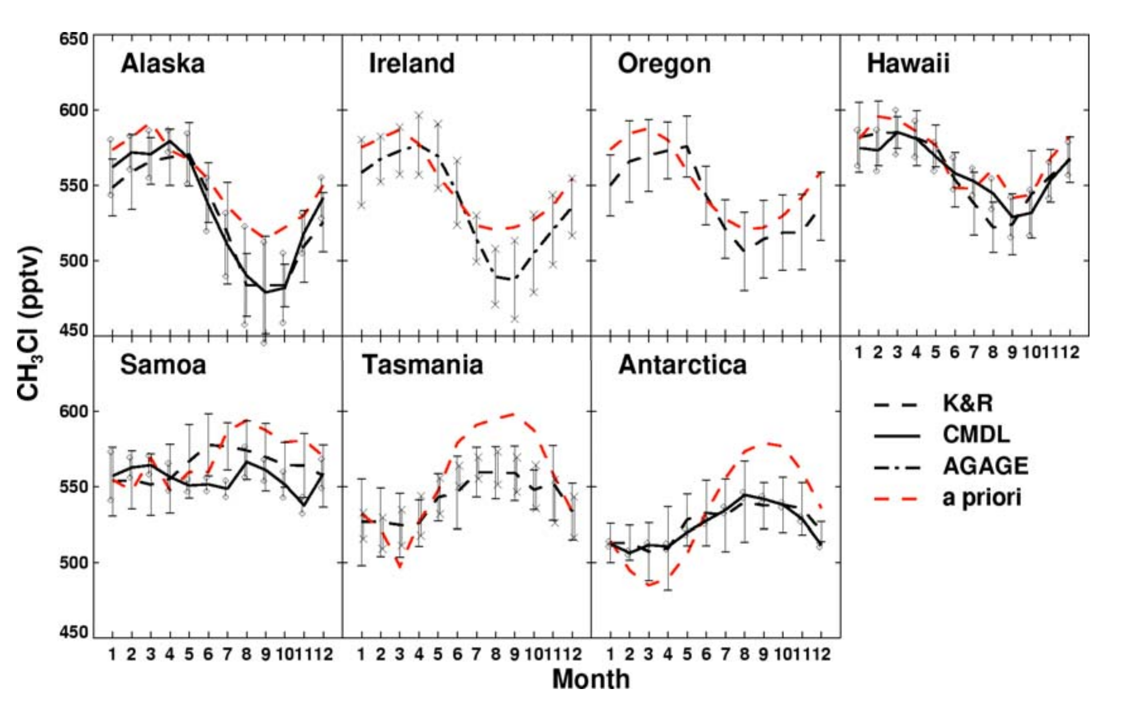
A priori vs. a posteriori flux estimates



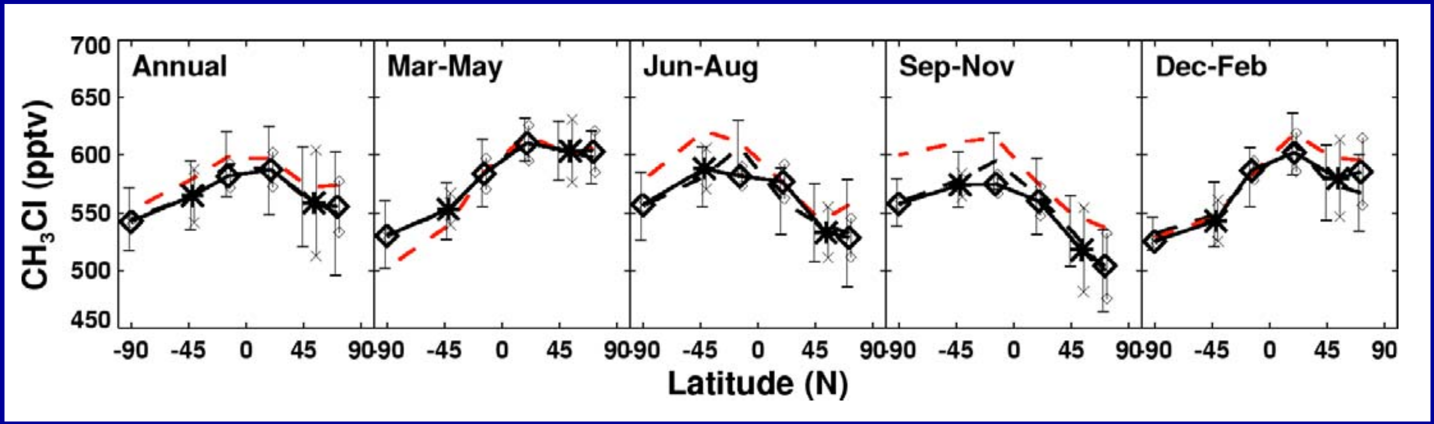
Seasonal variations



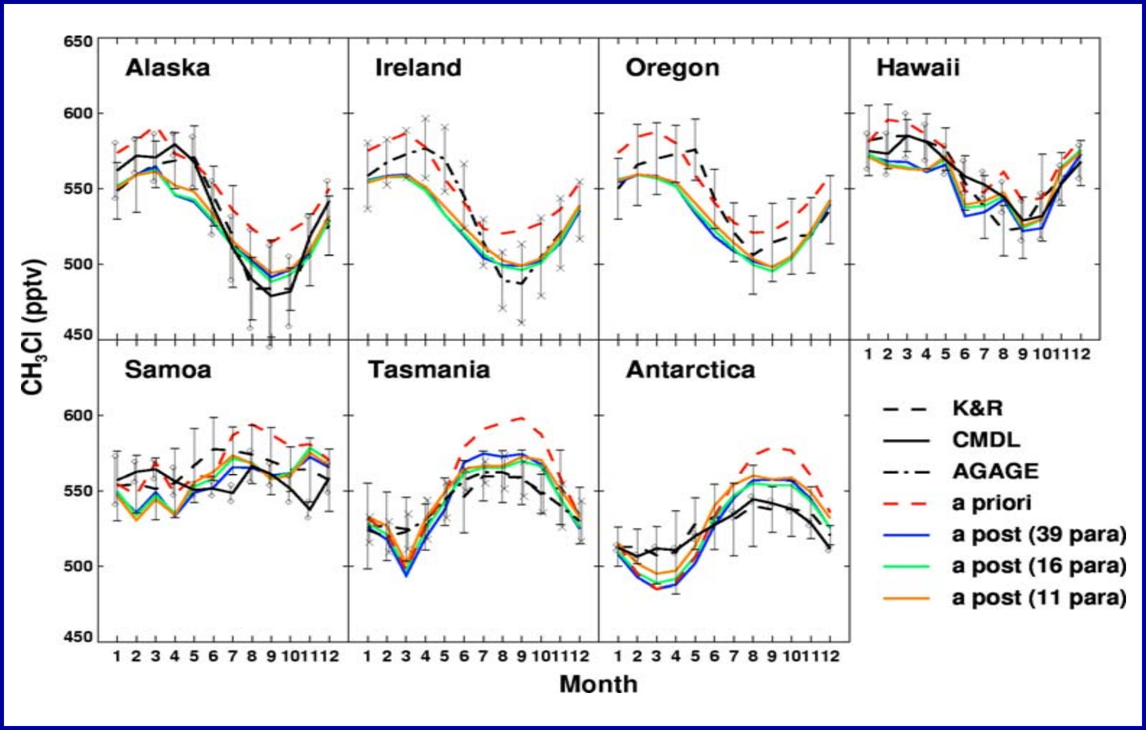
Seasonal variations



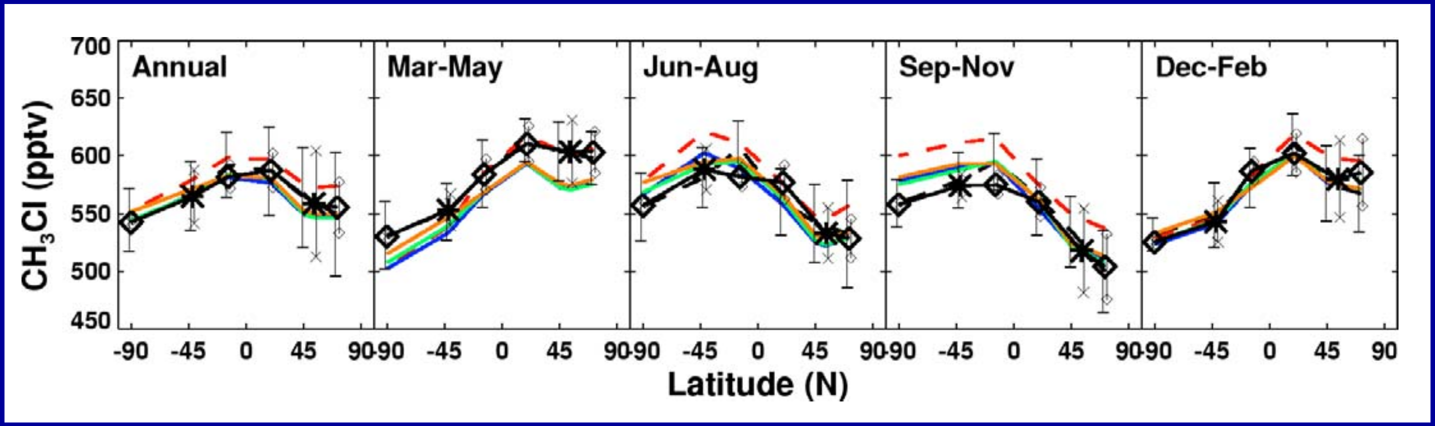
Latitudinal distributions



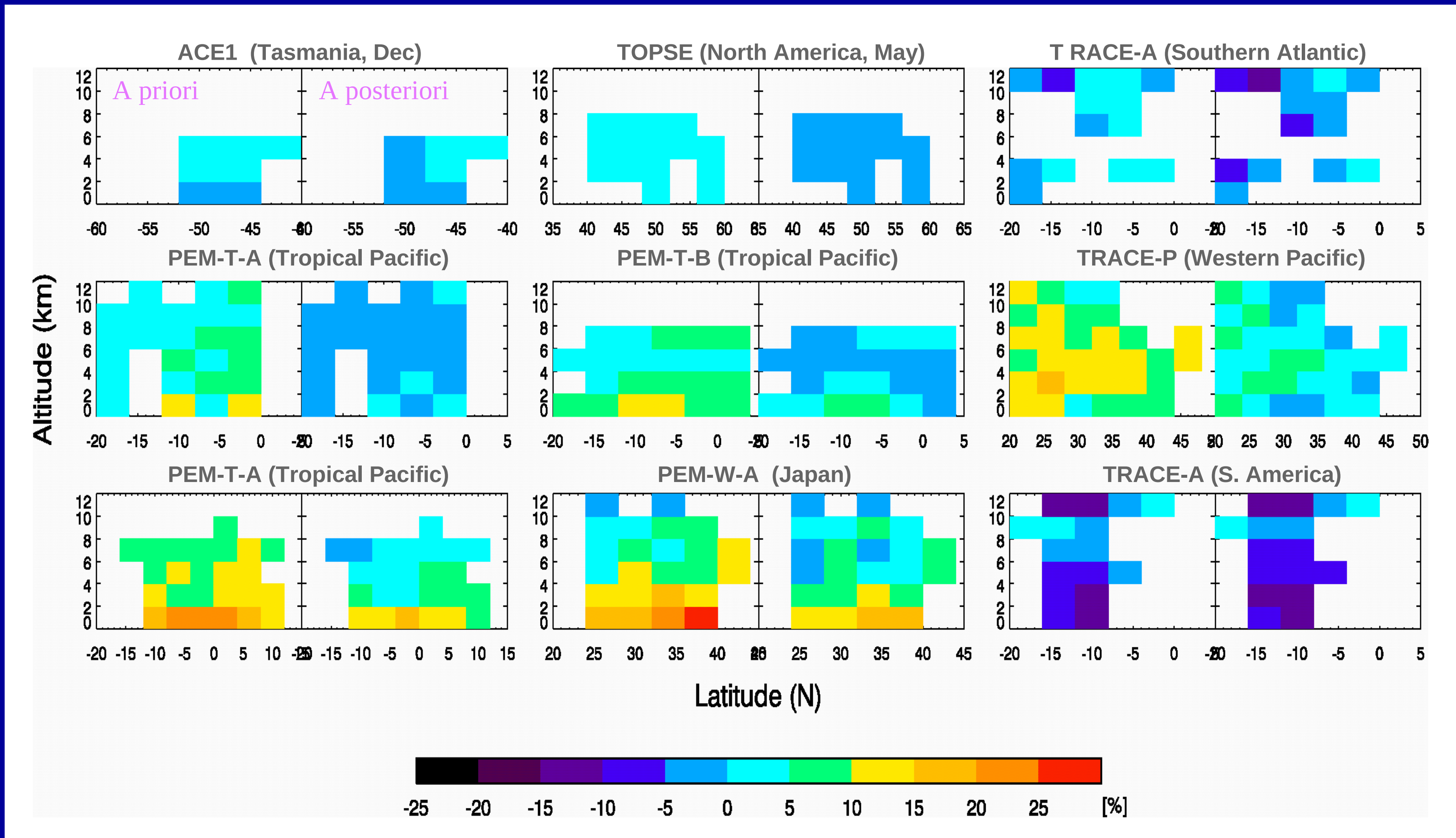
Seasonal variations



Latitudinal distributions



Differences between aircraft measurements and model



Conclusions

- We used an inverse modeling to understand better the seasonal characteristics of CH_3Cl emissions using measurements from seven surface stations and eight aircraft field experiments.
- We conducted sensitivity tests with three different model parameter sizes of 39, 16 and 11, and obtained largely compatible results.
- The apparent effect of changing the state vector size is on the uncertainties of the a posteriori fluxes especially for biogenic source.
- The a posteriori biogenic source of 2.5 Tg yr^{-1} shows a clear winter minimum in both hemispheres; the a posteriori uncertainty is generally 30-40%.
- The a posteriori biomass burning source decreases by 27% to 281 Gg yr^{-1} in the NH but increases by 17% to 264 Gg yr^{-1} in the SH.
- The a posteriori biomass burning emission is 545 Gg yr^{-1}, which is about 11% less than the a priori estimate.
- The a posteriori net ocean source increases by about a factor of 2 to 761 Gg yr^{-1}.

Acknowledgments

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CH_3Cl sources, sinks and their uncertainies

		A priori flux (Gg yr^{-1})	Reference	39 parameters			16 parameters			11 parameters		
				A posteriori flux (Gg yr^{-1})	A priori uncertainty (%)	A posteriori uncertainty (%)	A posteriori flux (Gg yr^{-1})	A priori uncertainty (%)	A posteriori uncertainty (%)	A posteriori flux (Gg yr^{-1})	A priori uncertainty (%)	A posteriori uncertainty (%)
Biogenic	NH spring	343[a]		342	100	91	359	100	61	406	100	38
	NH summer	343[a]		299	100	90	279	100	56	299	100	55
	NH fall	340[a]		355	100	73	407	100	45	401	100	38
	NH winter	336[a]		192	100	84	138	100	40	162	100	40
	NH total	1362[a]		1188			1183			1268		
	SH fall	388[a]		359	100	89	381	100	55	399	100	32
	SH winter	388[a]		254	100	85	187	100	47	127	100	42
	SH spring	383[a]		420	100	60	424	100	35	395	100	32
	SH summer	379[a]		354	100	84	359	100	40	320	100	39
	SH total	1538[a]		1387			1351			1241		
Biomass burning	NH spring	141[a,b]		143	70	61	138	70	63	119	70	55
	NH other seasons	244[a,b]		197	70	67	160	70	64	162	70	61
	SH spring	98[a,b]		115	70	65	115	70	66	116	70	62
	SH other seasons	127[a,b]		118	70	69	115	70	68	148	70	62
Ocean		507[a,c]	805[c]	760	70[c]	49	824	70[c]	52	806	70	51
Incineration/industry		162[d]	162[d]	129	80[d]	66	98	100	63	49	100	56
Salt marshes		170[e]	170[e]	96	100	91	103	100	63	51	100	56
Wetlands		48[f]	48[f]	41	100	98	29	100	63	14	100	56
Total source		4397	4525	4173			4116			3974		
Ocean sink		149[a,c]	150[c]	94	70[c]	50	88	70[c]	51	45	100	56
Soil sink		256[g]	256[g]	234	100	85	179	100	83	77	100	56
OH sink		3992[a]	3500[i]	3845			3848			3852		
Total sink		4397	3906	4173			4116			3974		

[a]*Yoshida et al*. [2004]
[b]*Lobert et al*. [1999]
[c]*Khalil et al*. [1999]
[d]*McCulloch et al*. [1999]
[e]*Rhew et al*. [1999]
[f]*Varner et al*. [1999]
[g]*Khalil and Rasmussen* [1999] and *Keene et al*. [1999]
[h]*Lee-Taylor et al*. [2001]
[i]*Koppmann et al*. [1993]