Environmental Transport and Fate of SOCs - Global Multicompartmental Modelling Using a GCM

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Motivation

1. Fundamental Science: Environmental chemistry on large spatial scales
2. Assist environmental risk assessment: State-of-the-art characterization of substance environmental fate (P_{ov}, LRTP)

Approach

Interdis

Global Multicompartment modelling

Model Application and Results

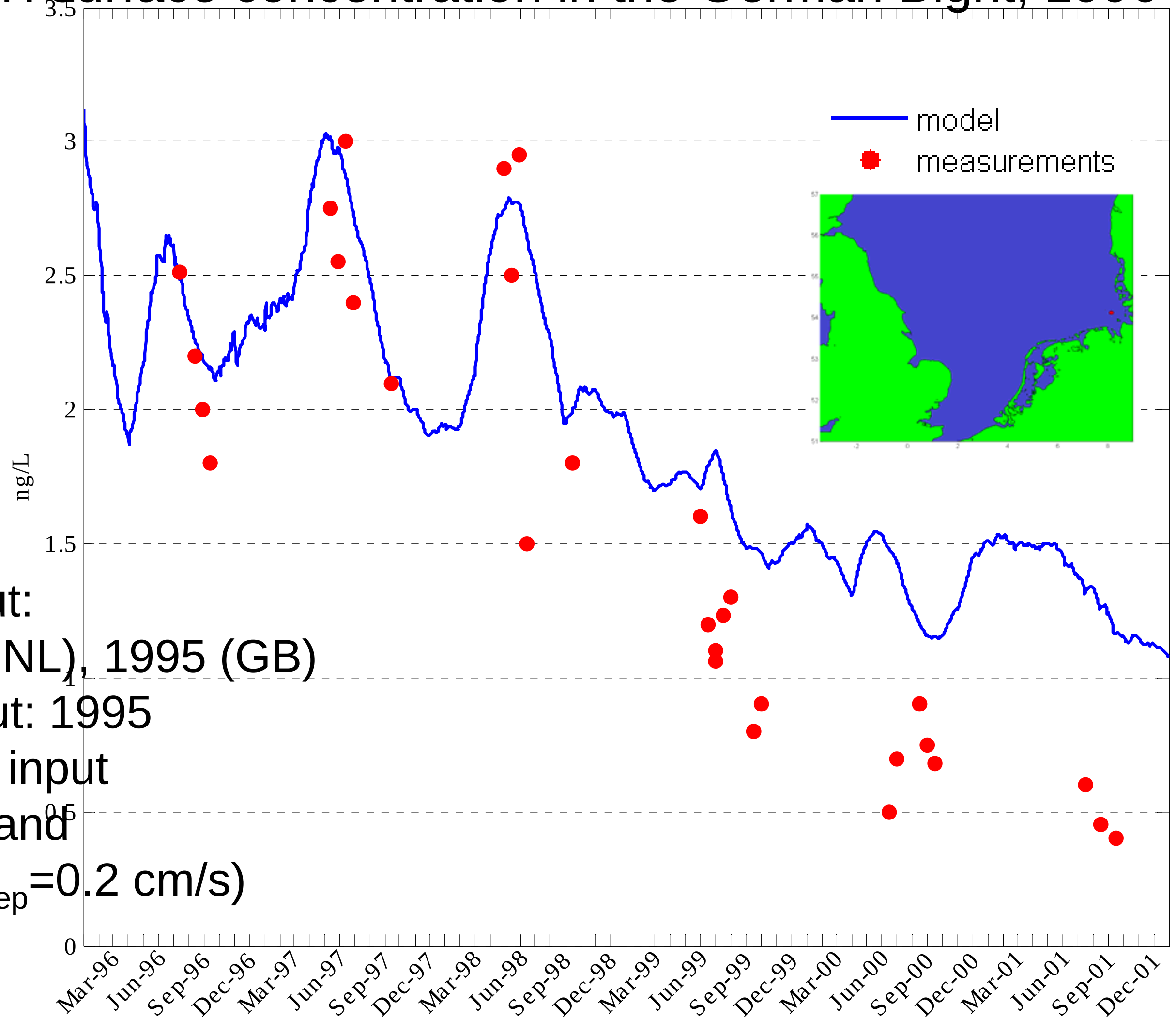
Geographic and compartmental distributions

'multi-hopping'

Substance characterisation: Persistence, long-range transport potential

State of Knowledge: Conclusions

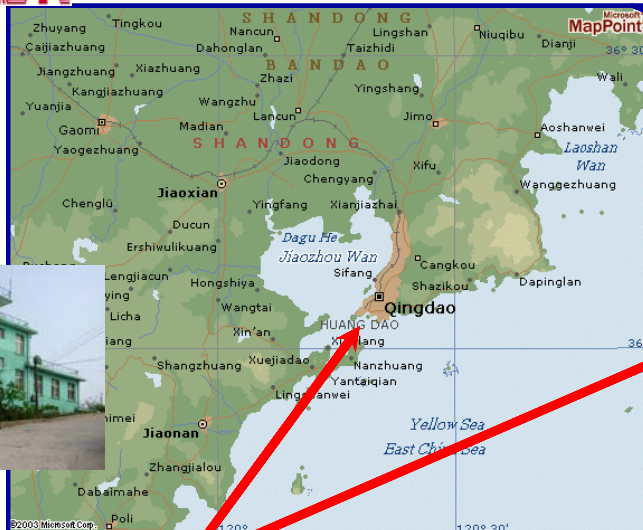
γ-HCH surface concentration in the German Bight, 1996-2001:

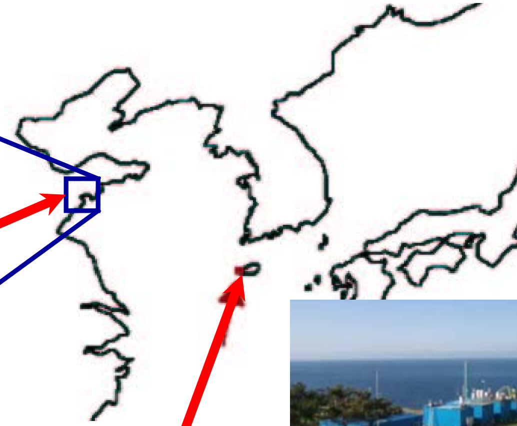


Riverine input:
transient (D, NL), 1995 (GB)
Channel input: 1995
Atmospheric input
wet: Westerland
dry: Lista (v_{dep}=0.2 cm/s)

(Ilyina, Pohlmann et al., 2005)

Yellow Sea campaign, 2003





Coastal Site
Qingdao, Shandong, China

Island Site
Gosan, Jeju Island, Korea

Preliminary Results

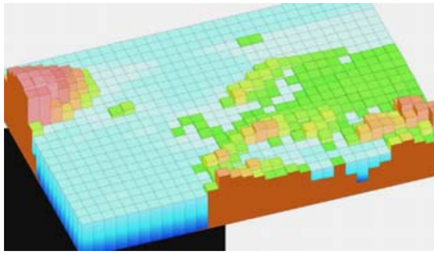
	Coastal Site		Island Site
HCHs	205 pg/m³ (α/γ=1.2)	HCHs	125 pg/m³ (α/γ=0.95)
PCBs	115 pg/m³	PCBs	2.4 pg/m³
DDTs	90 pg/m³ (DDE/DDT=0.8)		
Ca	1270 ng/m³	Ca	58 ng/m³
Cu	44 ng/m³	Cu	1.7 ng/m³
Pb	111 ng/m³	Pb	4.3 ng/m³

(Lammel, Ghim et al., 2005)

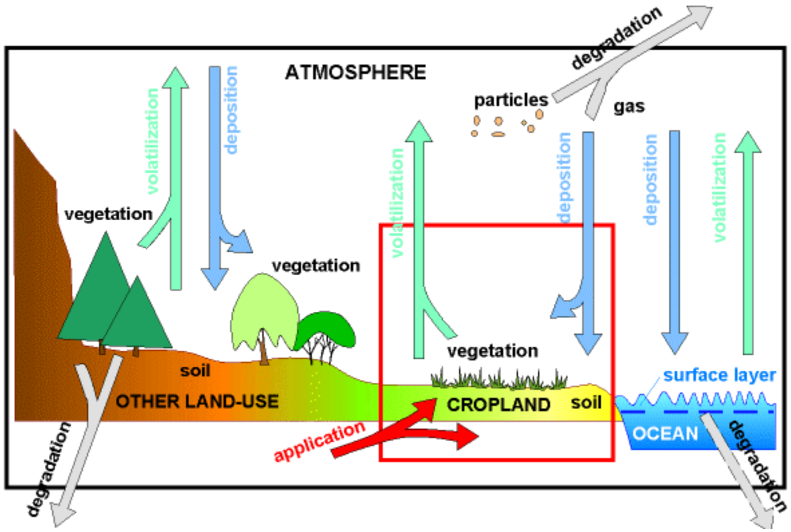
Approach

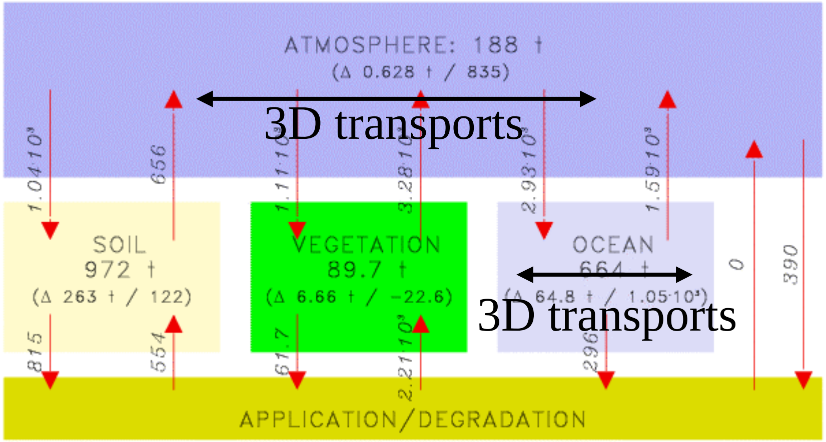
Interdisciplinary Shelf Sea Research

Global Multicompartment Modelling



- Multicompartment chemistry transport model (MPI-MCTM: Lammel et al., 2001) based on the atmosphere general circulation model (AGCM) ECHAM5 (Roeckner et al., 2003)
- Resolutions: T42 horizontal resolution (2.75*2.75°), 19 vertical levels, time step = 30 min, processes: equilibrium and time-resolved (pseudo-1st order kinetics) intra- and intercompartmental mass exchange processes
- ground compartments gain by application, deposition,and lose due to degradation, volatilisation and transfer to lower levels





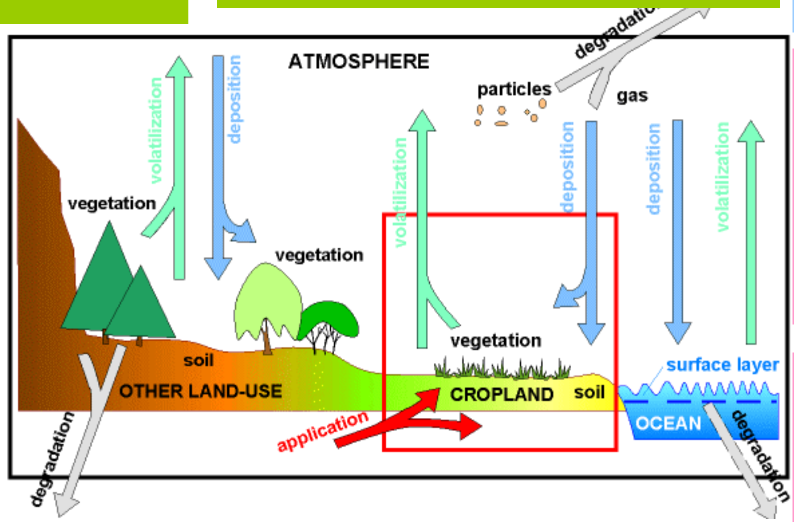
Approach (cont'd): Intracompartmental intercompartmental mass exchange processes

Degradation: Reaction with hydroxyl radical (and nitrate radical during the night), prescribed radical distributions (Roelofs & Dentener, 1997)

g/p partitioning: Empirically based adsorption **or** absorption, based on vapor pressure (Pankow & Bidleman, 1992) or K_{oa} (Finizio et al., 1997)

Aerosols: Prescribed climatology (GADS; Koepke et al., 1997) or fully dynamic microphysics (HAM; 4 size modes, 7 species; Stier et al., 2004)

Volatilisation from soils/vegetation: Loss of gaseous substance from the soil pore space/vegetation surface using empirically derived rates from pesticide application studies f(T, c_{om} , w_{soil} , w_{max}), K_{lg}, K_{sl}, p (Smit et al., 1997, 1998)



Dry deposition of g/p: Fixed deposition velocities v_{dep} or resistance scheme for gaseous molecules (Ganzeveld et al., 1997)/fixed for particle f(r)

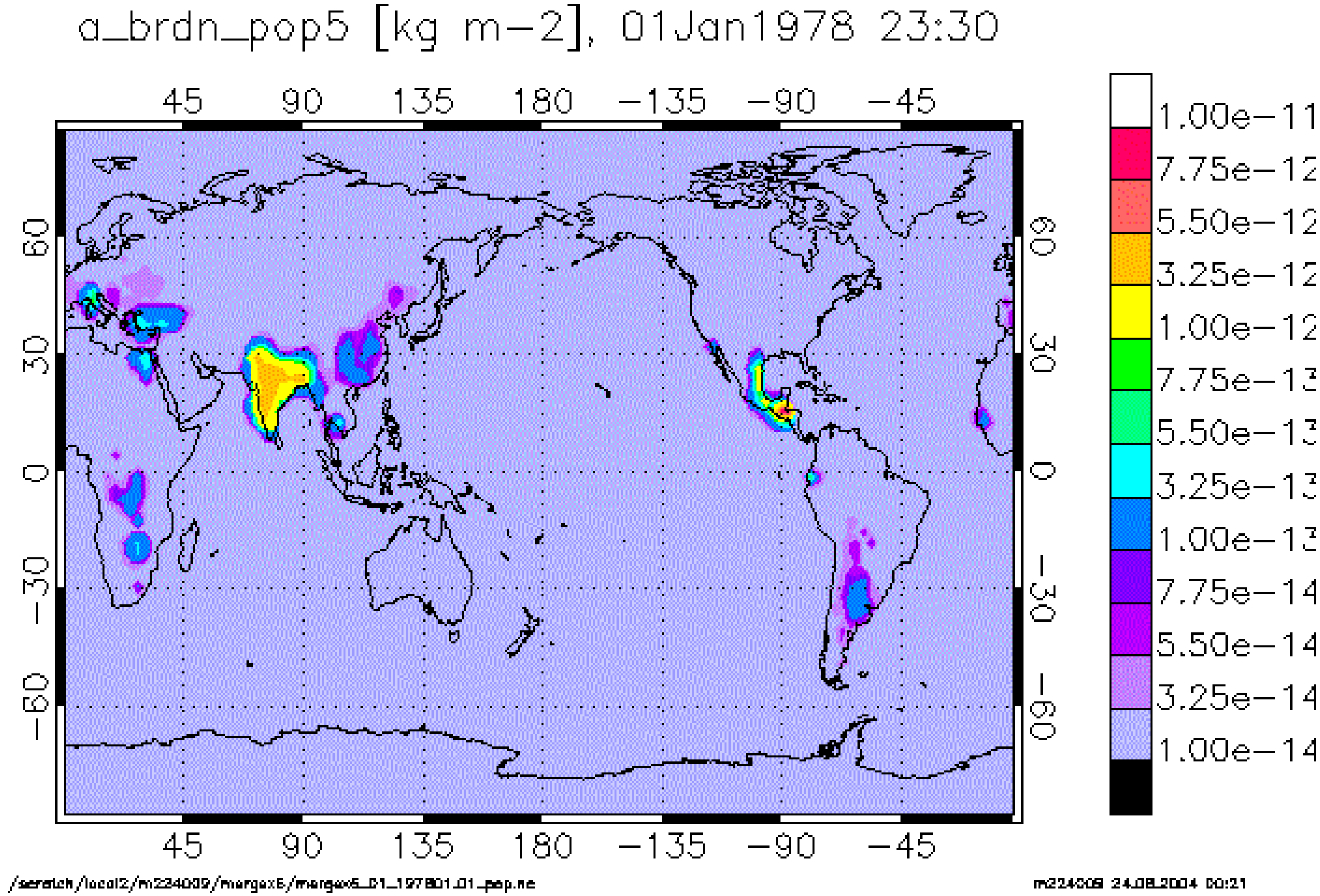
Air/sea exchange: Well mixed surface layer, stagnant film model

Degradation: Overall first-order rates, assumed to double per 10 K temperature increase

Partitioning in soils: Phase equilibrium in 3-phase soil system (soil hydrology represented by bucket model; Roeckner et al., 1996)

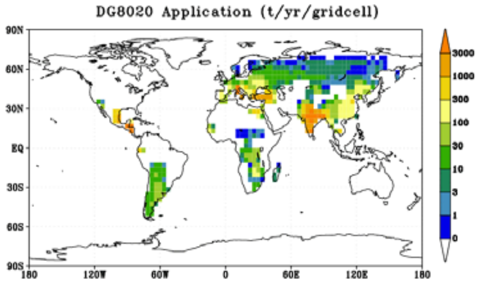
Approach (cont'd):

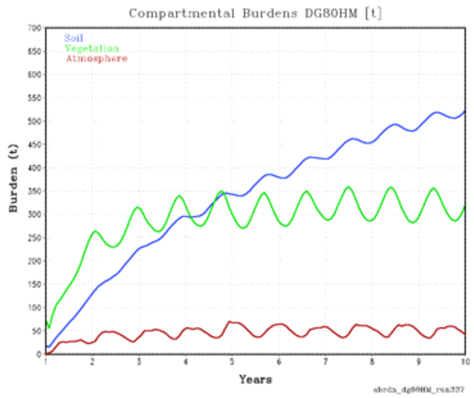
- 20-70 CPUh per simulated year on high-performance computing system NEC-SX6
- Entry of DDT into the model world: Application to ground compartments (here: vegetation 80%, soils 20%, according to 1980 global usage; FAO data), transport upon volatilisation according to local soil and air conditions

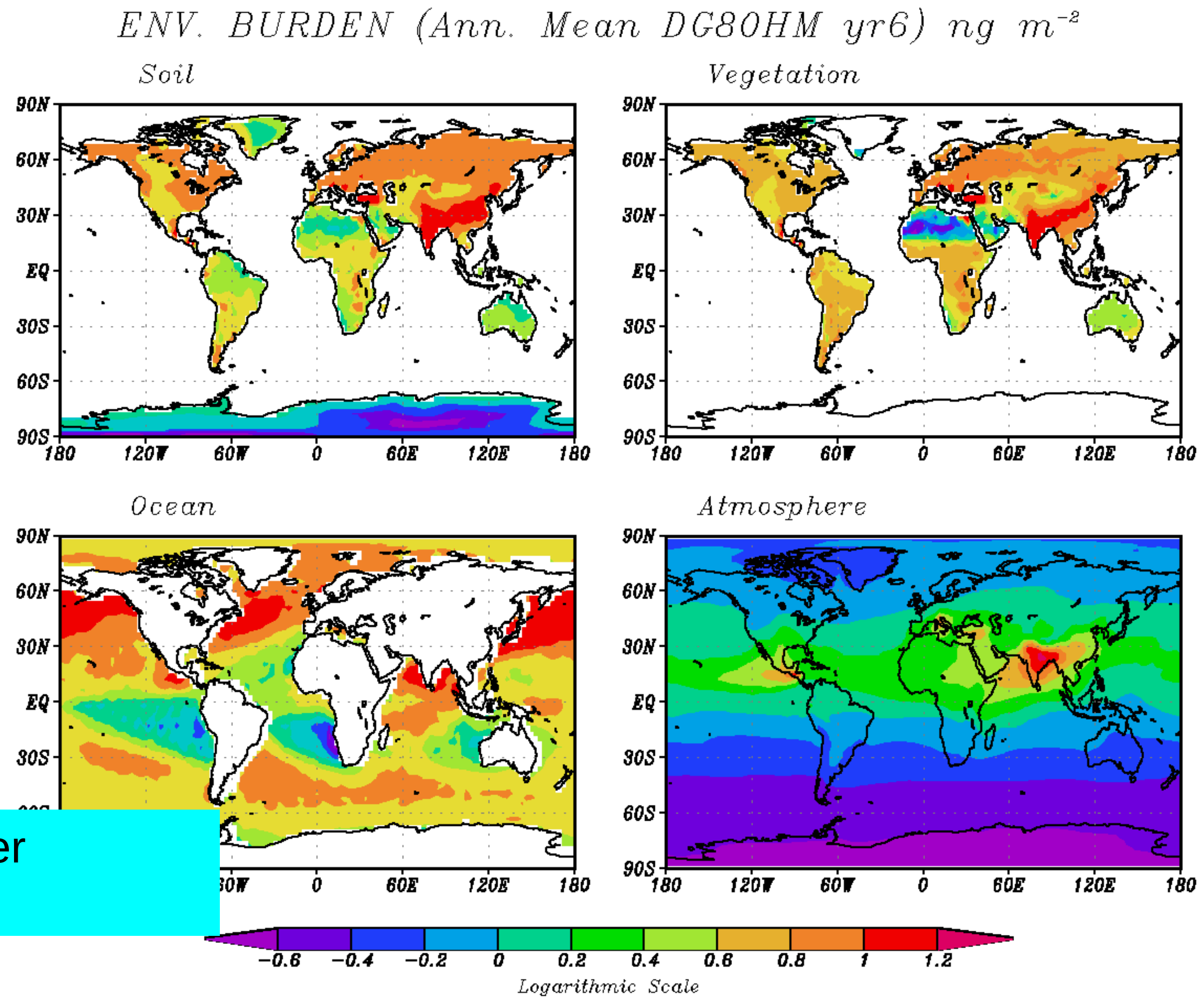


Multicompartmental distribution

1980 global usage DDT (Feb-Aug vegetation/soil/air = 80:20:0):



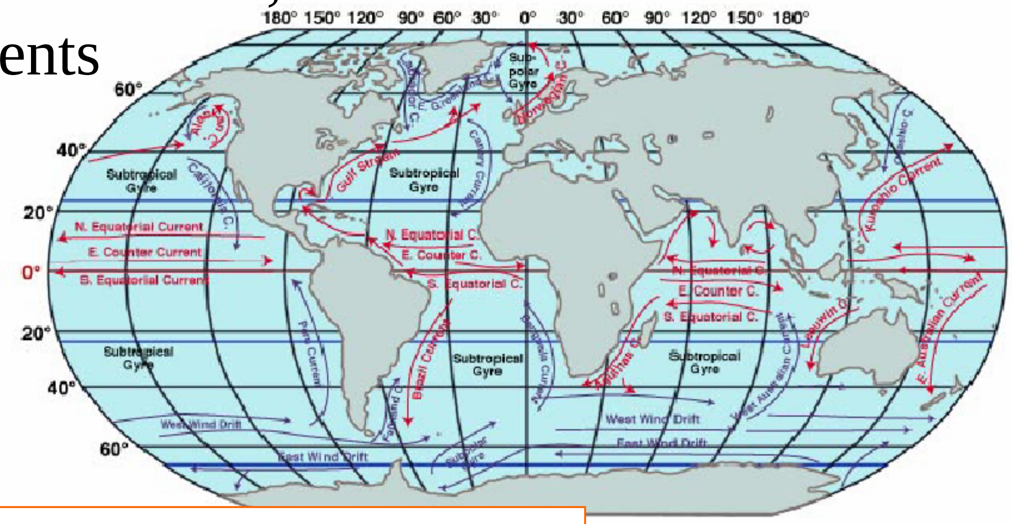




2 layer (surface mixed layer
+ deep sea) ocean

Coupling of the 3D ocean model, MPI-OM1, wind and salt-gradient driven currents

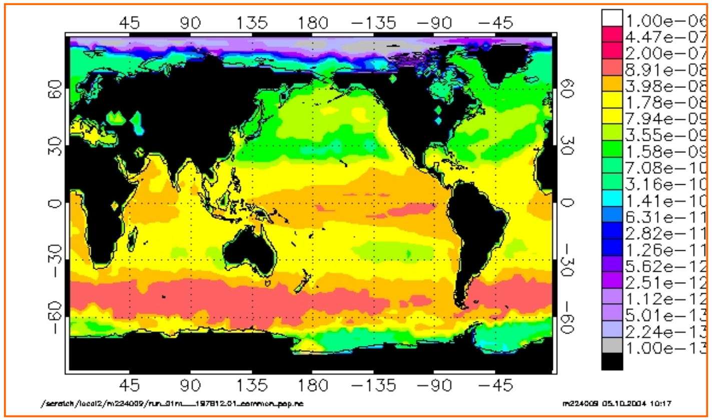
(Guglielmo, Maier-Reimer & Lammel)



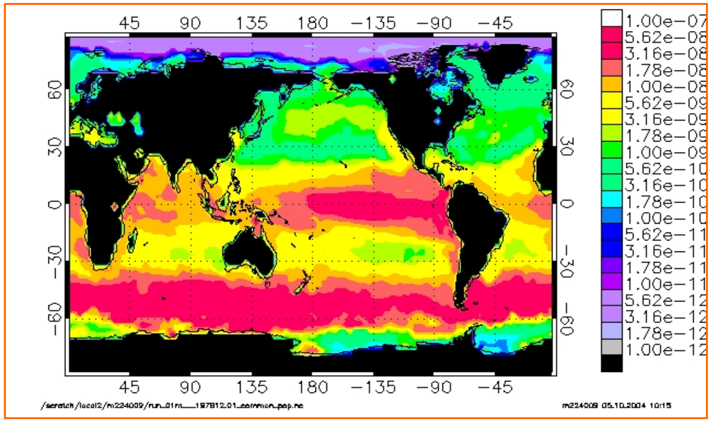
...and a biogeochemcal model,

HAMOCC: Nutrients, plankton, DOC, POC

(Six & Maier-Reimer, 1996)



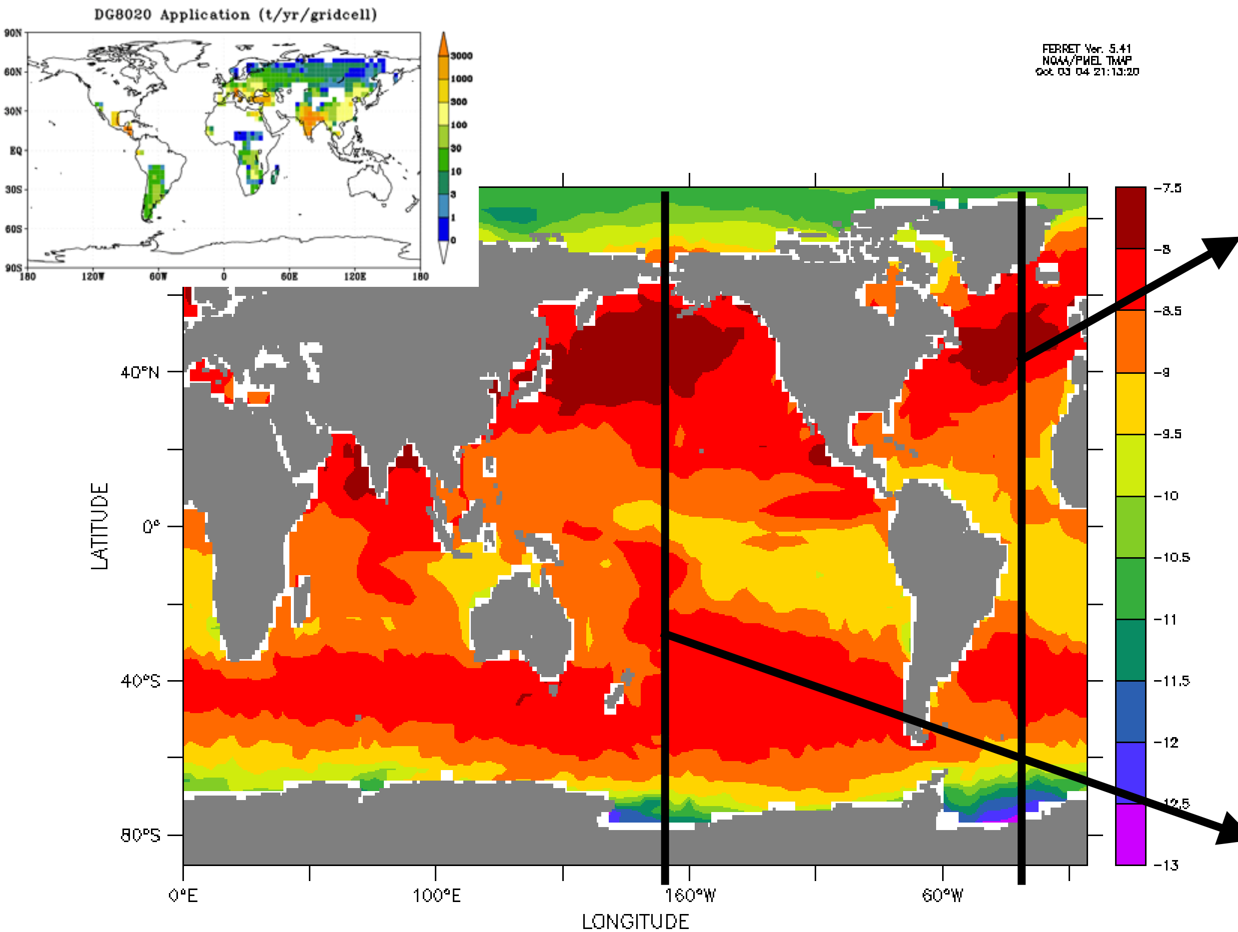
colloidal phase (kmol/m^3)

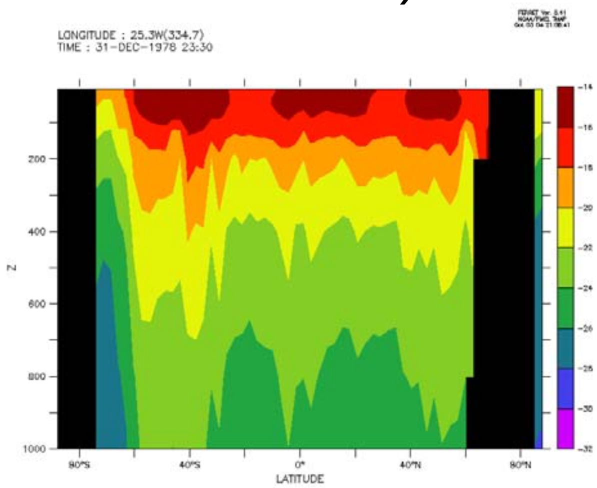


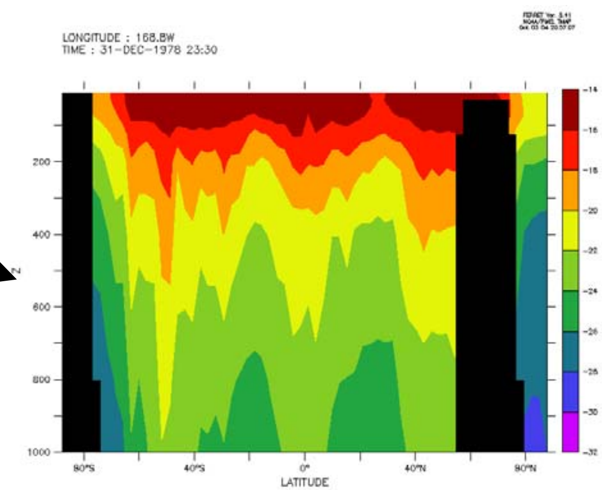
POM phase (kmol/m^3)

Cycling in the multicompartment system

1980 global usage DDT (Feb-Aug vegetation/soil/air = 80:20:0):







log(conc)

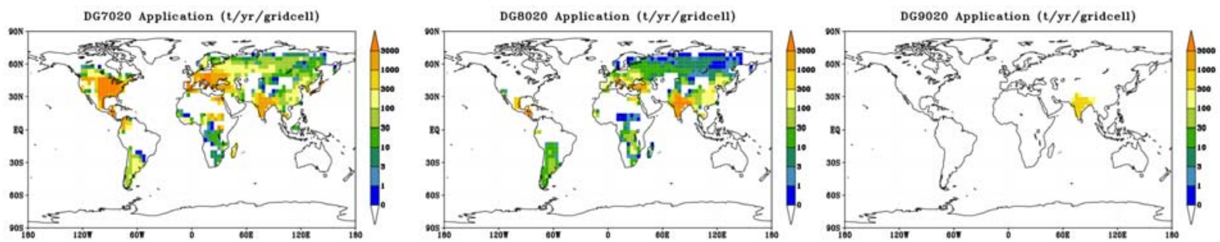
Compartmental distribution: Historic change

DDT, annual means, 2^{nd} year

Indicator for the long-range transport potential (LRTP)

Zonal width: ZW_{90} distance between 5 and 95%ile of zonal distribution

Zonal spreading: $ZS_{90}(t) = ZW_{90}(t) - ZW_{90}(t_0)$

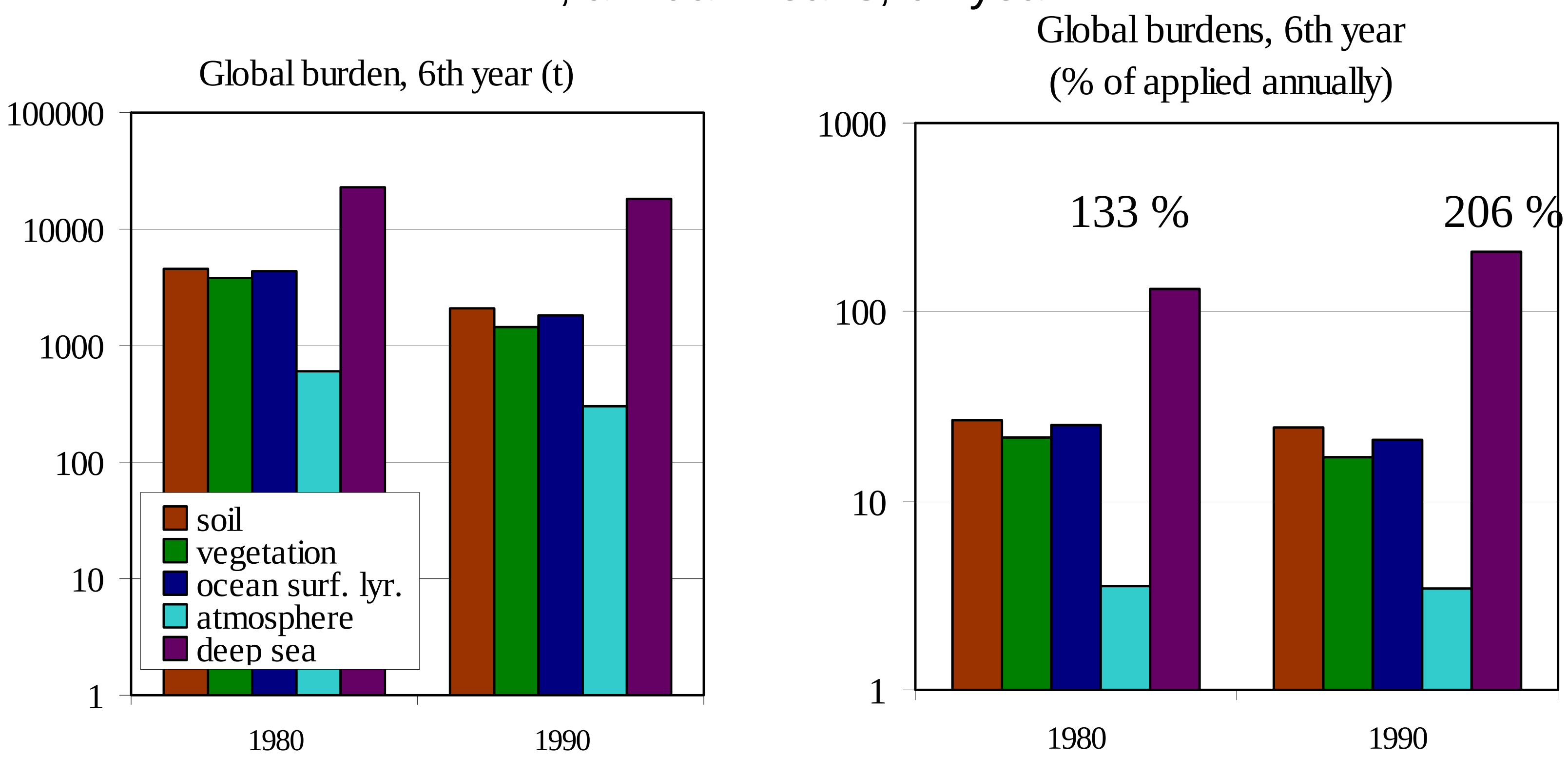


	1970	**1980**	**1990**
c_{air}/c_{total}	1.5	2.8	3.7 %
c_{ocean}/c_{total}	29	15	18 %
$P_{overall}$	550	660	540 d
ZS_{90}	520	1610	700 km

(Semeena & Lammel, 2003)

Compartmental distribution: Historic change

DDT, annual means, 6th year



Global DDT emissions in 1990 (dominated by usage in India) were suggestively less subject to transport into the Arctic and more subject to transport into the deep sea

Characterization of substance environmental fate

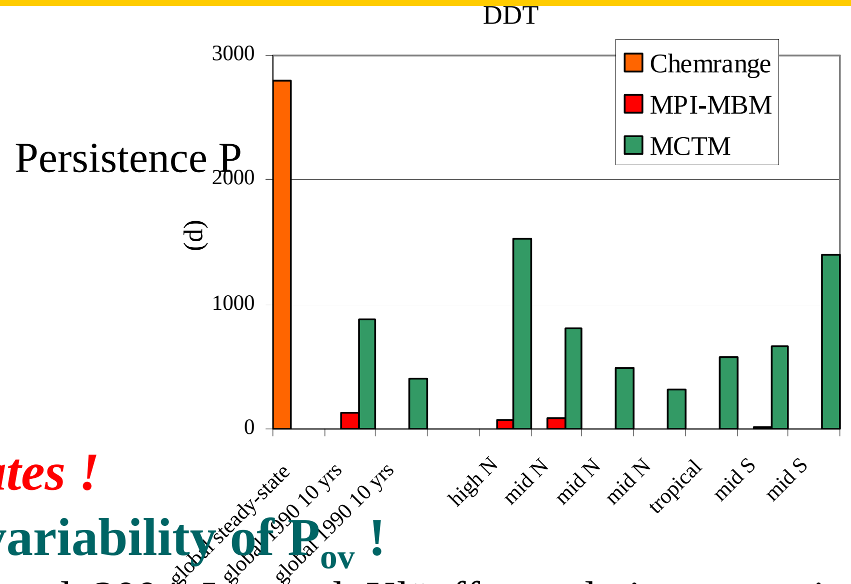
P_{ov} and P_{ov}-ranking are not independent of

...(1)geo-referencing and location of entry (emission):

Model intercomparison

Box models *vs.* ***GCM-based MCTM***

release into soil/vegetation scenarios



Partly underestimates, partly overestimates !

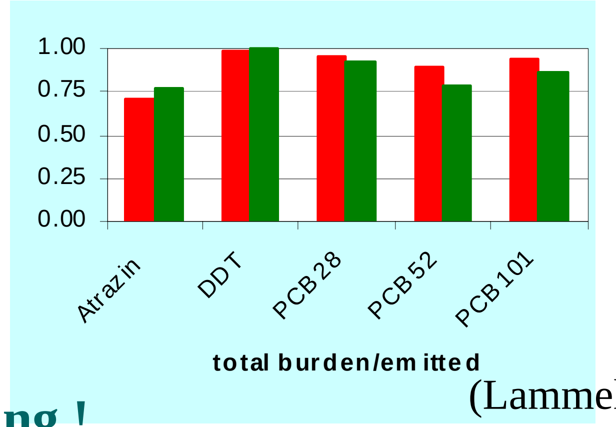
→ Location of entry introduces large variability of P_{ov} !

(Leip & Lammel, 2004; Lammel, Klöpffer et al., in preparation)

...(2)the temporal resolution:

constant mean value *vs.* ***time-resolved fit to observational data*** *(Δt = 1h)*

MPI-MBM, release into air scenario

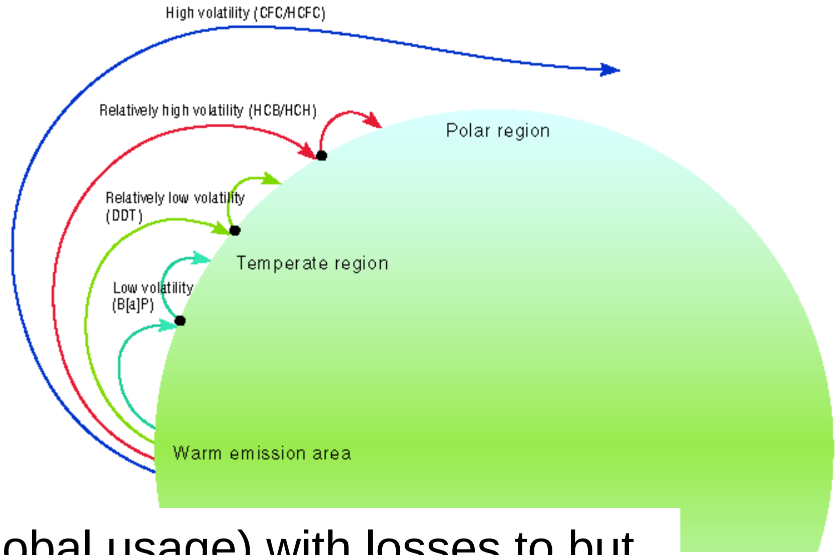


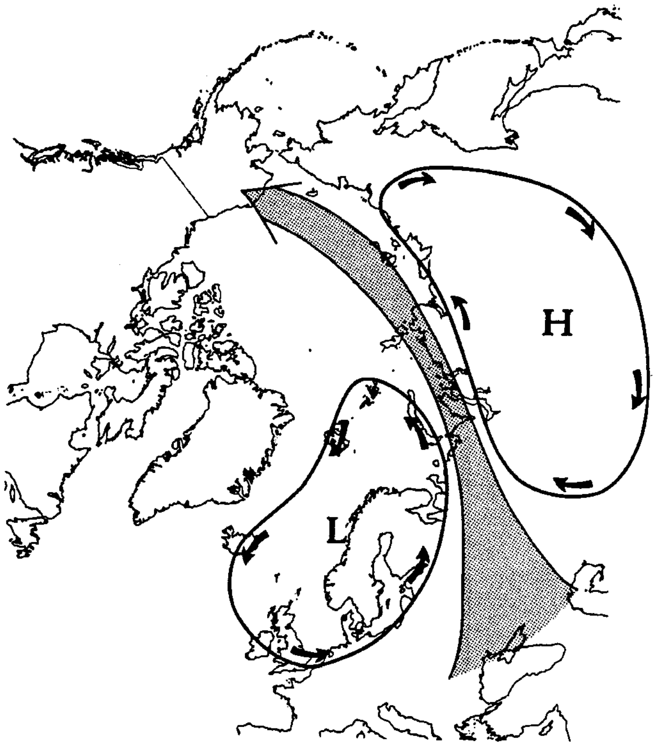
(Lammel, 2004)

Partly underestimates, partly overestimates !

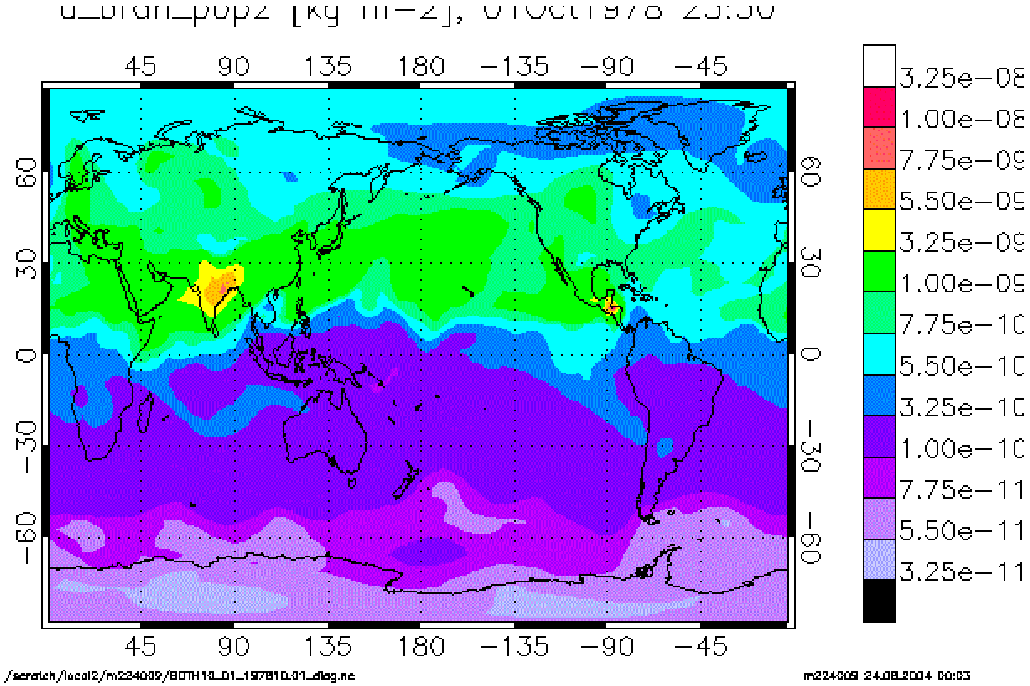
→ Steady-state / time averaging matters for ranking !

Long-range transport into the Arctic by singlehop or multihop ?





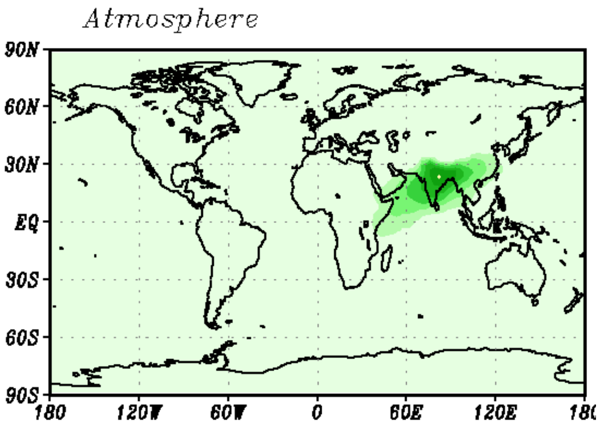
DDT (1980 global usage) with losses to but no re-gain from ground compartments

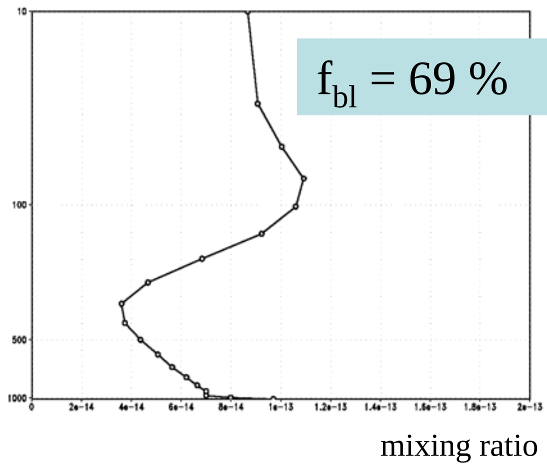


Longyearbyen (Foto: Rahmstorf)

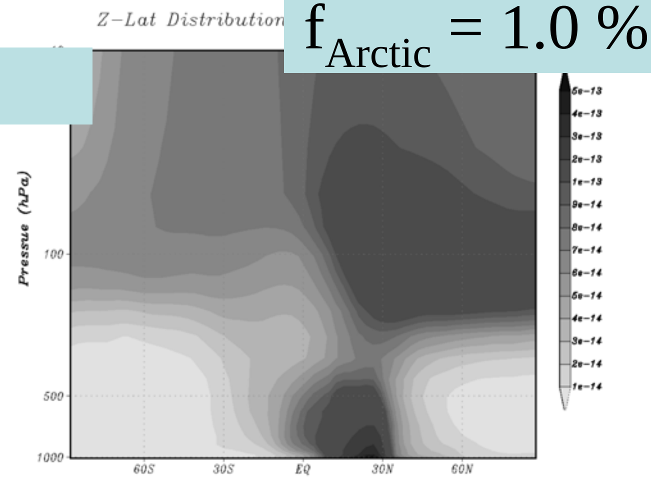
DDT, 1990 global usage, global annual mean

multi-hopping + single-hopping DDT:



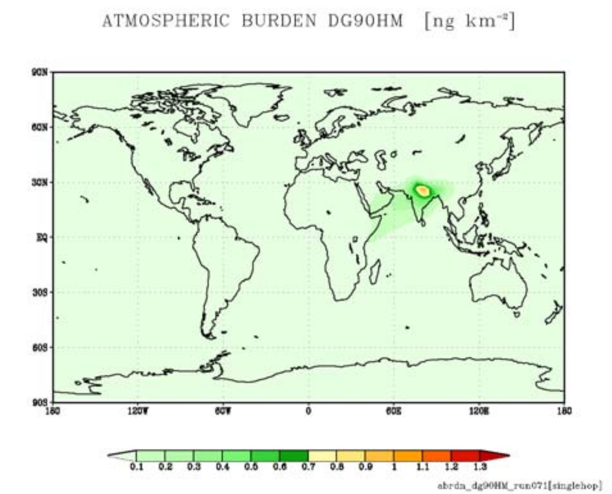


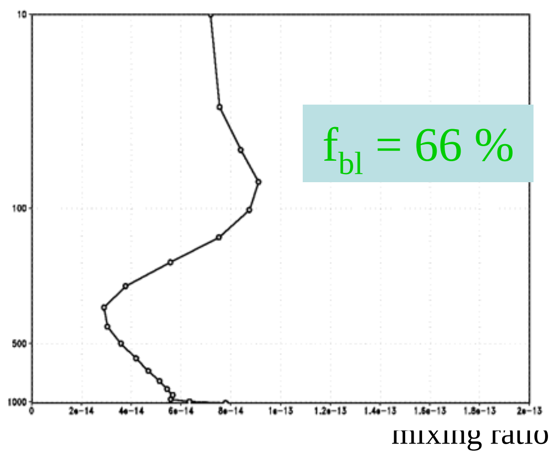
f_{bl} = 69 %



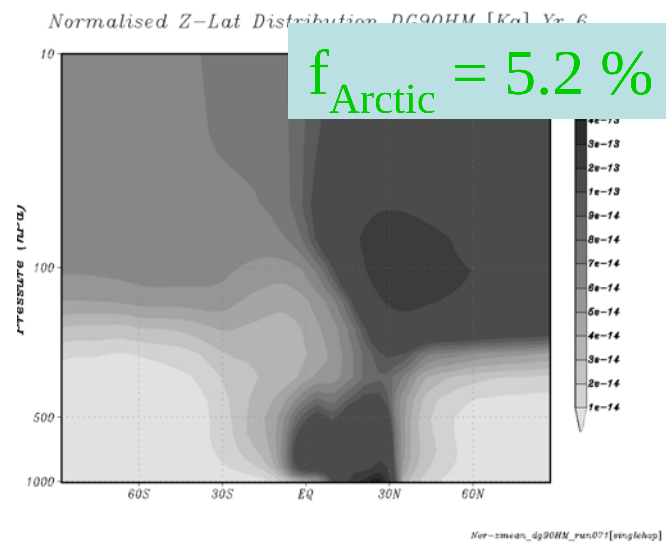
f_{Arctic} = 1.0 %

single-hopping:





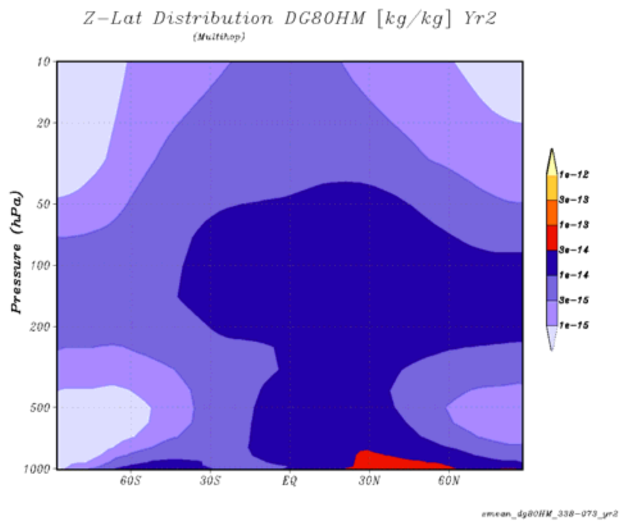
f_{bl} = 66 %

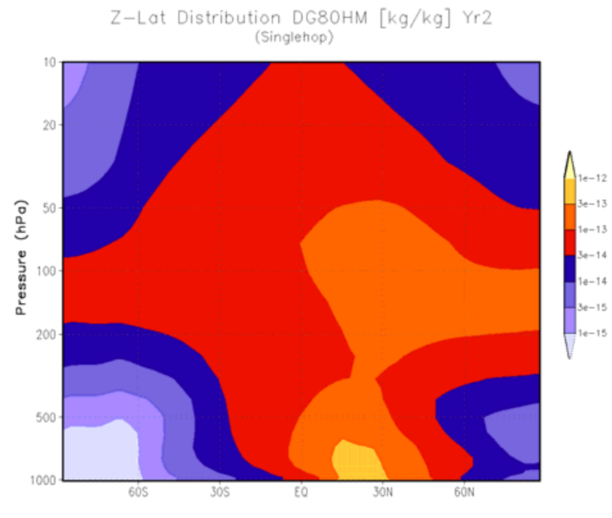


f_{Arctic} = 5.2 %

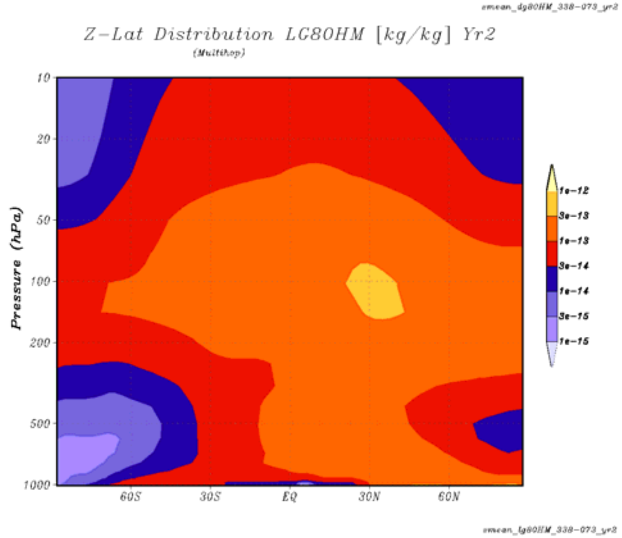
DDT and γ-HCH, 1980 global usage, 2nd yr
multi-hopping vs. single-hopping

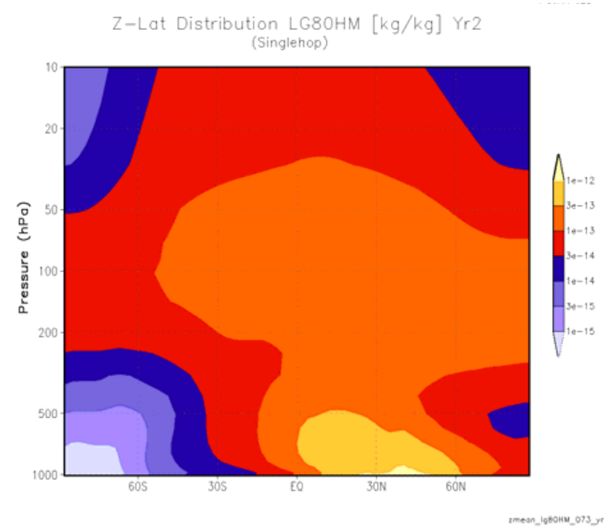
DDT





γ-HCH





(Semeena & Lammel, 2005)

DDT and γ-HCH, 1980 global usage multi-hopping vs. single-hopping

	DDT		γ-HCH	
	Multi-hopping	Single-hopping	Multi-hopping	Single-hopping
Total burden (t)	95	520	240	1000
Burden in the Arctic / Antarctic (t)	7 / 2	22 / 9.9	32 / 4	53 / 14
Burden in the boundary layer / stratosphere (t)	39 / 12	172 / 78	112 / 26	418 / 111
Total depositions (t)	2112	6540	6169	16010
Zonal /spatial spreading (km)	6987/ 5516	11982 / 7786	5794 / 4871	9834 / 7999
Deposition in the Arctic / Antarctic (t)	353 / 46	347 / 7.2	2038 / 85	2720 / 11
Ratio of depositions in the Arctic and Antarctic over their area share	2.1	0.7	4.1	2.1

→ Older and single-hopping substance is transported higher in the atmosphere

→ Expectedly, multi-hopping enhances persistence and long-range transport potential, but LRTP of multihopping < LRTP of singlehopping

→ Single-hopping substance reaches the Arctic and Antarctic, - lindane is even accumulated !

(Semeena & Lammel, 2005)

State of Knowledge: Conclusions

1. The discipline environmental chemistry is still in its infancy (processes, rates, validation)

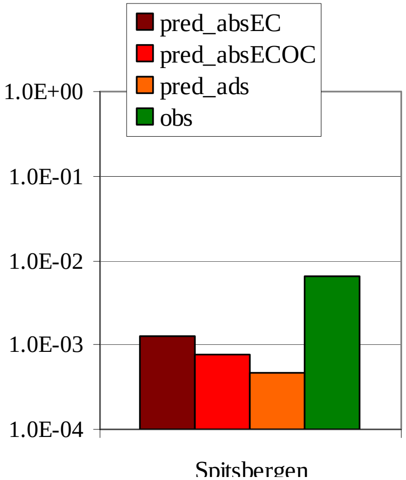
2. Major deficiencies relate to:
   - Emissions (usage) spatial and temporal patterns
   - Degradation in terrestrial and marine compartments, its temperature dependence
   - Gas/particle partitioning in air

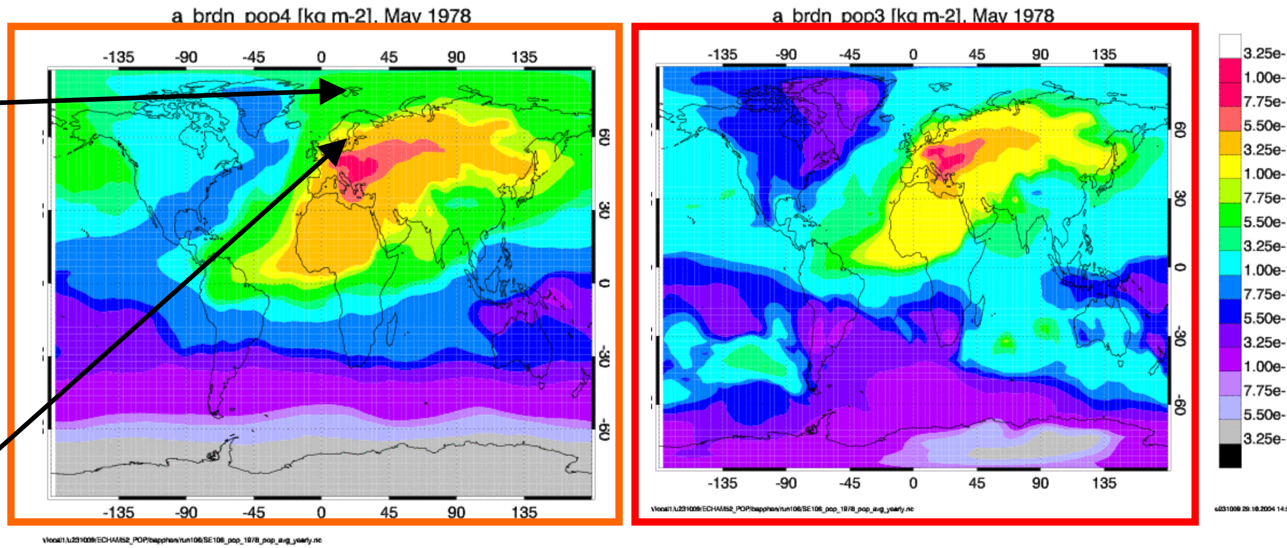
→ **Laboratory/greenhouse work**

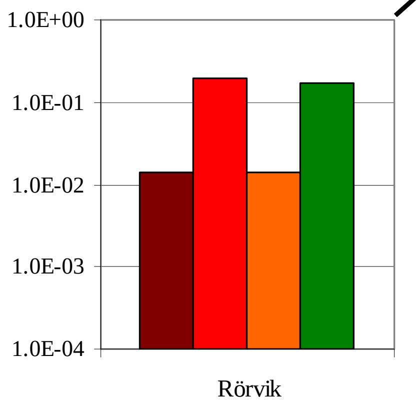
→ **Integrated approaches of measurement and modelling (include the tropics)**

Preliminary results: Gas/particle partitioning and LRT

B[a]P -Adsorption -Absorption







- PAHs are hemispherically distributed
- the contributions by adsorption and absorption to PM are not additive

(Lohmann & Lammel, 2004;
Lammel, Lohmann & Semeena, in preparation)