Biogeochemical effects of aerosols

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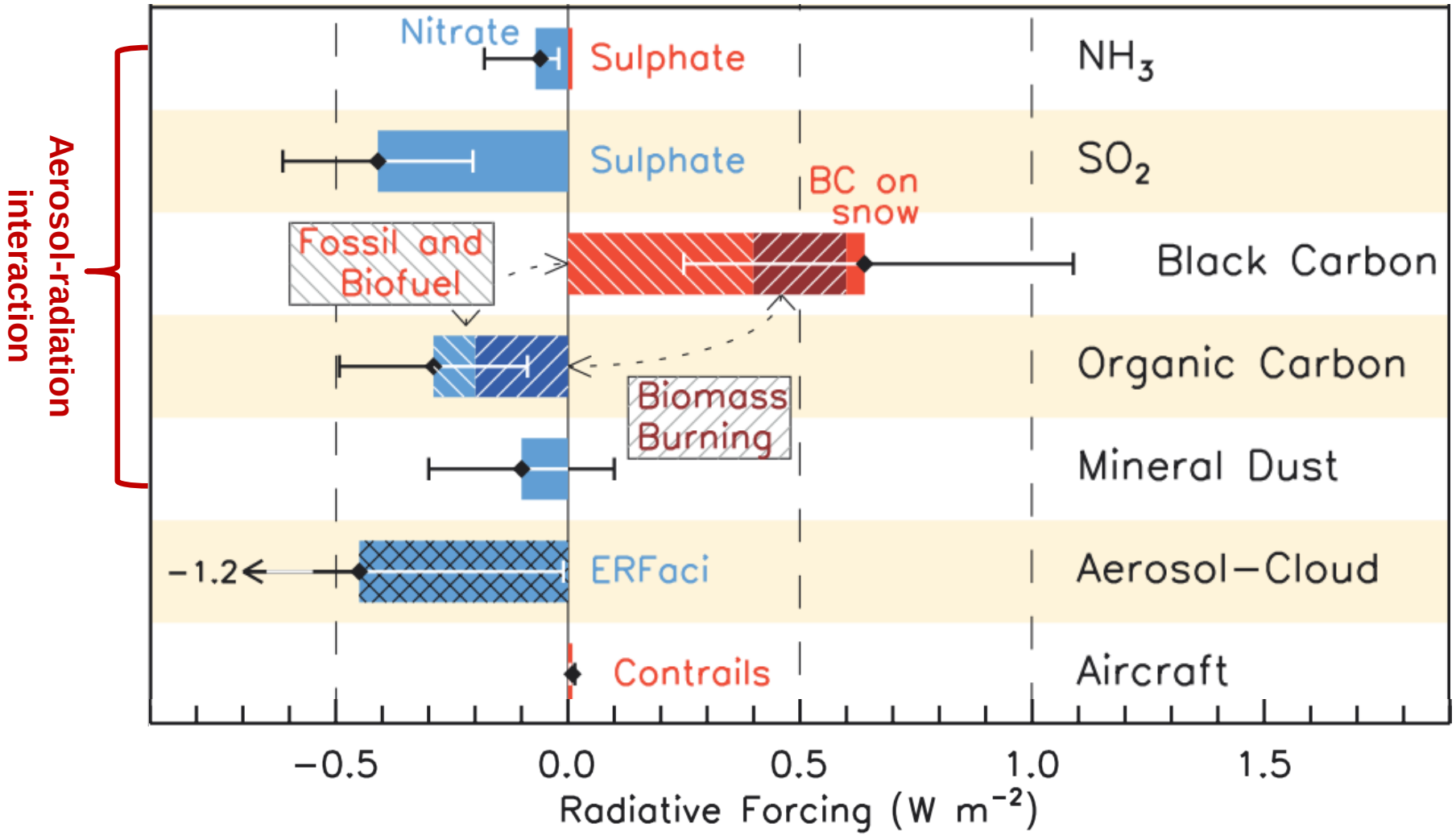
What is aerosol?

- An aerosol is a **C**olloidal **S**ystem of **S**olid or **L**iquid particles.

- Aerosols include a wide range of species: **S**ulfate, **N**itrate, **B**lack carbon, **O**rganic matter, **M**ineral dust and **S**ea-salt.

- Atmospheric processes of aerosols: **E**mission, **P**article growth, **T**ransport, **C**hemical reactions, **S**edimentation, **D**ry deposition, **I**n-cloud scavenging, **B**elow-cloud scavenging.

Effects of Aerosols on Radiation



from IPCC-AR5,2014

Effects of Aerosols on Ecosystems

① Aerosols change the physical climate of the oceans or land ecosystems, and then alter the biogeochemical processes.

②Toxic species (polycyclic aromatic hydrocarbons, heavy metals, …) or nutrients (N, P, Fe, Si, Ca, Mg, …) in aerosols can influence the primary production of ecosystems.

Effects of Aerosols on Ecosystems (Mahowald, Science, 2011)

- Aerosols provides nitrogen to land ecosystems, which is estimated to increase the carbon sink by **0.12 to 0.35 Pg C/year**.

- Phosphorus from deforestation in Amazon fertilizes the local ecosystems, which is estimated to increase the carbon sink by **0.2 to 0.5 Pg C/year**.

- Due to expansion of deserts, increase of iron emission in dust can fertilize the ocean biota and enhanced nitrogen fixing, which is estimated to reduce the atmospheric CO_2 by **~ 4 ± 4 ppm**.

However, the nutrients from combustion have not been considered in current models !!!

Effects of Aerosols on Ecosystems

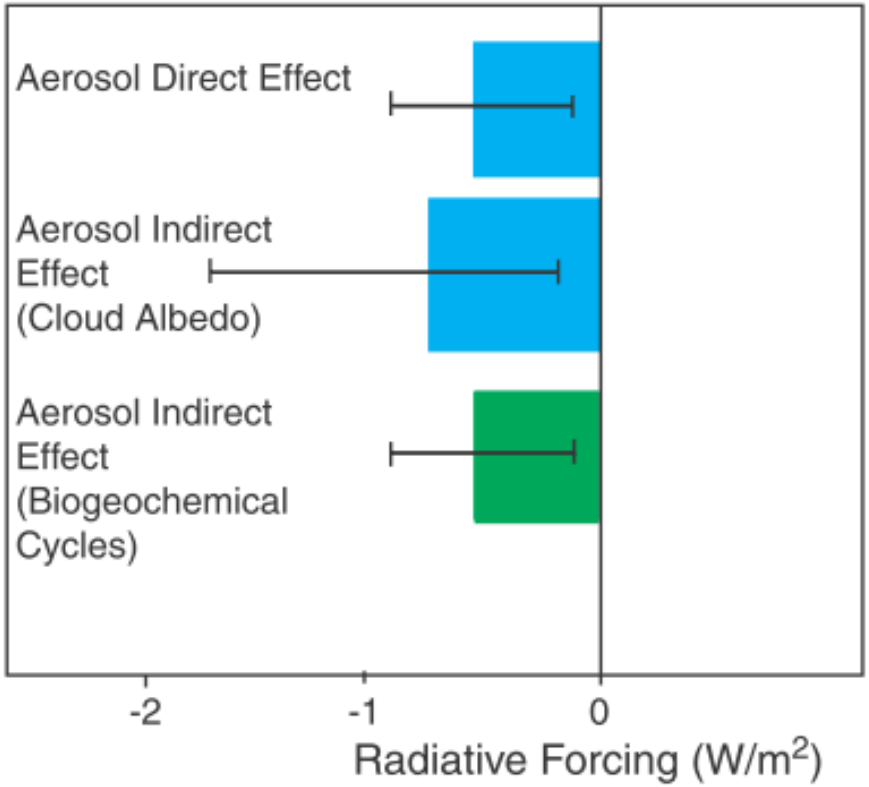


Fig. 1. Aerosol direct and indirect (cloud albedo) radiative forcing estimates (*1*) compared to the radiative forcing estimates from the indirect effect of aerosols from biogeochemical cycles.

Mahowald, Science, 2011

Part 1: Atmospheric cycle of phosphorus

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Significant contribution of combustion-related emissions to the atmospheric phosphorus budget

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Atmospheric phosphorus fertilizes plants and contributes to Earth's biogeochemical phosphorus cycle. However, calculations of the global budget of atmospheric phosphorus have been unbalanced, with global deposition exceeding estimated emissions from dust and sea-salt transport, volcanic eruptions, biogenic sources and combustion of fossil fuels, biofuels and biomass, the latter of which thought to contribute about 5% of total emissions. Here we use measurements of the phosphorus content of various fuels and estimates of the partitioning of phosphorus during combustion to calculate phosphorus emissions to the atmosphere from all combustion sources. We estimate combustion-related emissions of 1.8 Tg P yr^{-1}, which represent over 50% of global atmospheric sources of phosphorus. Using these estimates in atmospheric transport model simulations, we find that the total global emissions of atmospheric phosphorus (3.5 Tg P yr^{-1}) translate to a depositional sink of 2.7 Tg P yr^{-1} over land and 0.8 Tg P yr^{-1} over the oceans. The modelled spatial patterns of phosphorus deposition agree with observations from globally distributed measurement stations, and indicate a near balance of the phosphorus budget. Our finding suggests that the perturbation of the global phosphorus cycle by anthropogenic emissions is larger than previously thought.

Previous budget of P estimated in Mahowald et al. (2008)

The estimated emission into the atmosphere

- 1.15 Tg P yr^{-1} from dust;
- 0.164 Tg P yr^{-1} from primary biogenic particles;
- 0.025 Tg P yr^{-1} from biomass burning;
- 0.024 Tg P yr^{-1} from fossil fuel burning;
- 0.021 Tg P yr^{-1} from biofuel burning;
- 0.006 Tg P yr^{-1} from volcanoes;
- 0.0049 Tg P yr^{-1} from sea-salts.

As a result, the total source is **1.39** Tg P yr^{-1}.

However, based on the observed deposition of P, the total atmospheric sink of P:

- **4.5** Tg P yr^{-1} from Graham and Duce, 1979;
- **3.7** Tg P yr^{-1} from Tippling *et al.*, 2014

Obviously, the budget of P in the atmosphere is not balanced!

New method to estimate the emissions of P

Previous studies estimate the emissions from data on the P content of fine particulate matter (PM10), P content of PM10, and PM10 emission factors.

Our estimate is based on the P content of fuel and on data showing the partitioning of P during combustion into that released to the atmosphere and that retained in combustion residues

$$E = a \cdot b \cdot c \cdot (1 - f) \cdot \sum_{x=1} J_x \cdot \left[\sum_{y=1}^{4} A_y \cdot (1 - R_{x,y}) \right]$$

x represents a given particle size
y represents a specific control device (cyclone, scrubber, ESP, …)
a is the consumption of fuel
b is the rate of combustion
c is the content of P in fuel
f is the fraction of P retained in the residue ash
J_x is the fraction of P emitted in particle size *x*
A_y is the fraction of a given type of control device
$R_{x,y}$ is the removing efficiency of the control device for particles in the size

A new budget of P is provided in our new study (Wang et al., Nature Geoscience, 2015)

	Fluxes and the 90% CI, Tg P yr^{-1}
Sources	
Combustion (present study) [a]	**1.8 (0.5 to 4.4)**
Anthropogenic (including deforestation fires)	**1.1 (0.3 to 3.1)**
Natural	**0.7 (0.2 to 1.3)**
Mineral dust input [b]	0.93 (0.23 to 2.1)
Primary biogenic aerosol particles [c]	0.58 (0.16 to 1.0)
Volcanoes [d]	0.006 (0.003 to 0.009)
Sea salt [e]	0.16 (0.0049 to 0.33)
Phosphine from marshes & paddies [f]	0.00020 (0.000038 to 0.00036)
Total sources	**3.5 (0.9 to 7.8)**
Sinks	
Total sinks from the model	3.5 (0.9 to 7.8)
over land	2.7 (0.7 to 6.2)
over oceans	0.8 (0.2 to 1.6)
From Graham and Duce, 1979 (ref. 10)	4.5
From Tippling *et al.*, 2014 (ref. 13)	3.7

Modeling of Phosphorus in the atmosphere

A global atmospheric general circulation model LMDz-OR-INCA at a horizontal resolution of 0.94° latitude by 1.28° longitude with 39 vertical layers:

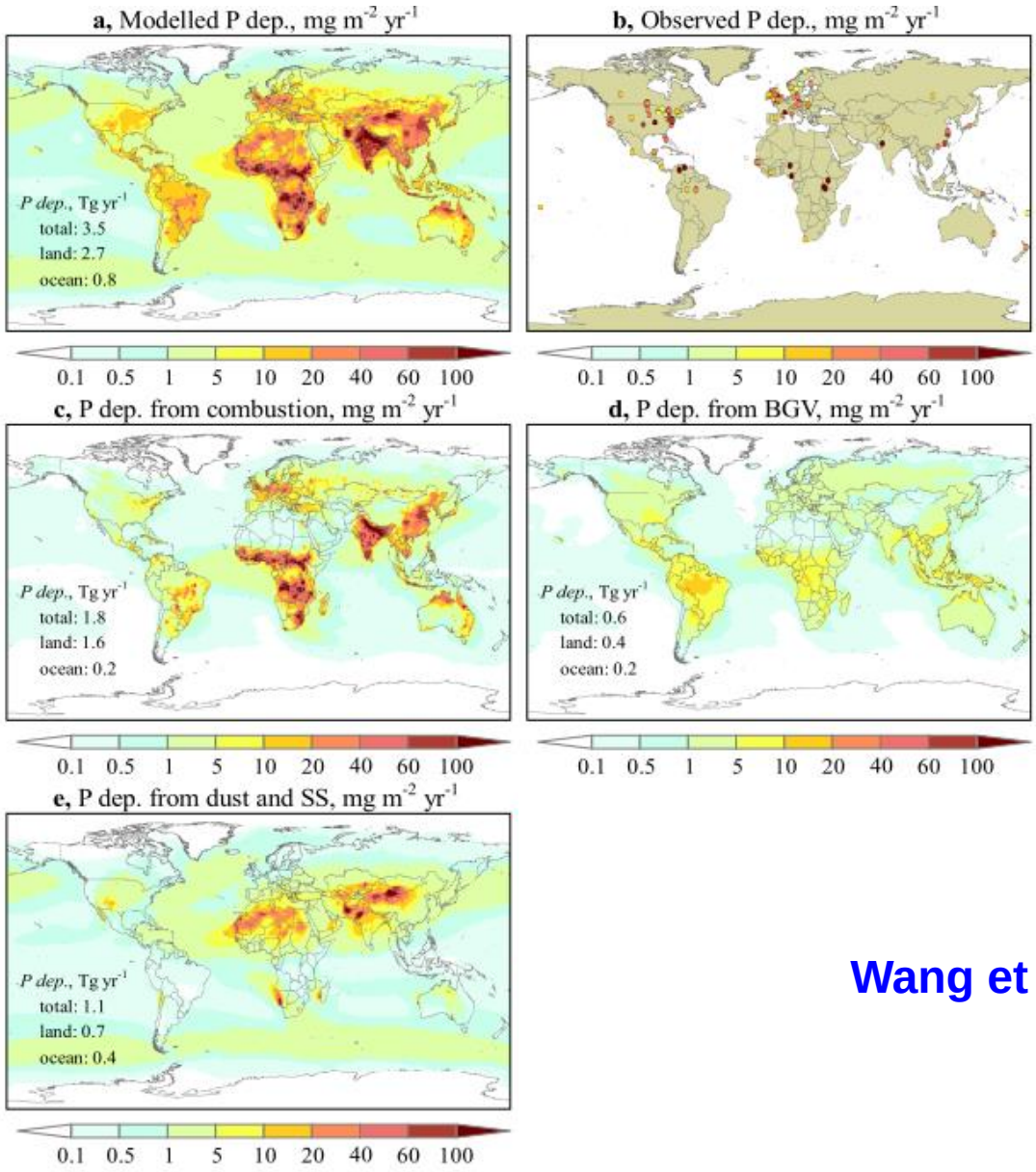
- ✓ P emitted from combustion sources were modelled in one fine mode (size = 0.34 um) and two coarse modes (size = 2.5 um and 10.0 um)

- ✓ P emitted from biogenic sources (small pollens, etc) and volcanoes were modelled in one coarse mode (size = 5 um)

- ✓ P emitted from mineral sources were modelled as dust (size = 2.5 um)

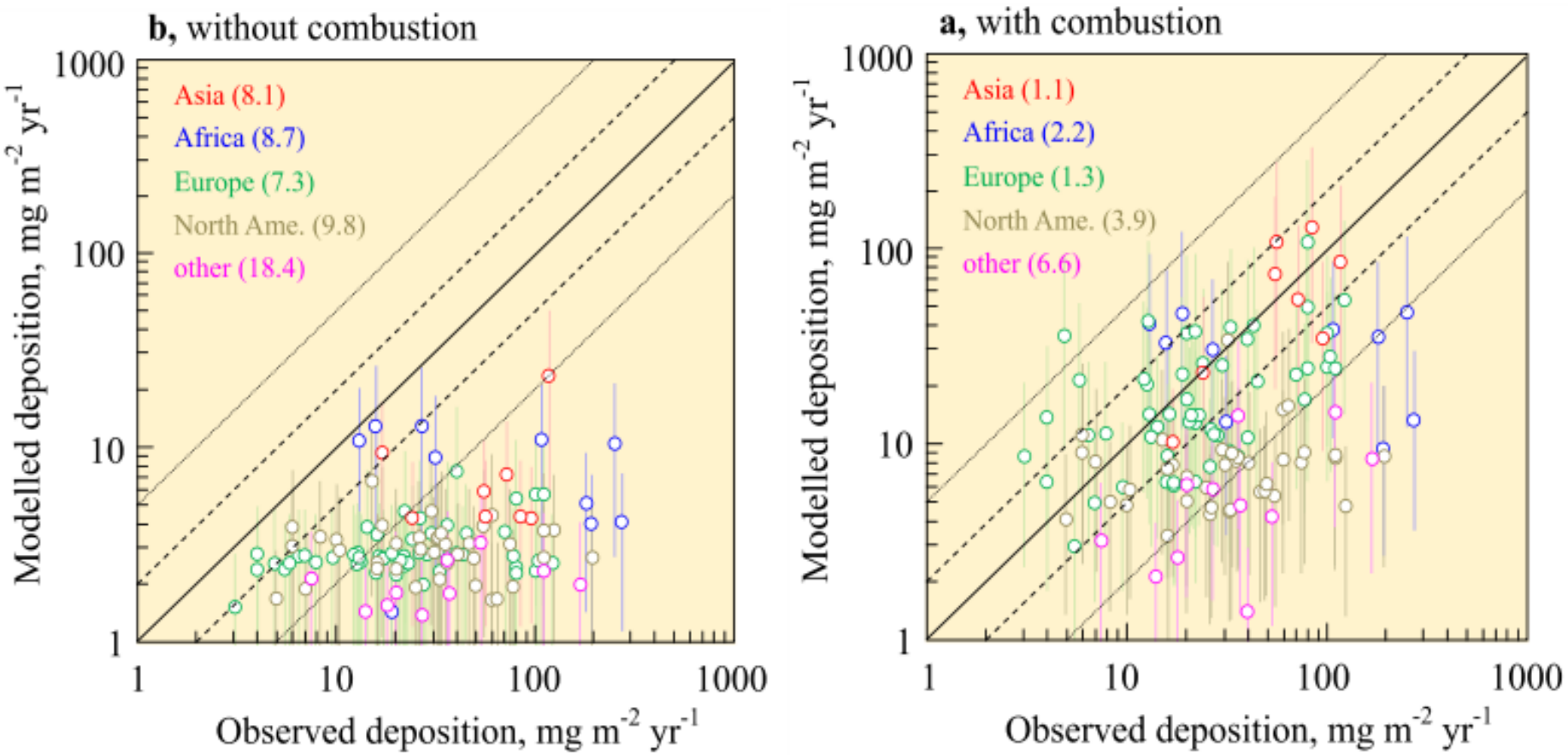
- ✓ P emitted from sea-salt were modelled as sea-salt.

Atmospheric deposition of phosphorus



Wang et al., Nature Geoscience, 2014;

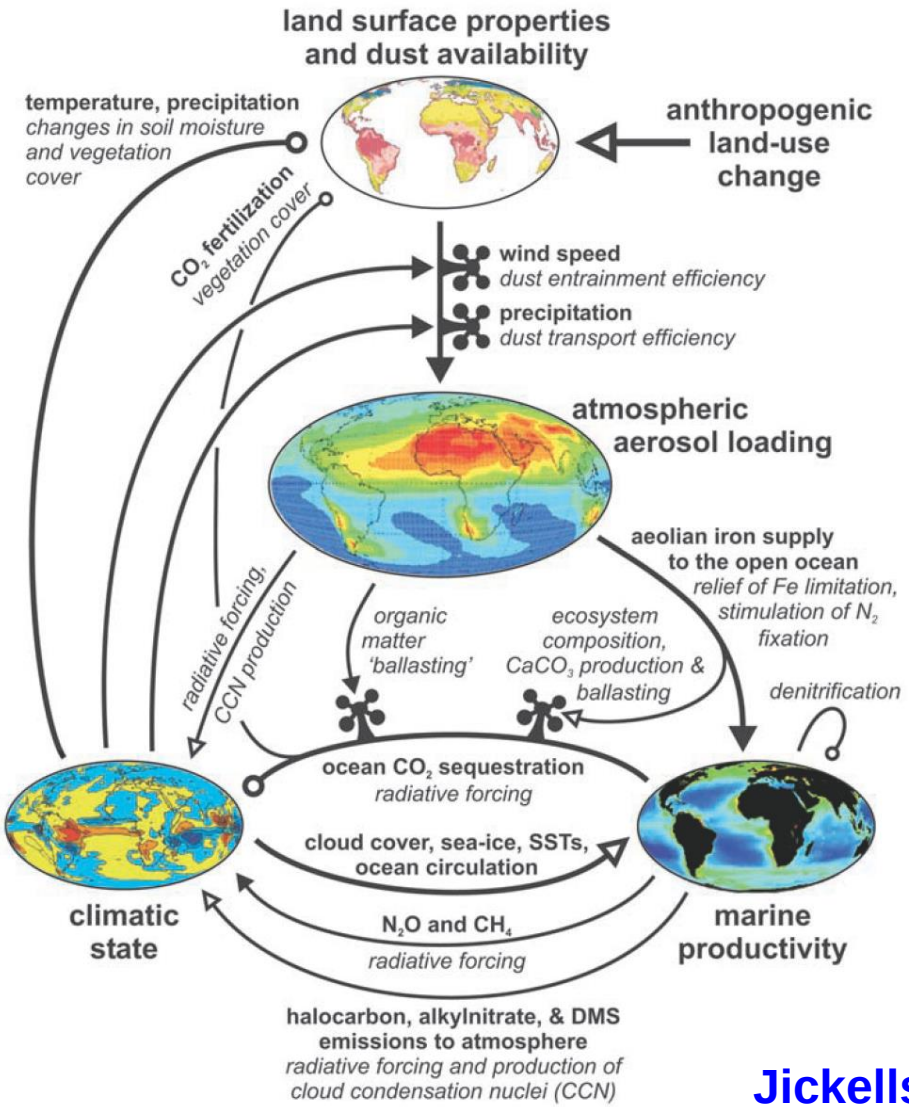
Model Validation with / without combustion sources



Wang et al., Nature Geoscience, 2014;

Part 2: Atmospheric cycle of iron (Fe)

Iron (Fe) is the most important element for ocean biogeochemistry



Jickells et al., Science, 2005;

Methods: Emission of Iron in combustion

$$E = a \cdot b \cdot c \cdot (1 - f) \cdot \sum_{x=1} J_x \cdot \left[\sum_{y=1}^{4} A_y \cdot (1 - R_{x,y}) \right]$$

x represents a given particle size
y represents a specific control device (cyclone, scrubber, ESP, …)
a is the consumption of fuel
b is the combustion rate
c is content of Fe in fuel
f is the fraction of Fe retained in the residue ash
J_x is the fraction of Fe emitted in particle size *x*
A_y is the fraction of a given type of control device
$R_{x,y}$ is the removing efficiency of the control device for particles in the size

Methods: Atmospheric Transport of Fe

A global aerosol model LMDz-INCA at a horizontal resolution of 0.94° latitude by 1.28° longitude and 39 vertical layers from the surface to 4.3Pa.

The model couples a General Circulation Model LMDZ (Hourdin et al., 2006) with a aerosol module INCA (Balkanski et al., 2004, 2007).

Fe emitted from combustion sources, three size bins was considered:

- Fe in PM_1 as a fine mode (MMD = 0.34 μm, σ = 1.59);
- Fe in PM_{1-10} as a coarse mode (MMD = 3.4 μm, σ = 2.0);
- Fe in $PM_{>10}$ as a coarse mode (MMD = 34 μm, σ = 2.0).

Fe emitted from dust sources:

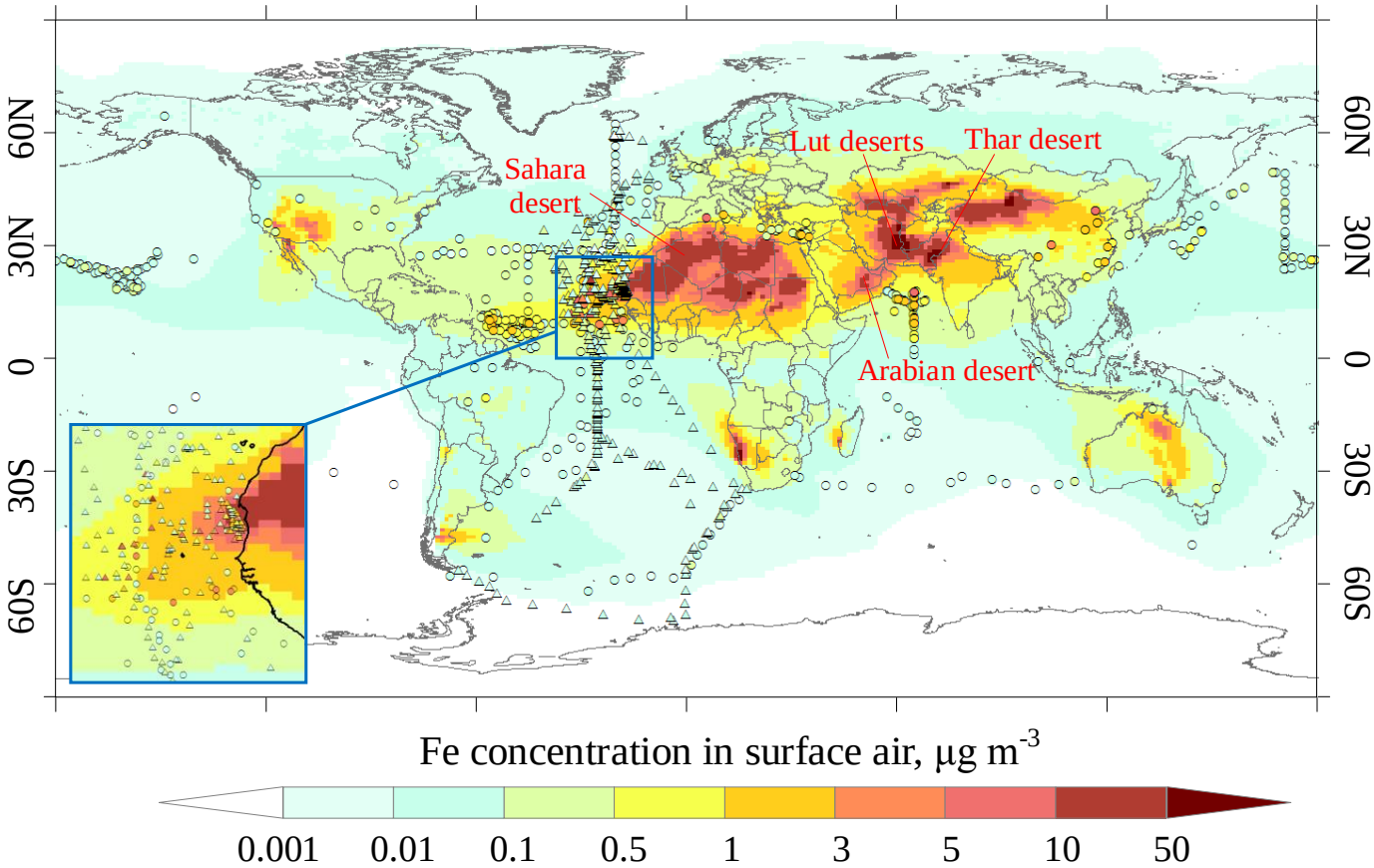
- the content of Fe in dust was estimated based on a lastest soil mineralogy database (Journet et al., 2014).
- the transport of Fe is treated the same as dust (MMD = 2.5 μm, σ = 2.0).

Results - Emission Estimates:

Table 1. Comparison of Fe emissions in our work and previous studies ("**B71**" for Bertine and Goldberg, 1971; "**Luo08**" for Luo et al., 2008; and "**Ito13**" for Ito, 2013). Unit: **Tg yr^{-1}**.

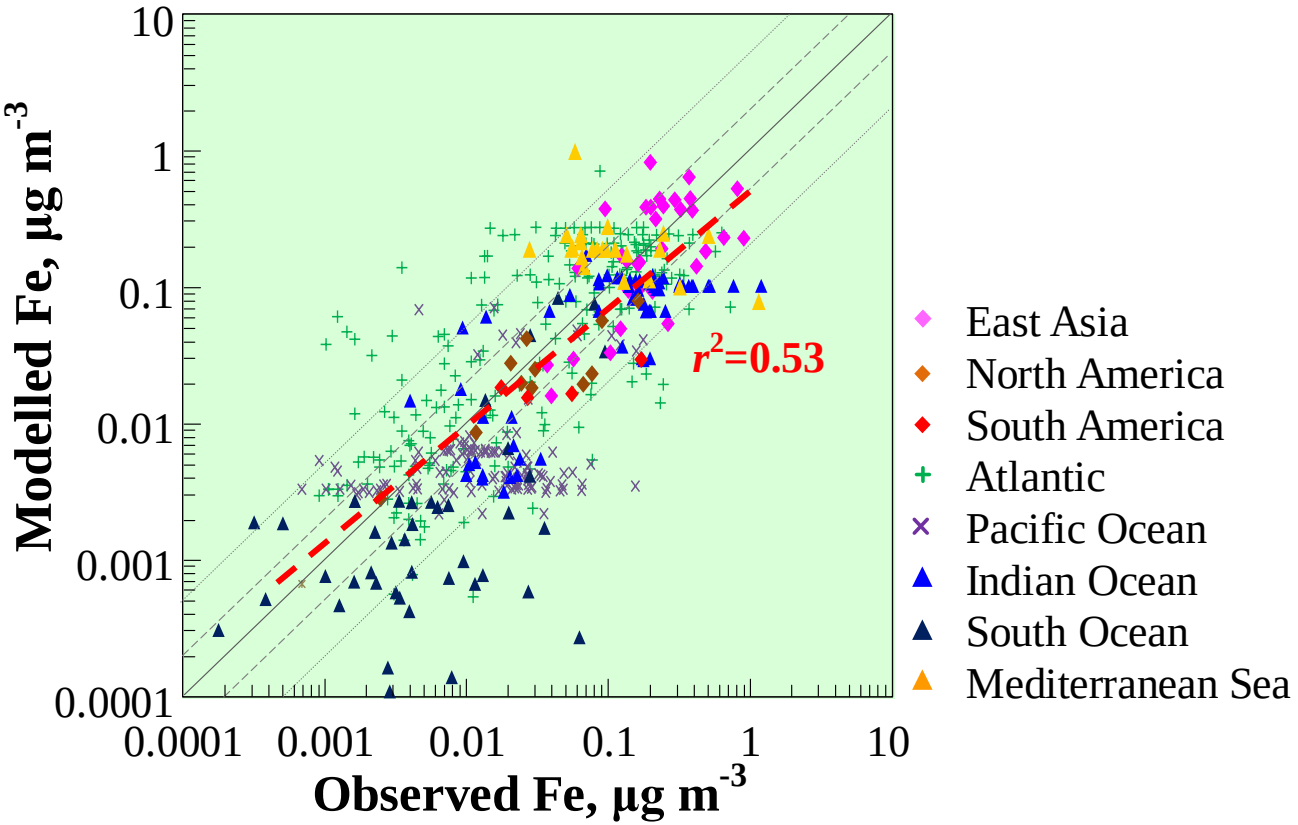
	Years	**Fossil fuels**	**Biomass**	**Dust**
B71	1967	1.4 (all sizes)		
Luo08	1996	0.56 (PM_{1-10})	0.86 (PM_{1-10})	55 (using a Fe content of 3.5%)
		0.10 (PM_1)	0.21 (PM_1)	
Ito13	2001	0.44 (PM_{1-10})	0.92 (PM_{1-10})	74 (using a Fe content of 3.5%)
		0.07 (PM_1)	0.23 (PM_1)	
Our work	1967	2.32 ($PM_{>10}$)		
		0.64 (PM_{1-10})		
		0.017 (PM_1)		
	1996	1.14 (PM_{1-10})	0.31 (PM_{1-10})	
		0.036 (PM_1)	0.012 (PM_1)	
	2001	0.83 (PM_{1-10})	0.31 (PM_{1-10})	
		0.035 (PM_{1-10})	0.012 (PM_1)	
	2007			35 (using a Fe content of 3.5%)
	2007			38 (using the mineralogy data)

Results – Modelled Surface Concentrations:



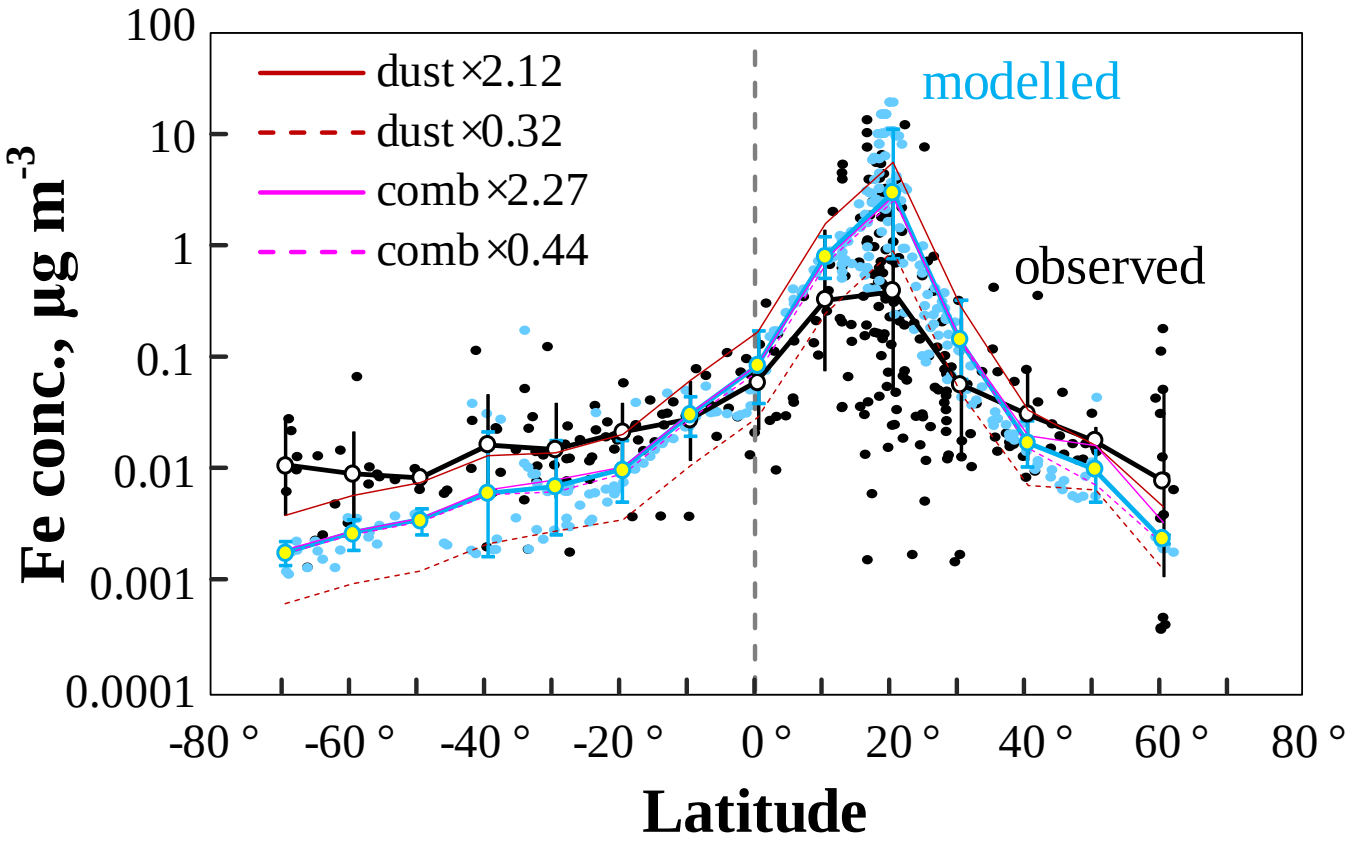
Wang et al., ACP, 2015

Results – Validation of modelled Surface Concentrations:



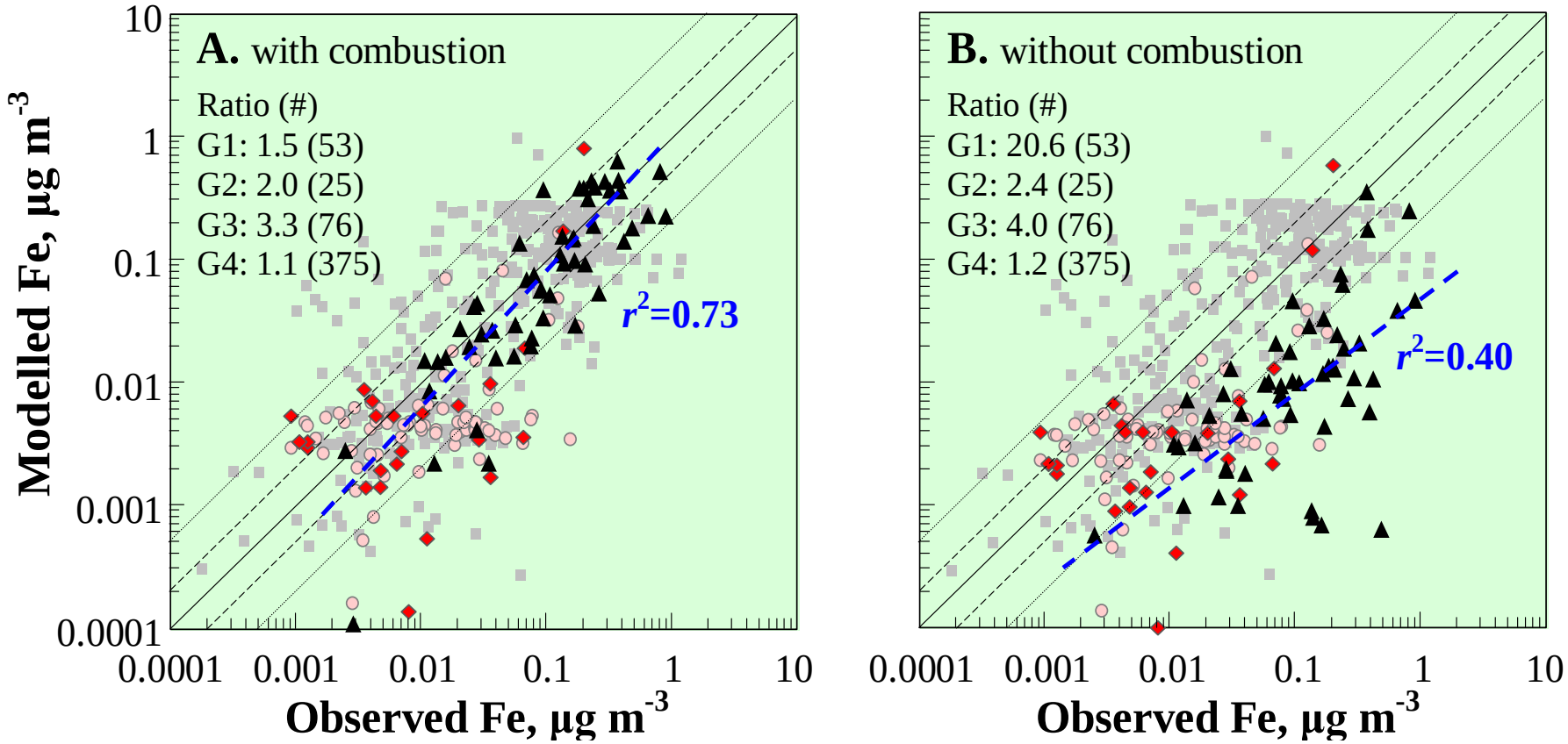
Wang et al., ACP, 2015

Results – Latitudinal Distribution of Fe over the Atlantic Ocean



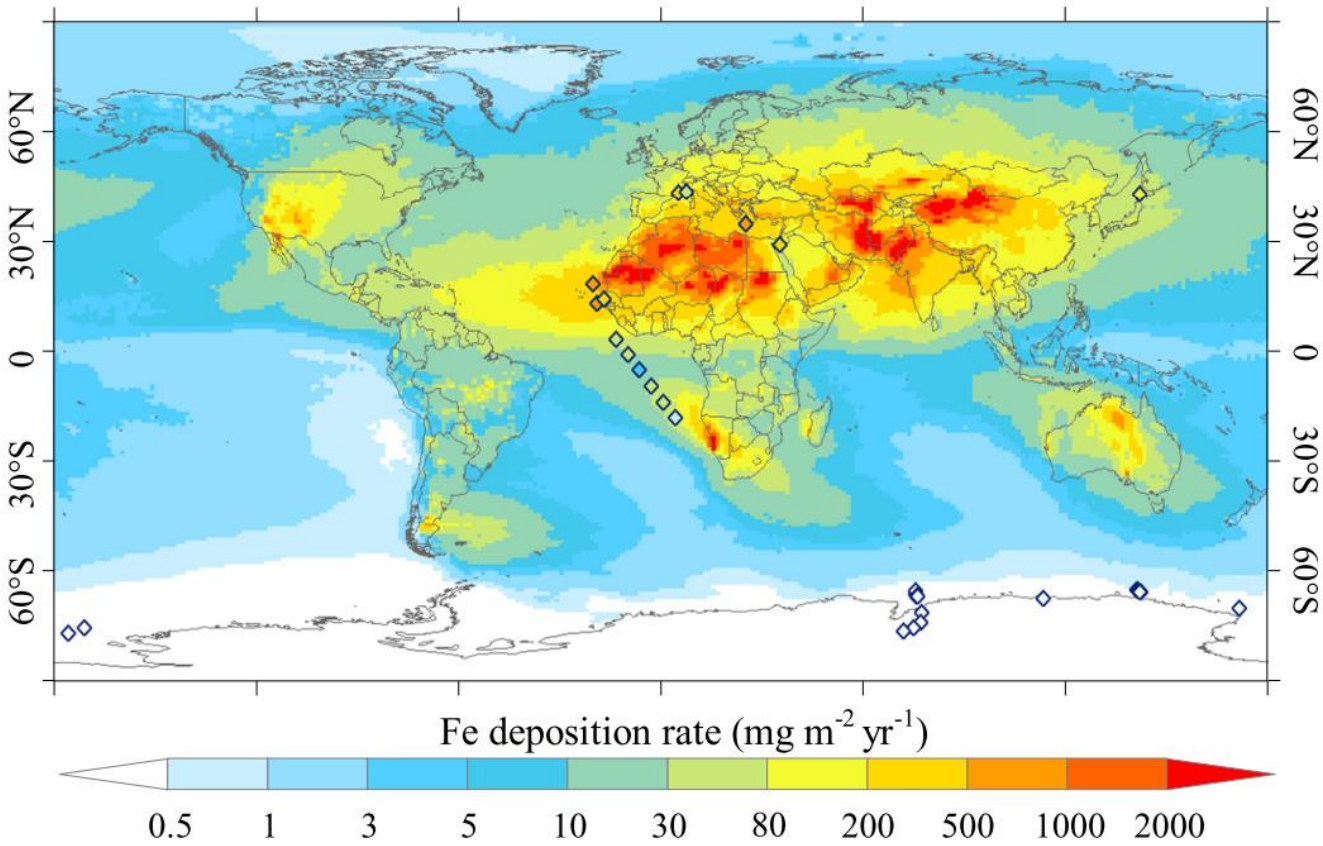
Wang et al., ACP, 2015

Results – Comparison to sites dominated by combustion sources



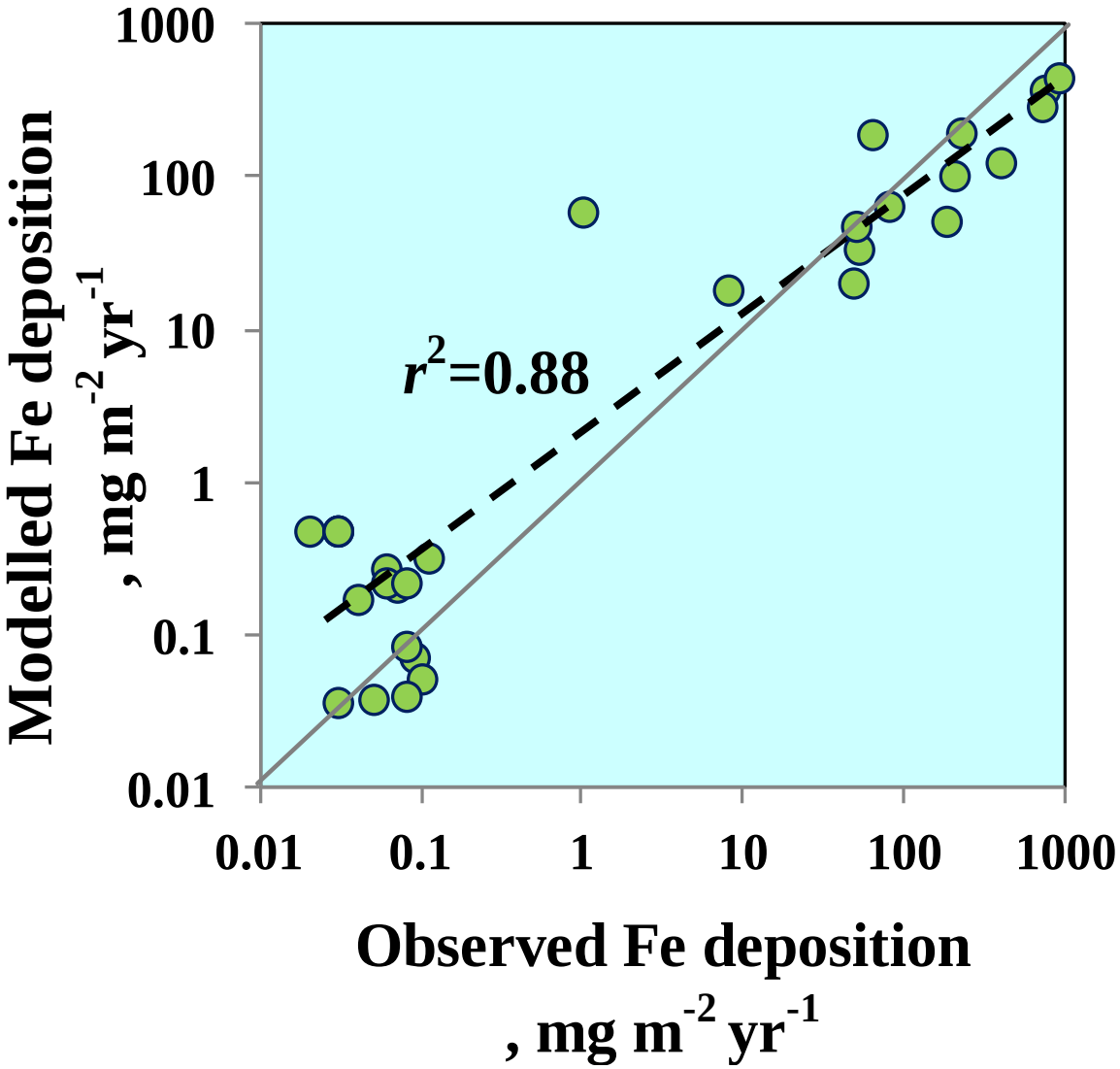
Wang et al., ACP, 2015

Results – Modelled Fe deposition rate:



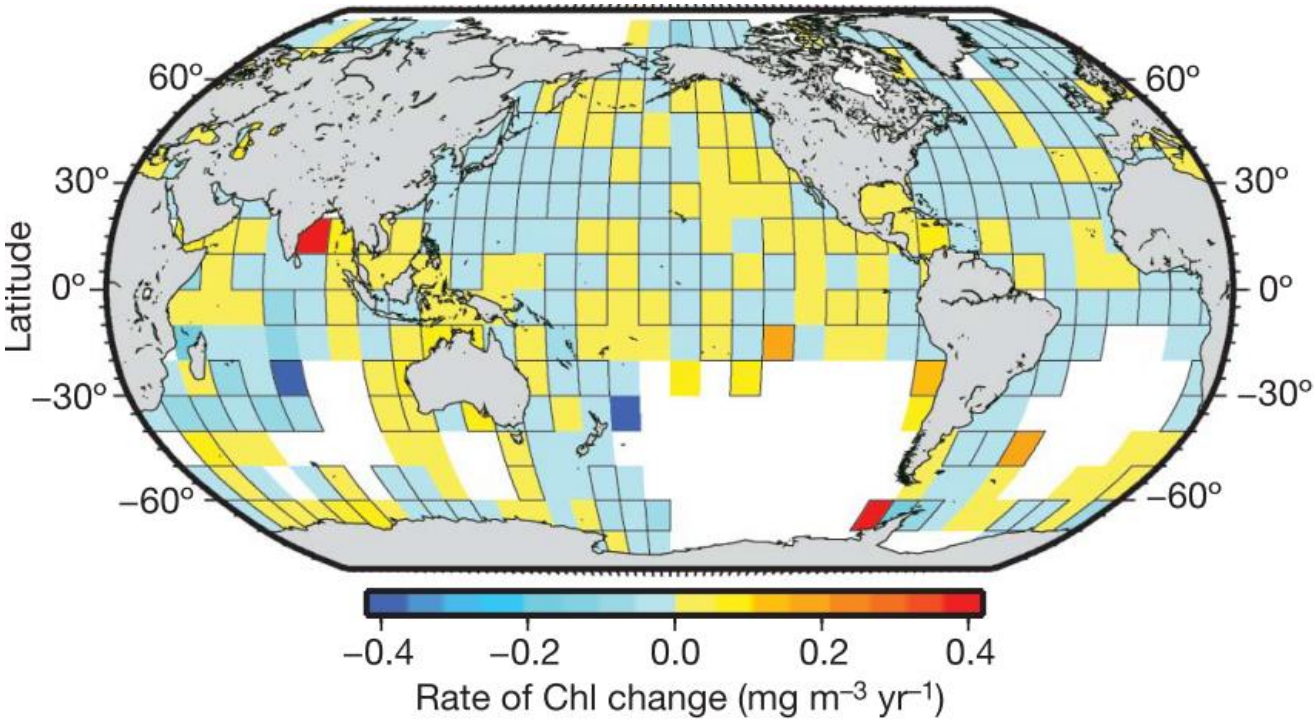
Wang et al., ACP, 2015

Results – Validation of modelled Fe deposition rates



Part 3: Global ocean model

Ocean phytoplankton might be decreasing in the last century

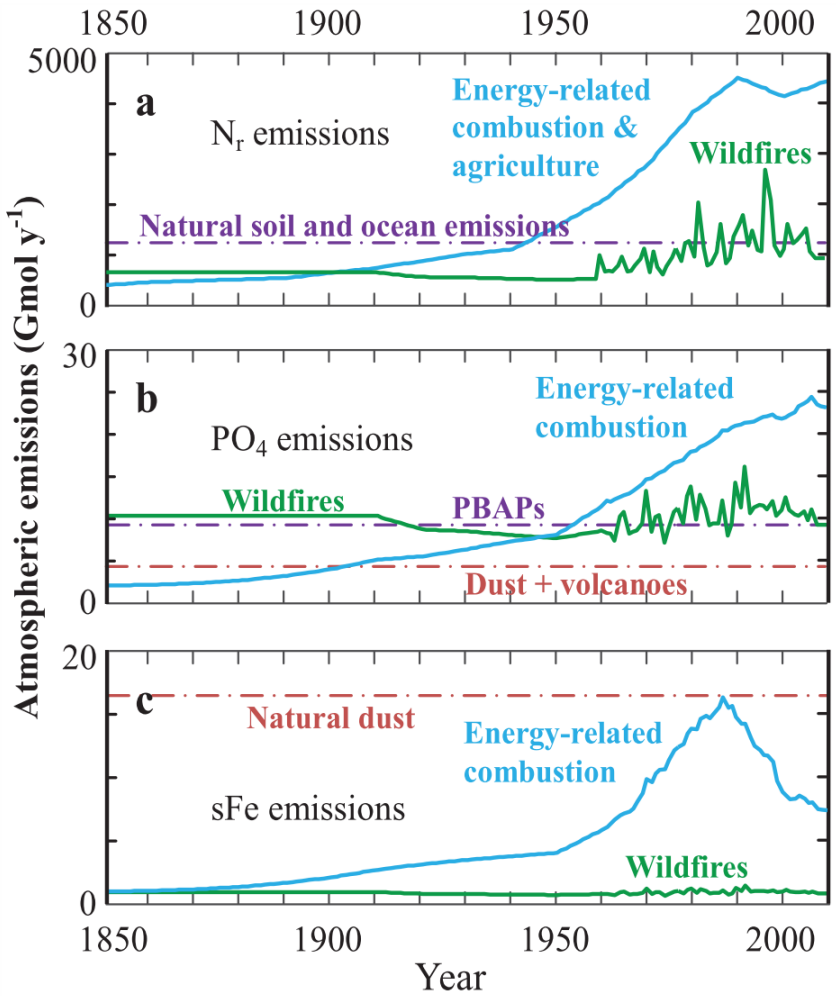


Boyce et al., Nature, 2010

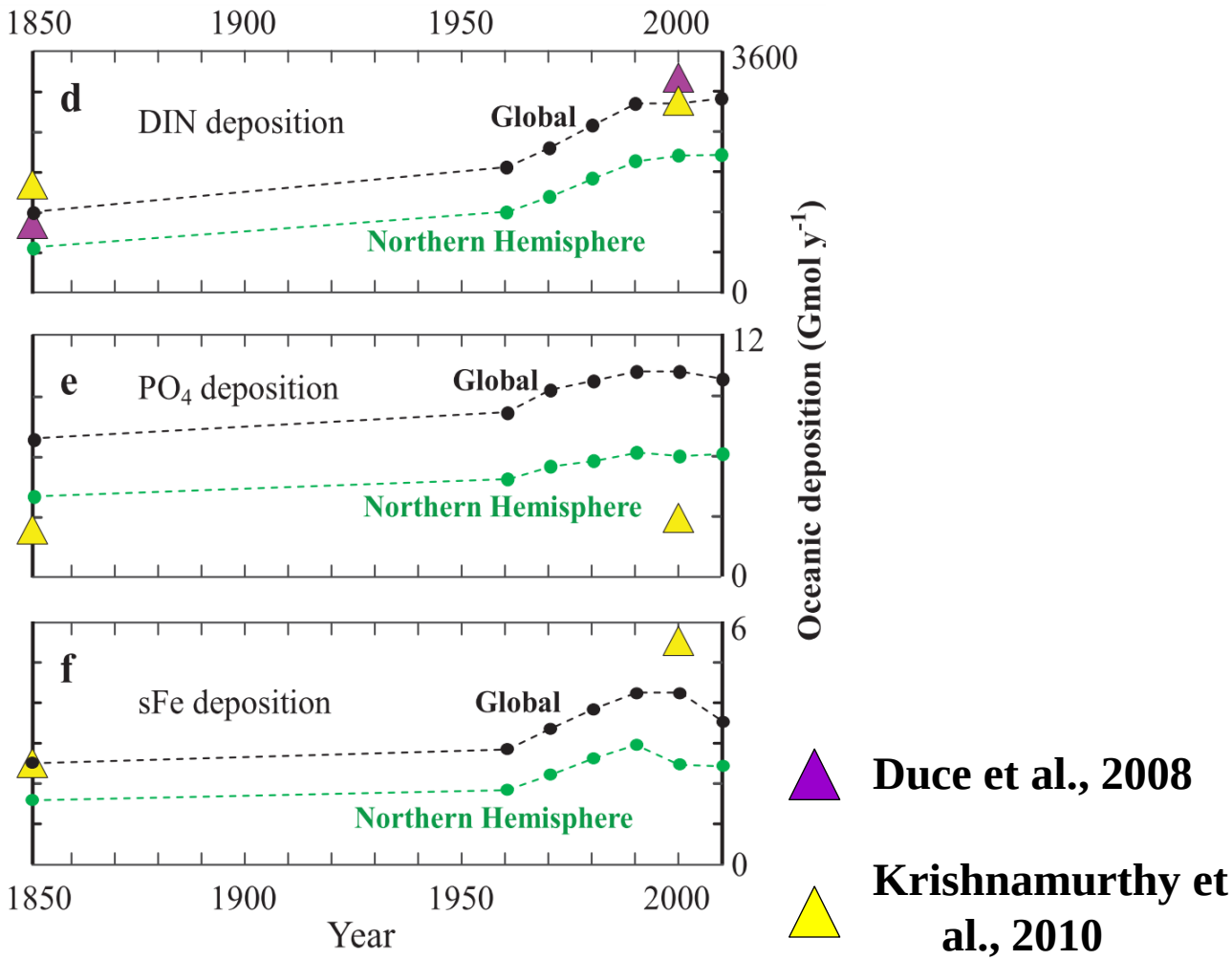
Field and model simulations suggest a decline in marine phytoplankton and the net primary production (NPP), because global warming has led to the increasing stratification of water columns and reduction of the supply of nutrients from subsurface waters (IPCC, 2013).

Our question: can anthropogenic aerosols provide additional nutrients and change the temporal trend of marine phytoplankton?

Historical emissions of reactive nitrogen (N_r), phosphate(PO_4) and soluble iron (sFe) - Reconstructed in our study



Oceanic deposition of dissolved inorganic nitrogen (DIN), phosphate (PO_4) and soluble iron (sFe) - Simulated in our study



Methods: Global Carbon-Climate Coupled Ocean Model NEMO-PISCES

A state-of-the-art carbon-climate coupled ocean model NEMO-PISCES (version 2) was used to simulate the fate of nutrients (N, P, Fe and silicon) and the carbon cycle in global oceans. The model was run with the ocean dynamics simulated by an ocean physical model ORCA2-LIM (version 3.2), which couples an oceanic general circulation model OPA9 (Ocean PArallelise) with a sea-ice model LIM2 (Louvain-la-Neuve). The horizontal resolution of the model is 2° ×2° cos (latitude), with a zoomed resolution of 0.5° ×0.5° over the equatorial oceans. There are 30 vertical layers from the ocean surface down to a depth of 5000 m, varying from a vertical resolution of 10 m at the surface to one of 500 m at the bottom. External sources of nutrients include: atmospheric deposition, rivers, exchange between the sediments and the water, exchange between the sea ice and the water, and hydrothermal vents.

Two Experiments:

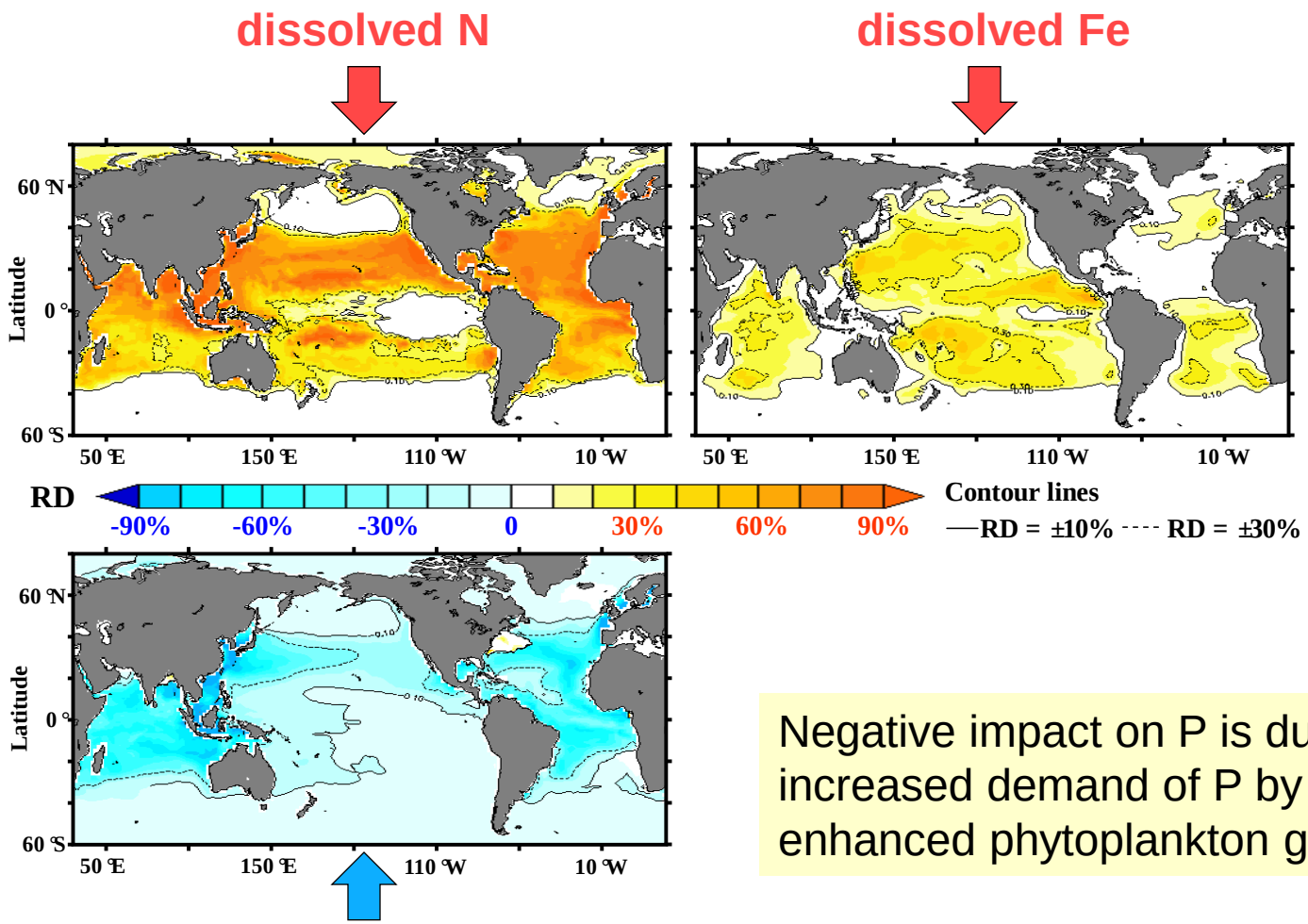
Without anthropogenic aerosol deposition (CTL):

- ✓ We used the standard model configuration as done in Bopp et al., 2013.
- ✓ The deposition of N, P and Fe was fixed at the 1850 levels.

With anthropogenic aerosol deposition (DEP):

- ✓ We used the standard model configuration as done in Bopp et al., 2013.
- ✓ The monthly deposition of N, P and Fe simulated by our 3-D atmospheric transport model (LMDZ-ORCHIDEE-INCA) from 1850 to 2010 was prescribed to NEMO-PISCES.

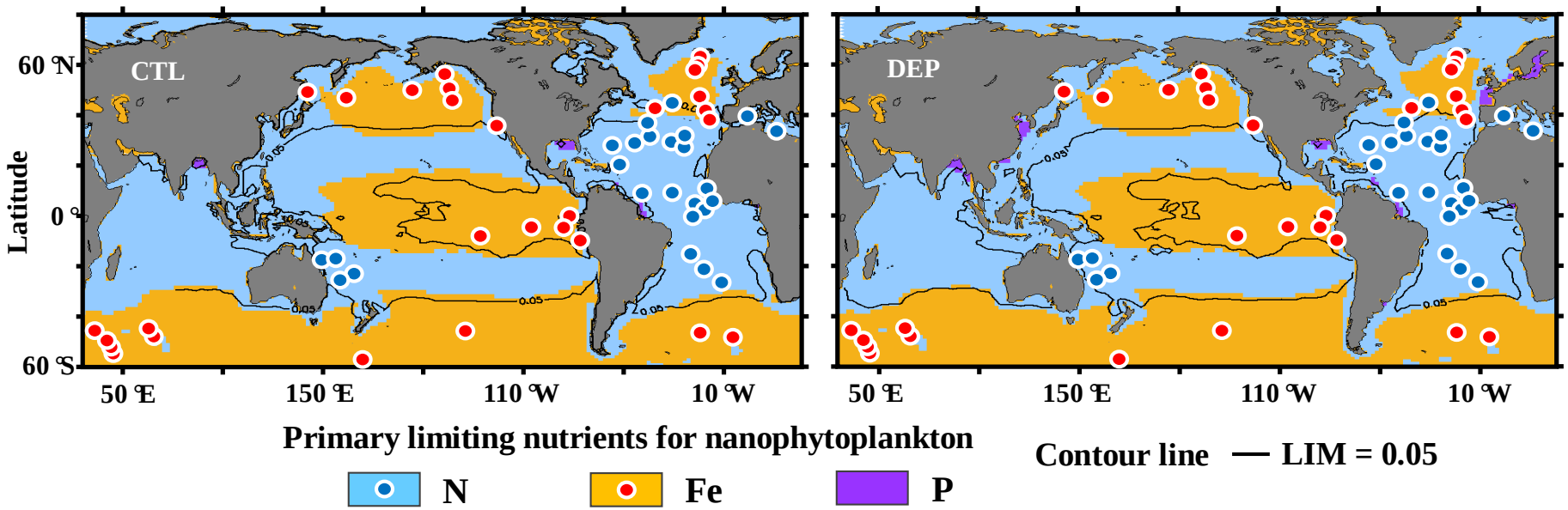
Impact of anthropogenic aerosols on ocean nutrient concentrations as difference between CTL and DEP



Negative impact on P is due to increased demand of P by enhanced phytoplankton growth.

Impact of anthropogenic aerosols on nutrient limitation to phytoplankton: the high-limitation area (limitation factor < 0.05) is shrinking due to anthropogenic aerosols

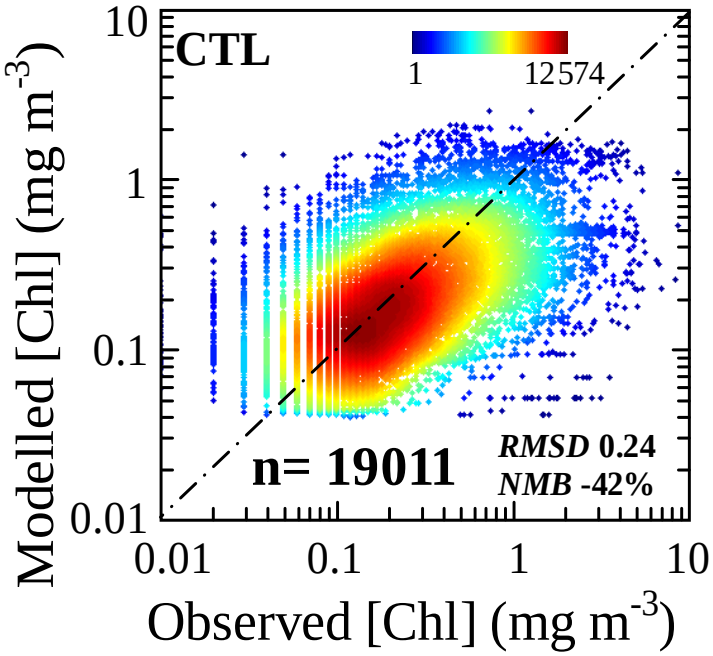
Without anthropogenic aerosols | **With anthropogenic aerosols**



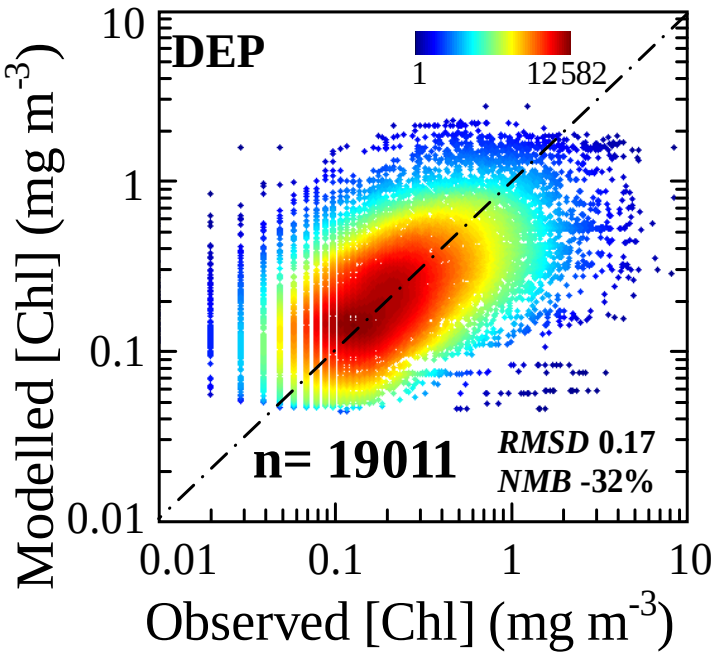
Circles show the primary limitation nutrient observed in Moore et al, 2013

Impact of anthropogenic aerosols on oceanic chlorophyll concentrations

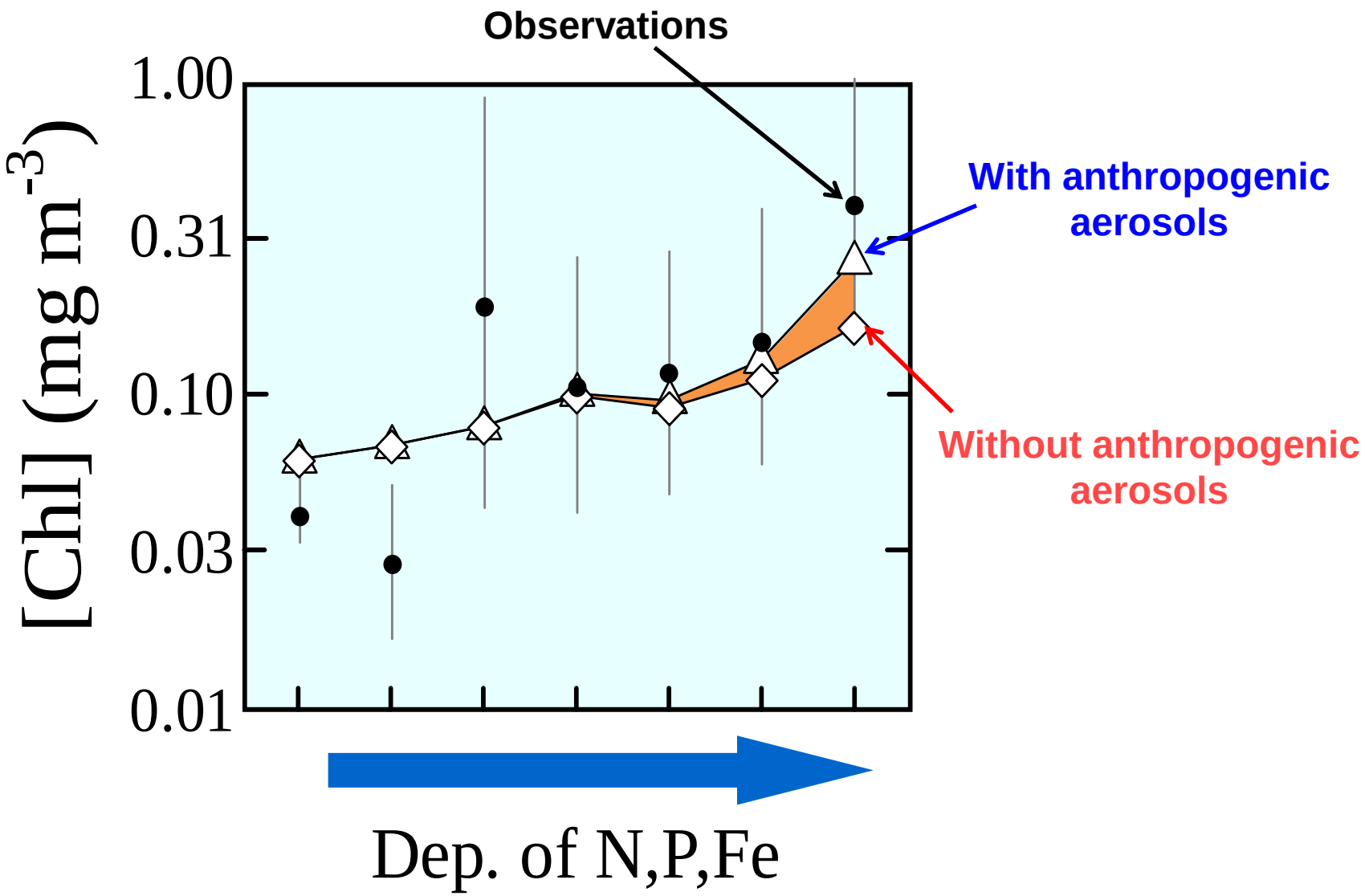
Without anthropogenic aerosols



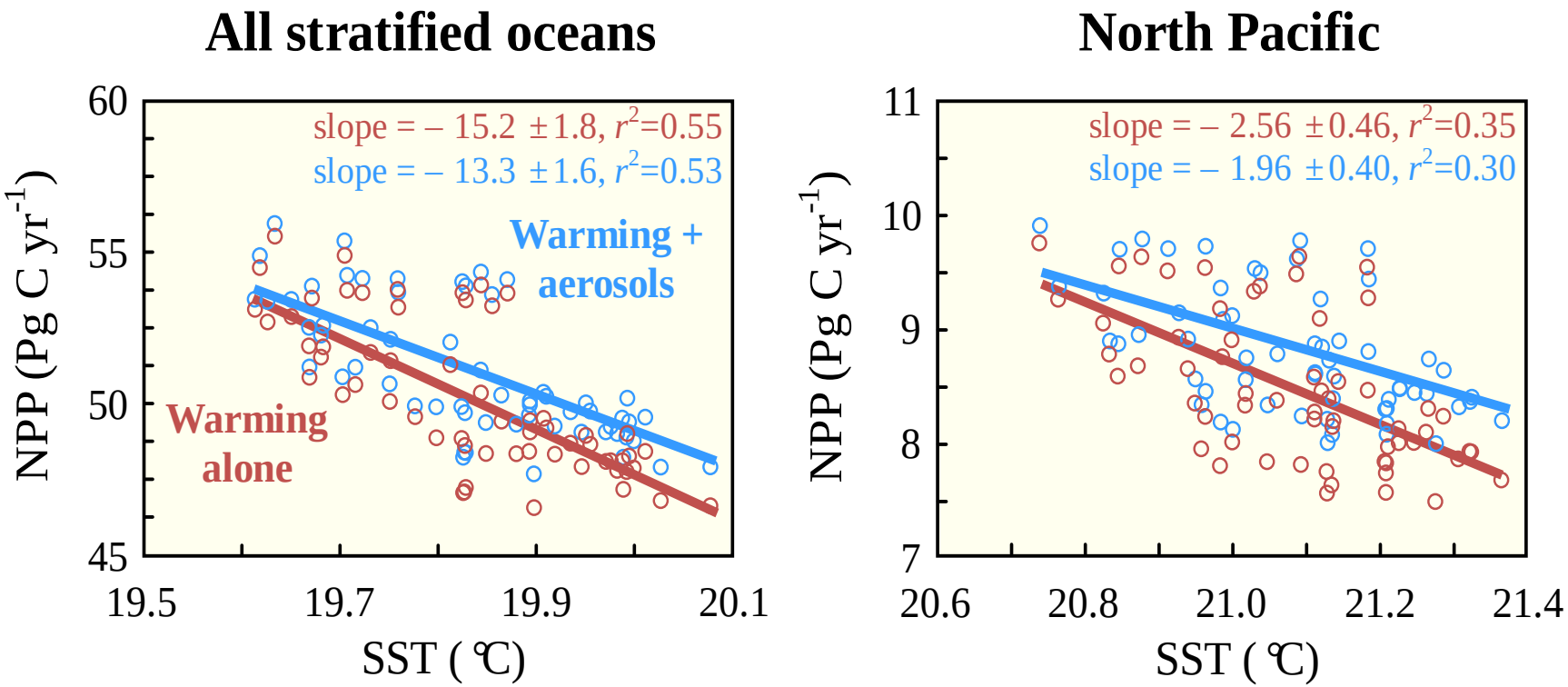
With anthropogenic aerosols



Observations from global ocean database (Wang et al., 2013).


Observations
With anthropogenic aerosols
Without anthropogenic aerosols
1.00
0.31
0.10
0.03
0.01
[Chl] (mg m-3)
Dep. of N,P,Fe

Impact of anthropogenic aerosols on the sensitivity of marine NPP to sea-surface temperature (SST) from 1948 to 2007



Part 4: Global land dynamic model (I will present these results in the next year)

Thank for your attention!!!