

Chemistry and deposition of mineral aerosol over the Southern oceanic area Study on Kerguelen and Crozet Islands



université
PARIS
PARIS 7
DIDEROT

lisa | Laboratoire Inter-Universitaire
des Systèmes Atmosphériques

UPEC
Connaissance - Action

UNIVERSITÉ
PARIS-EST CRÉTEIL
VAL DE MARNE

Alexie Heimburger
Thesis defense



INSU
Institut national des sciences de l'Univers

UPMC
SORBONNE UNIVERSITÉS

surface ocean **solas** 2019 lower atmosphere study



Scientific context

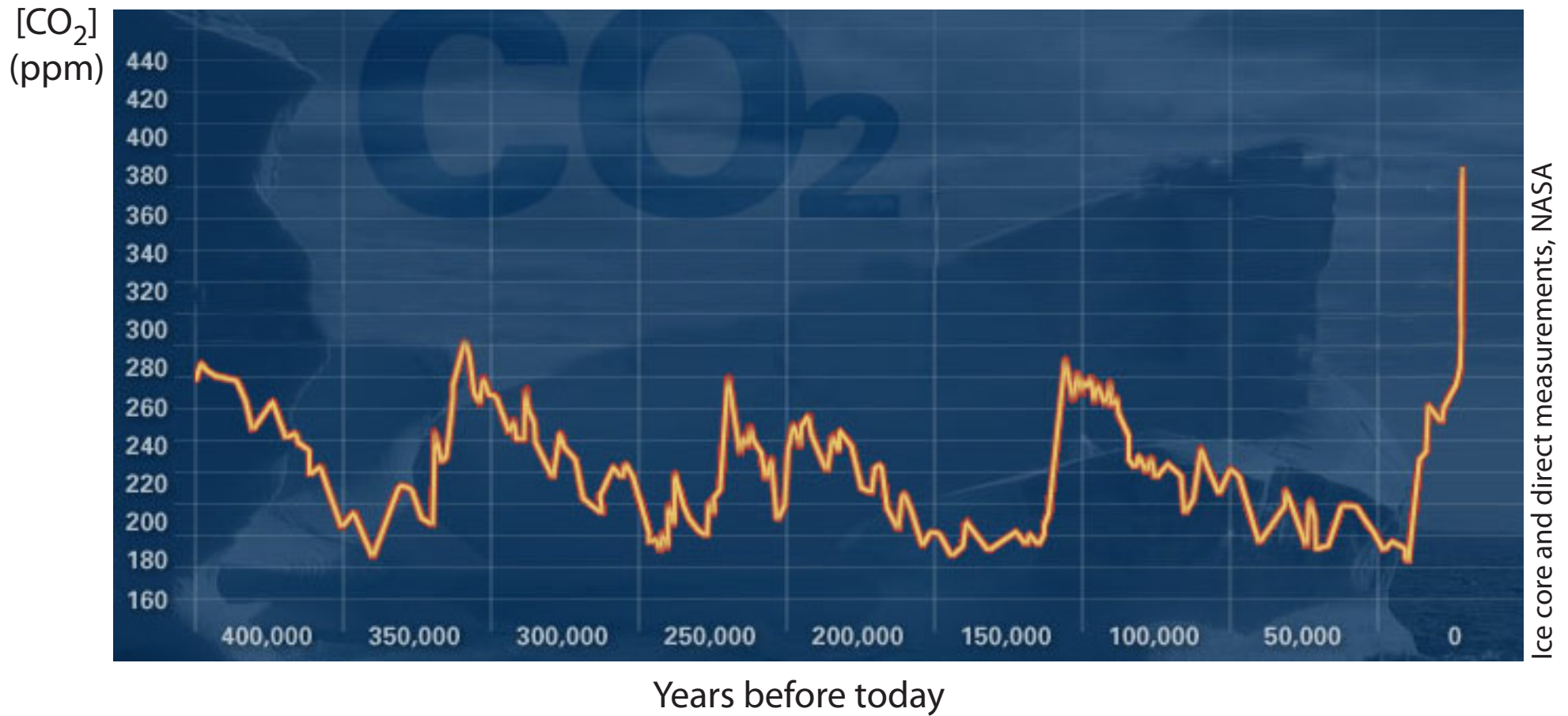
Scientific context

Issues

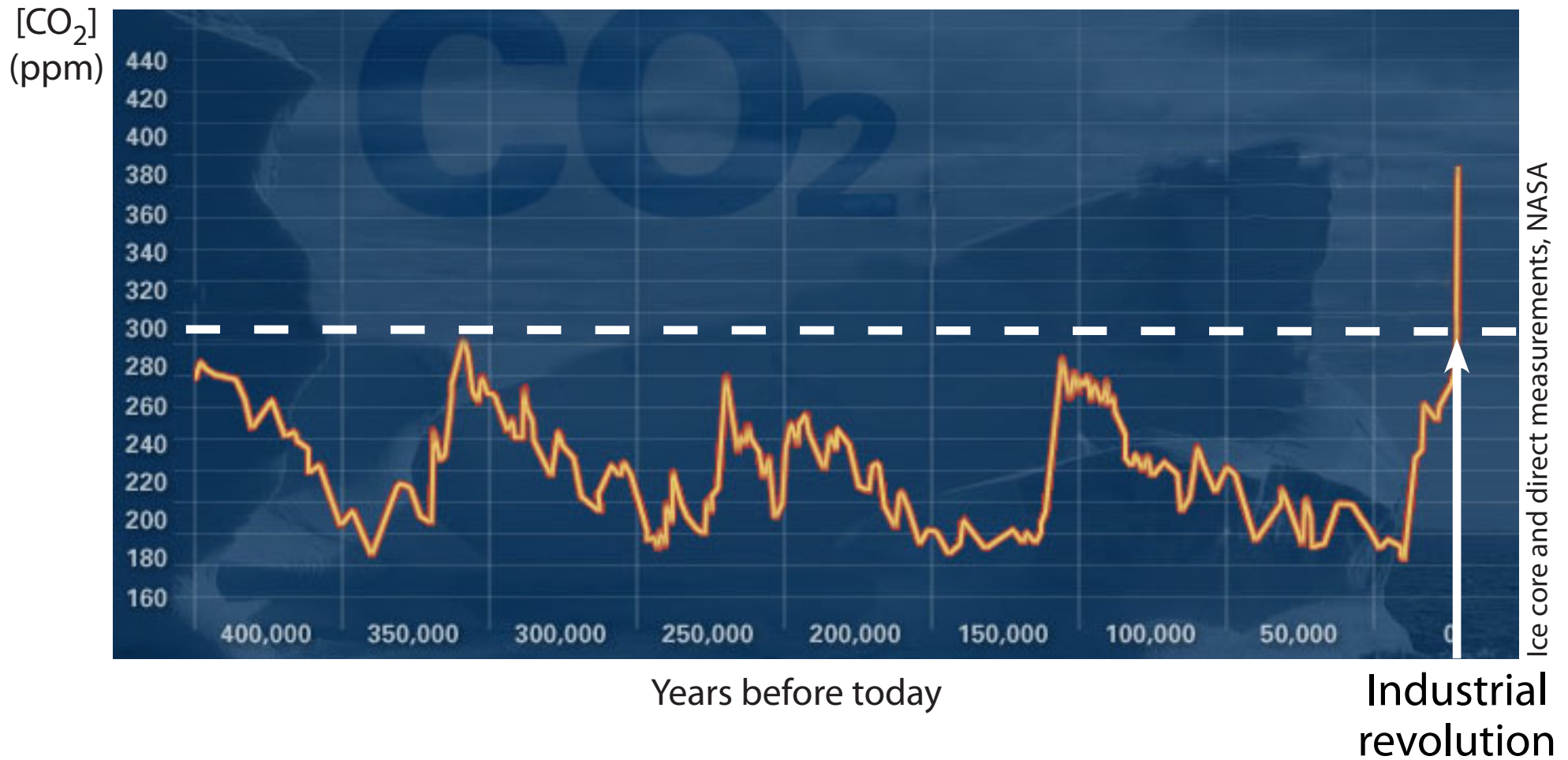
Methods

Results

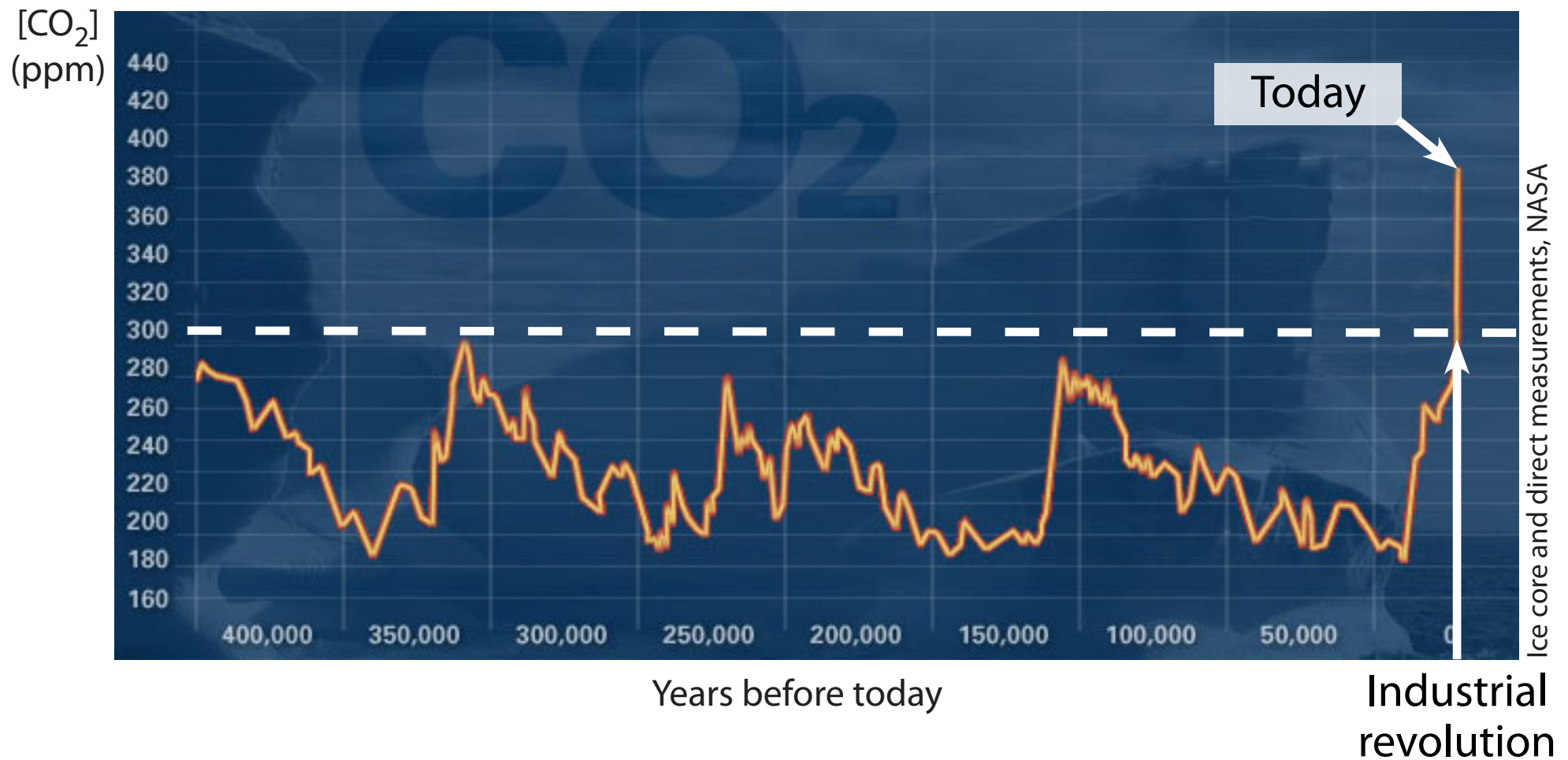
CO₂ level in the atmosphere since 400 000 years



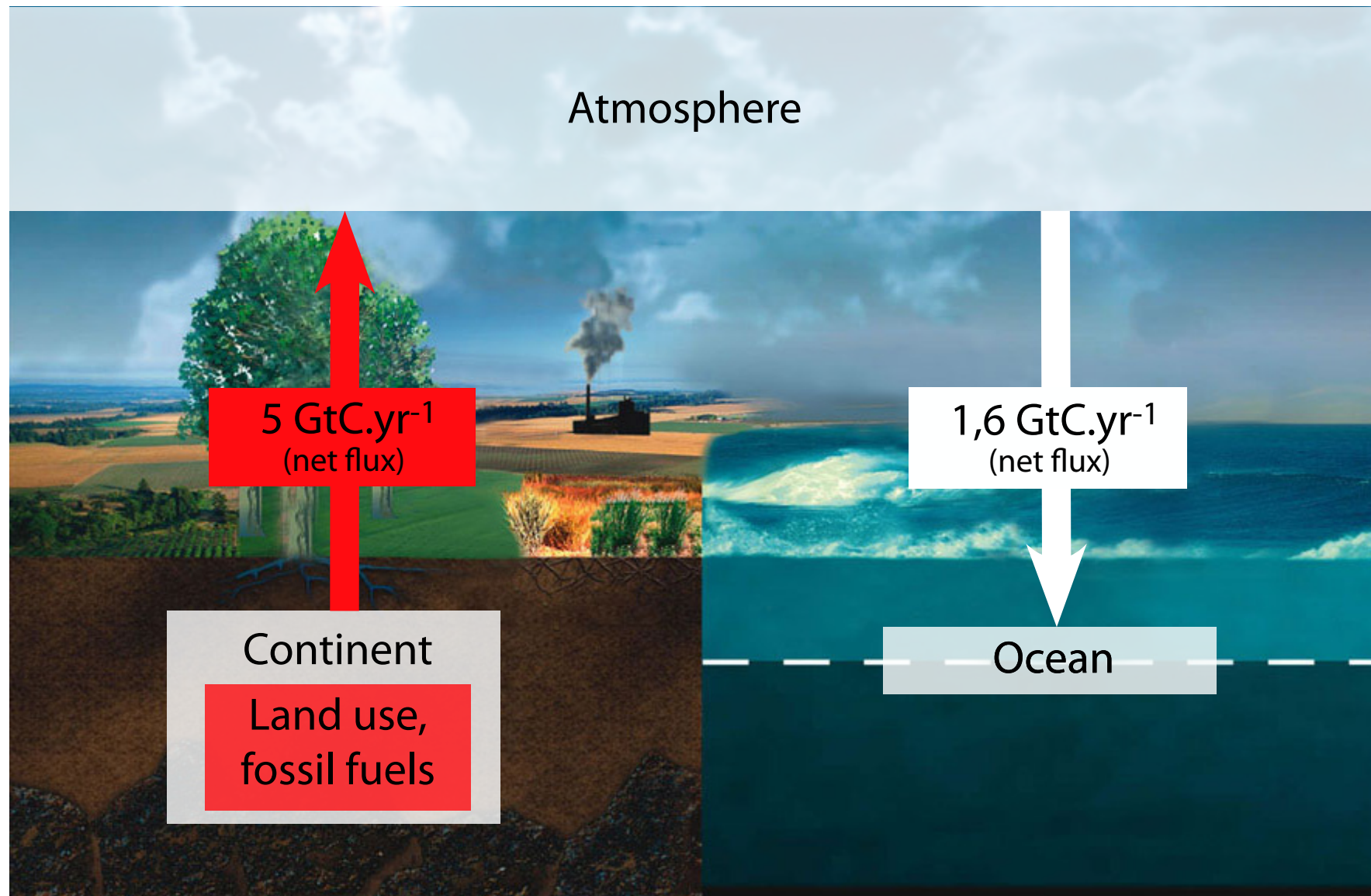
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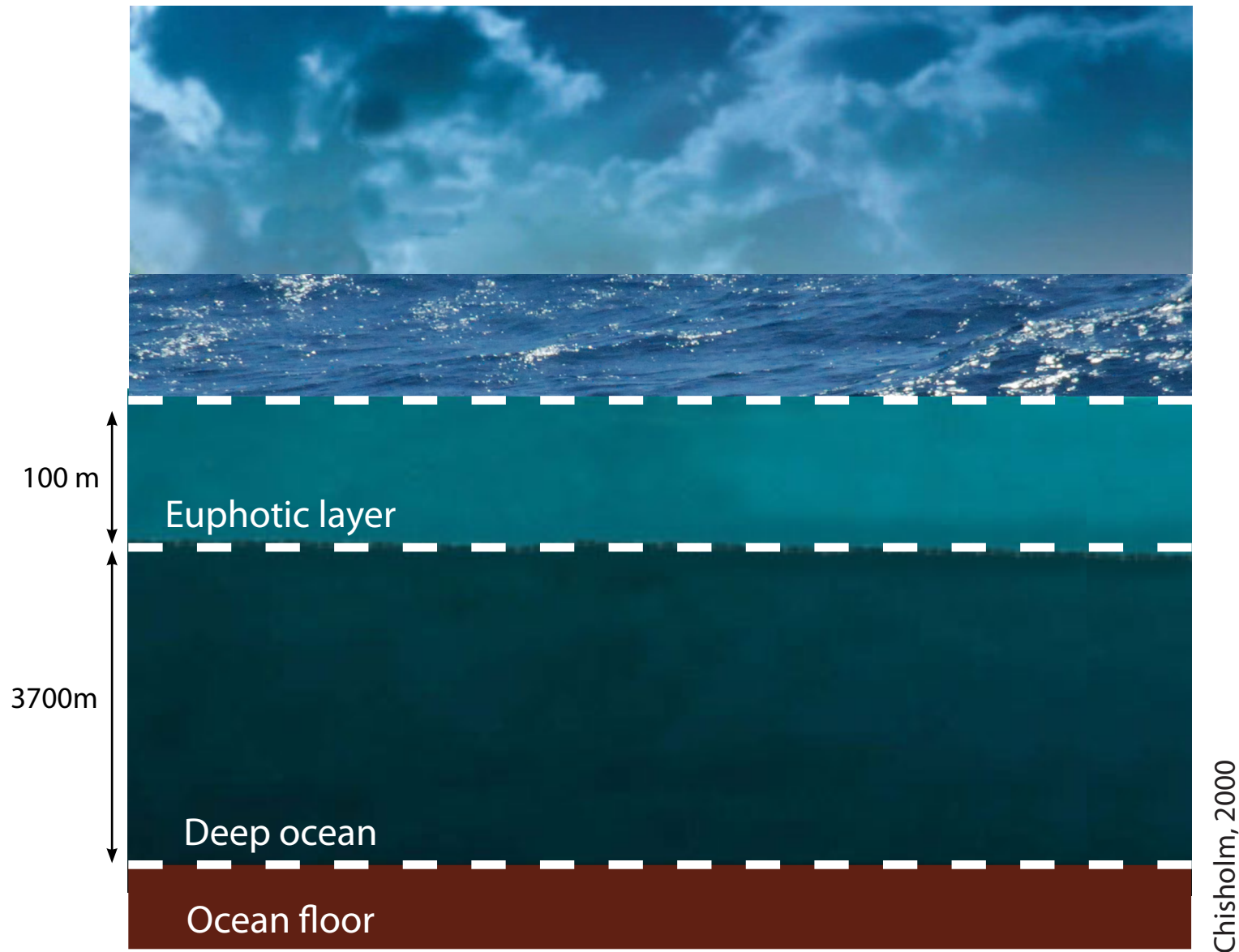
CO₂ level in the atmosphere since 400 000 years



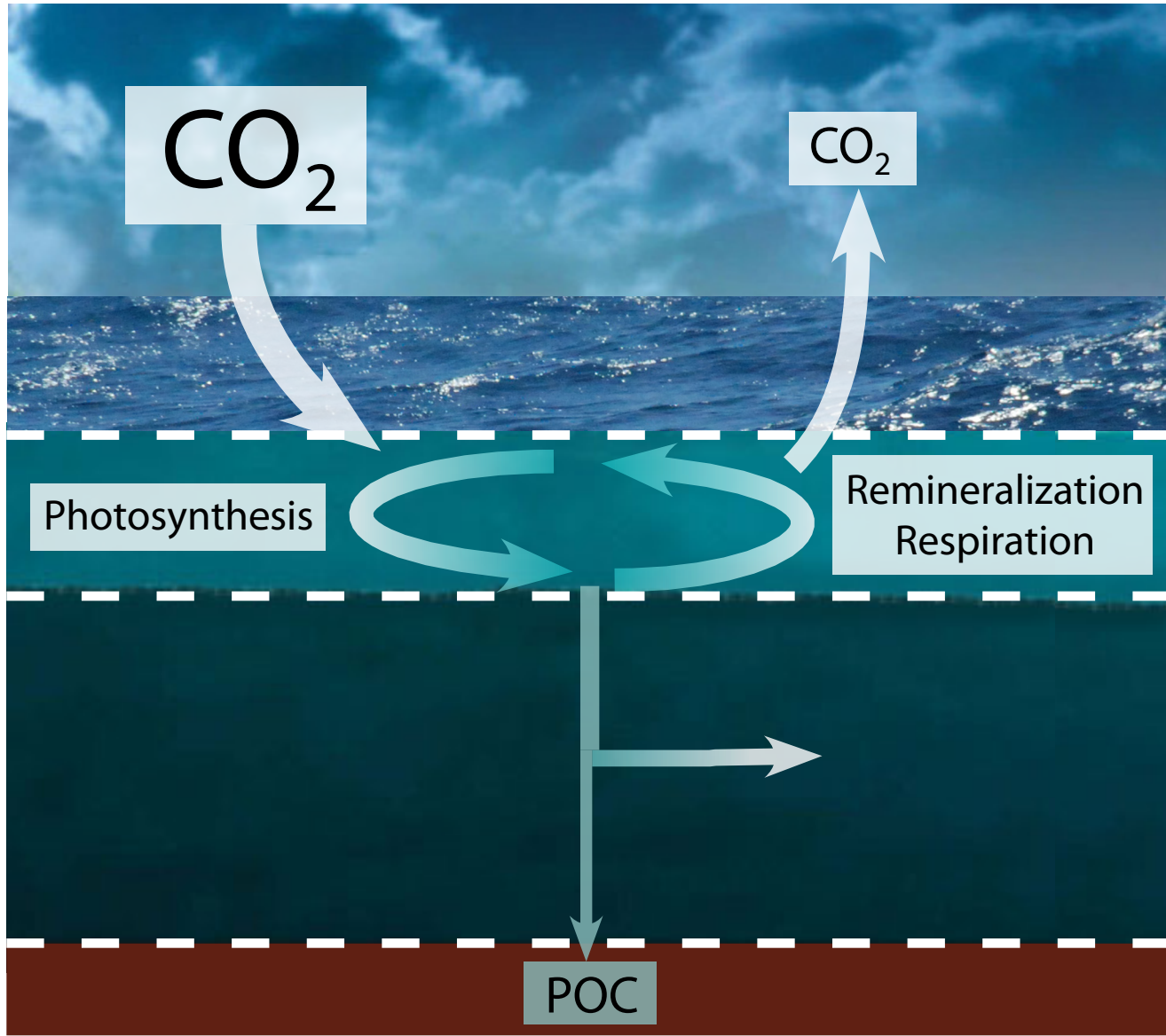
Carbon cycle



Oceanic CO₂ biological pump

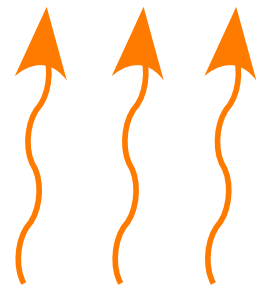
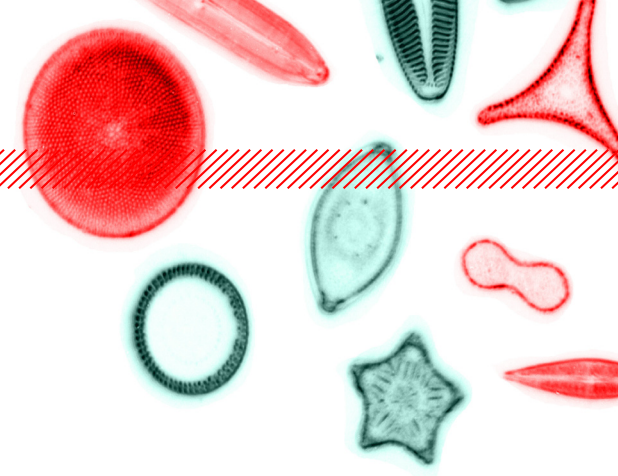


Oceanic CO₂ biological pump



Chisholm, 2000

Photosynthesis reaction



Sunlight



Jacques, 2006

Photosynthesis

Redfield ratio: C:N:P (106:16:1)

Redfield, 1963

Proportionnal uptake of:

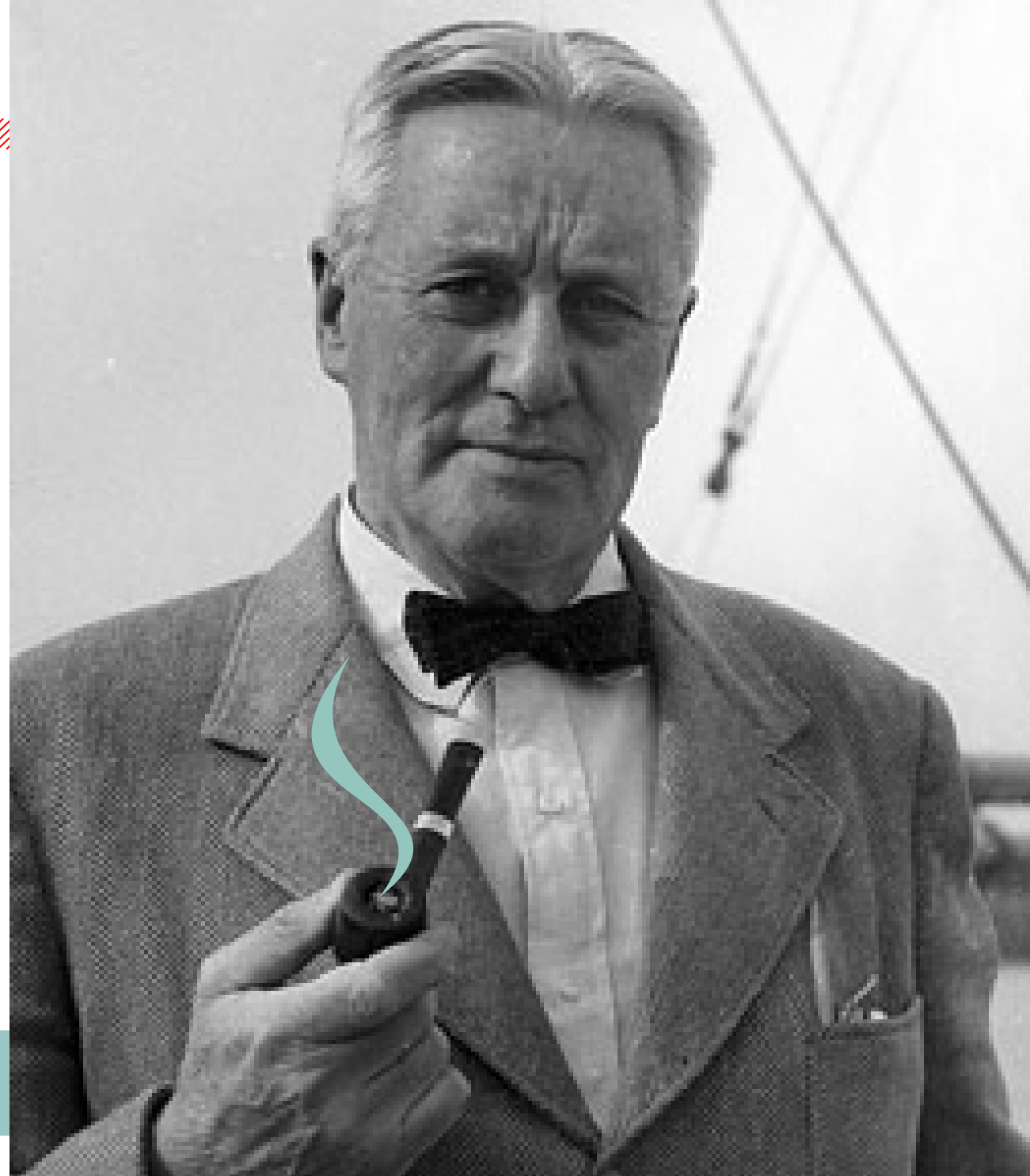
$\text{CO}_2 = C_{\text{inorganic}}$

Nitrogen

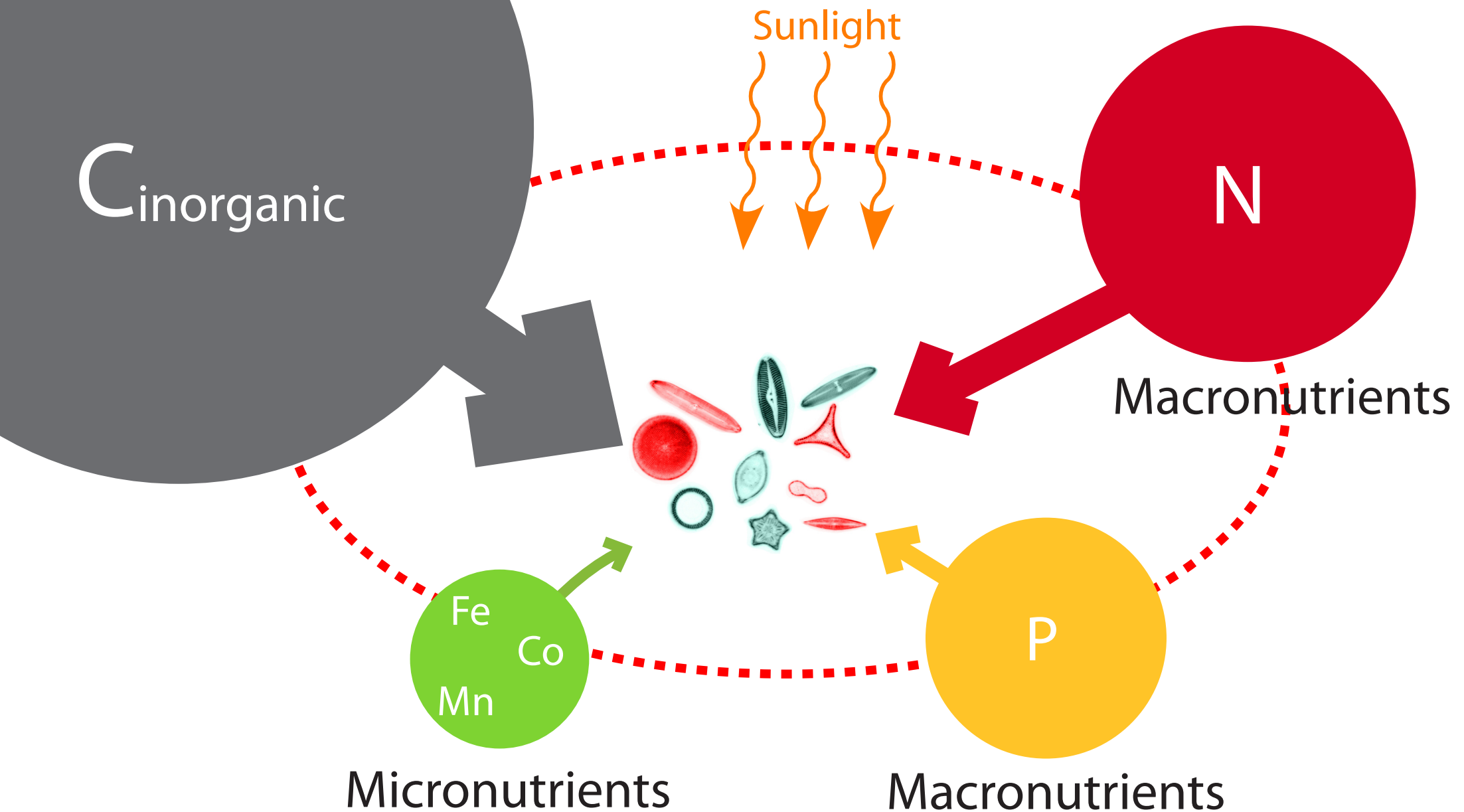
Phosphorous

Micronutrients

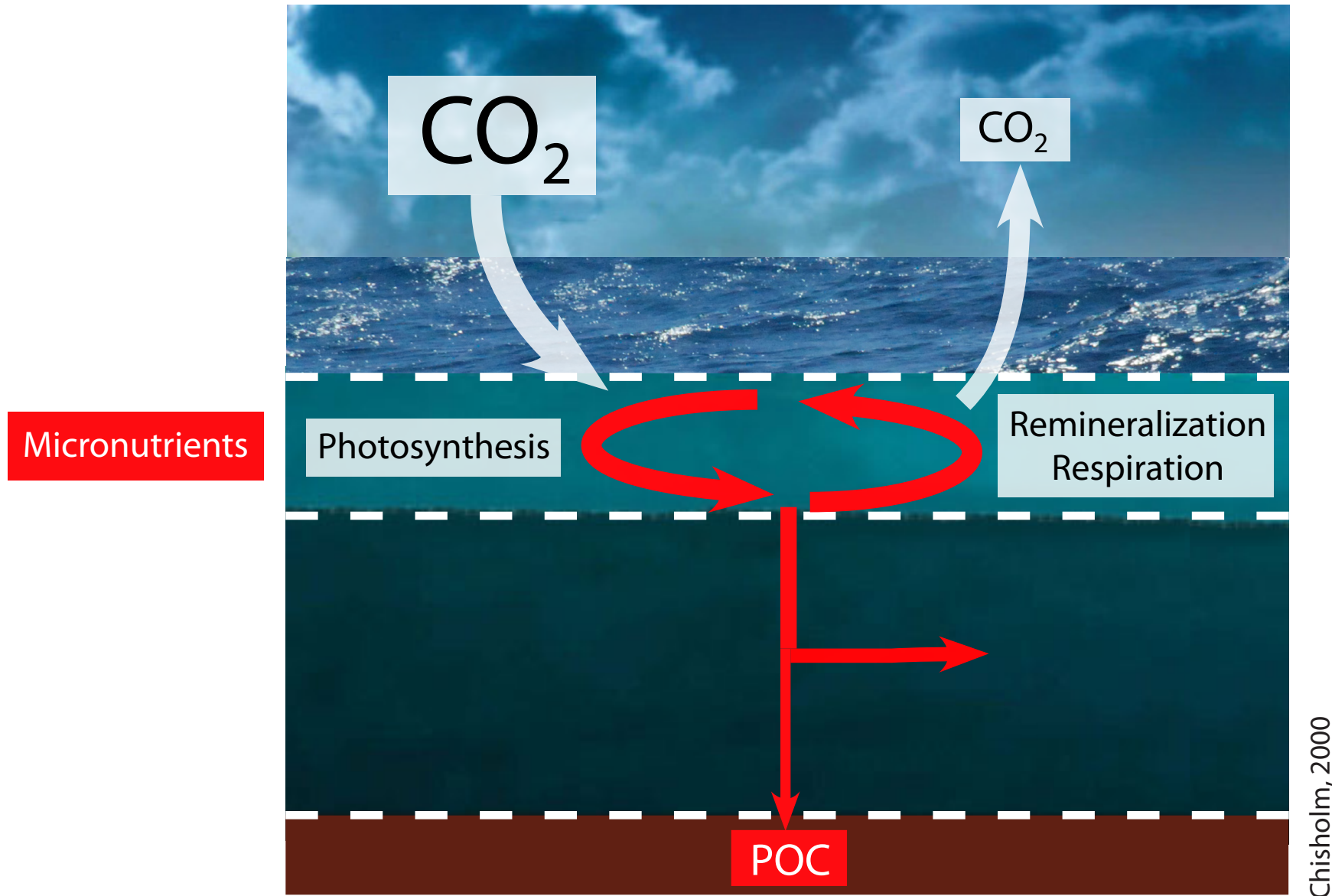
by phytoplankton.



Oceanic fluxes



Oceanic CO₂ biological pump



Bioavailability

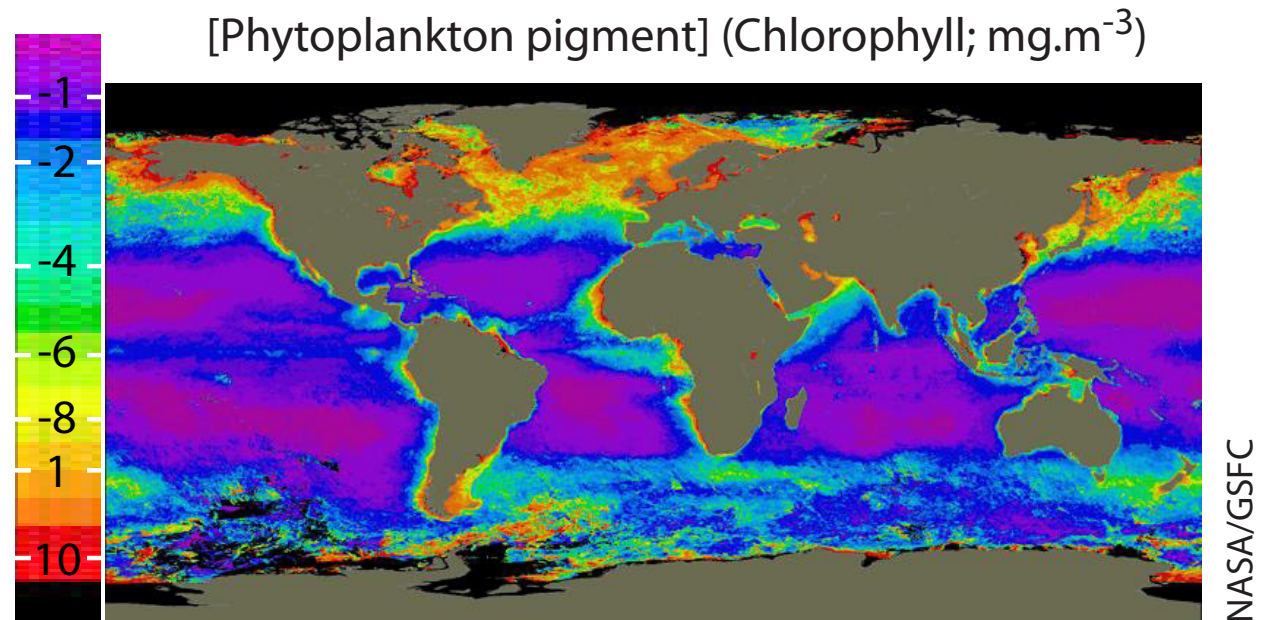
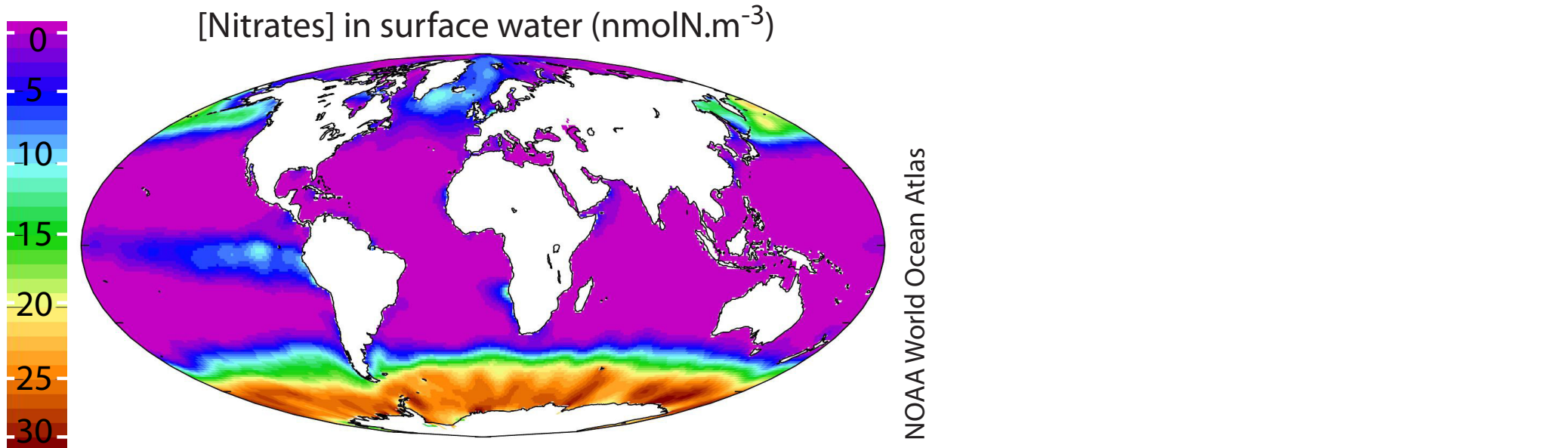


Sunlight

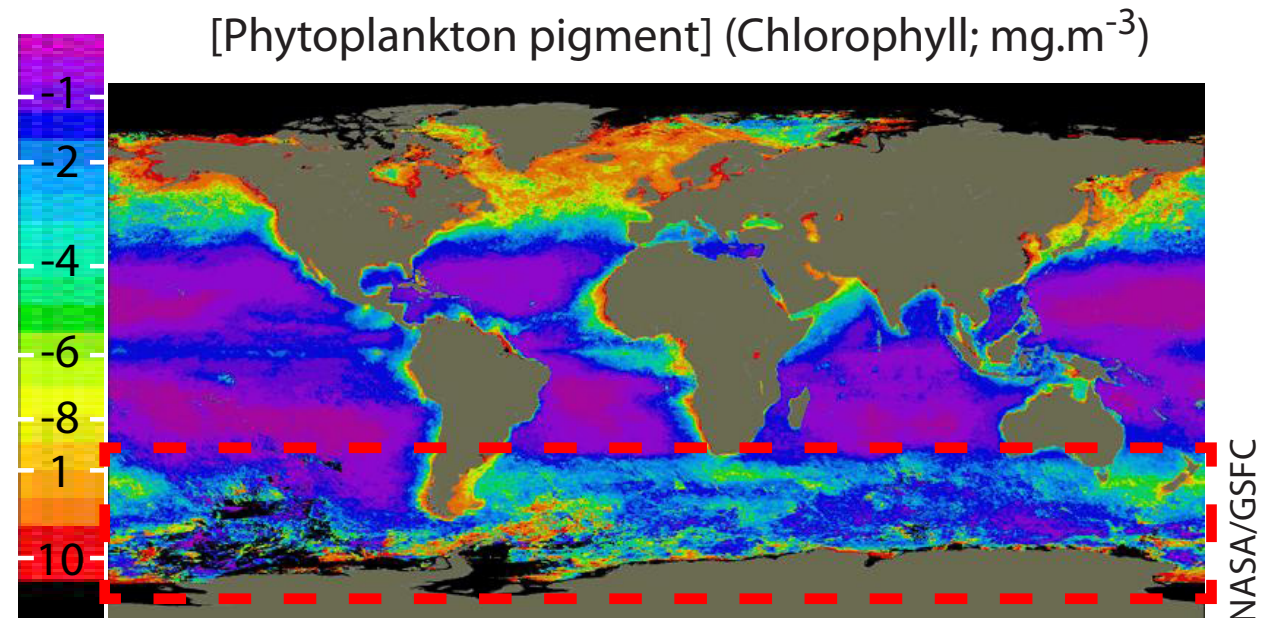
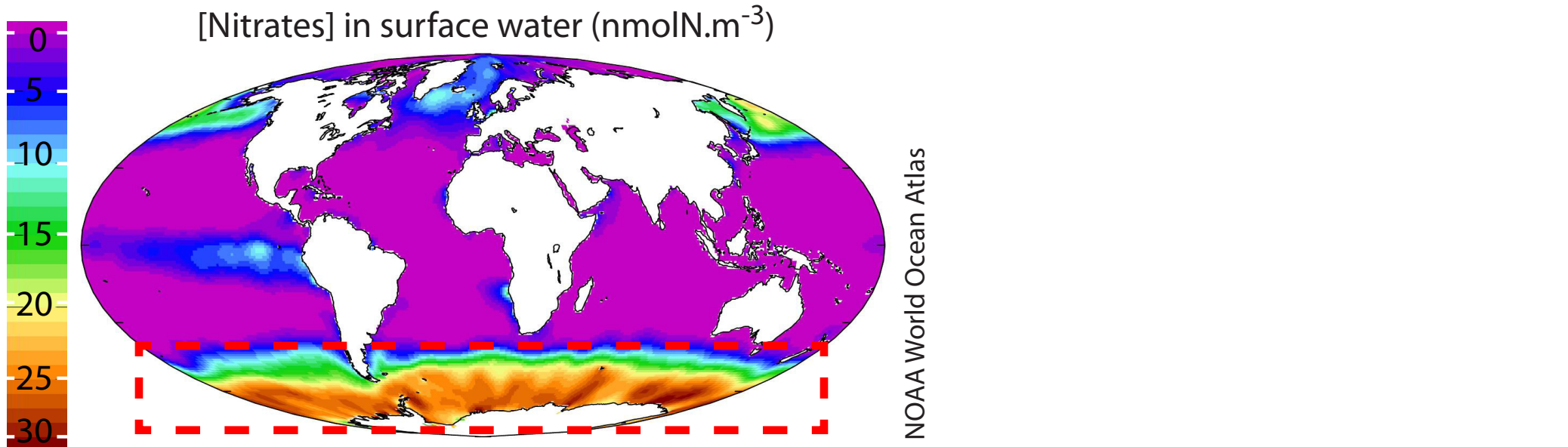


Soluble

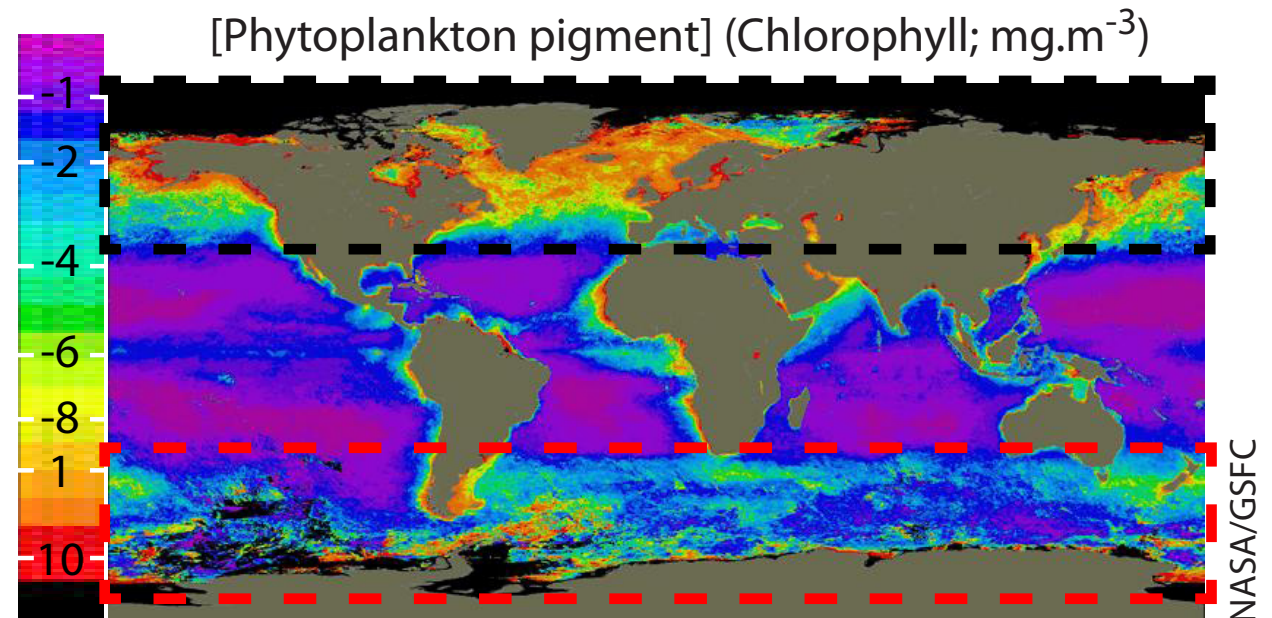
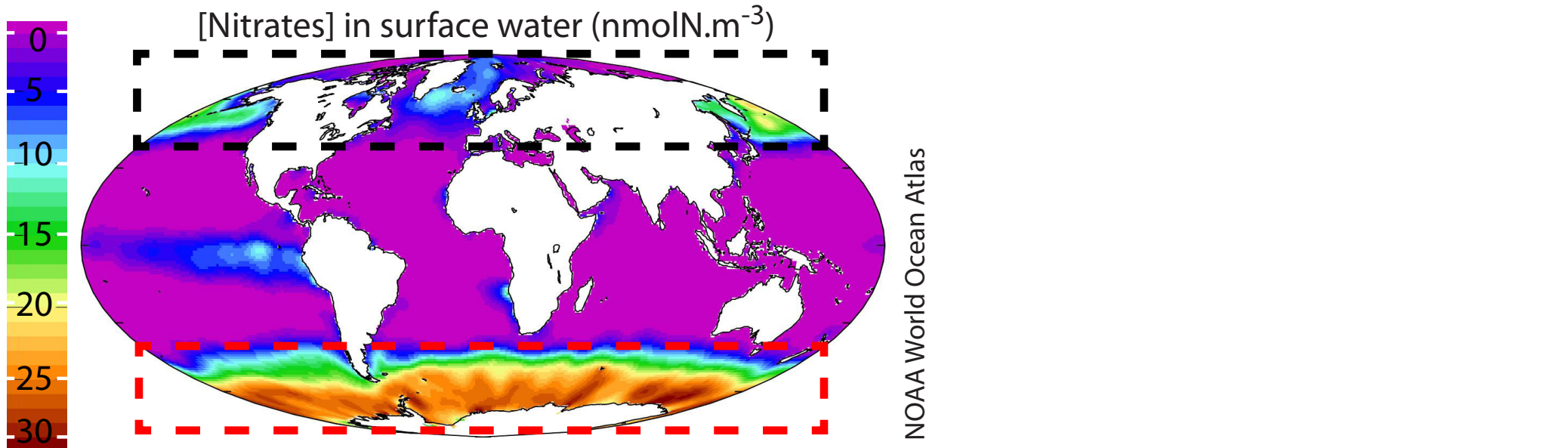
The Southern Ocean: HNLC area



The Southern Ocean: HNLC area



The Southern Ocean: HNLC area



Oceanic fluxes

C
inorganic

Sunlight

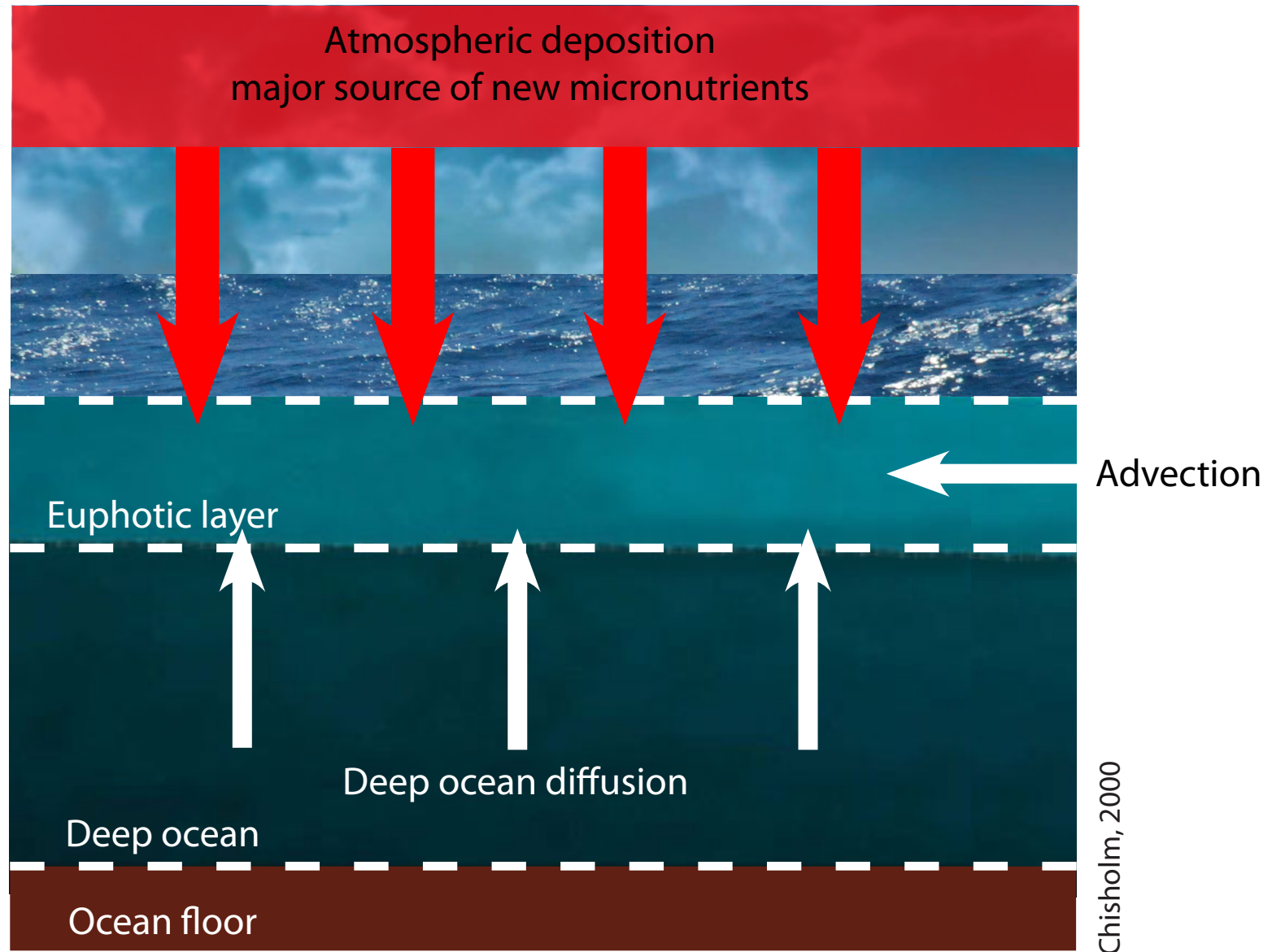
N

P

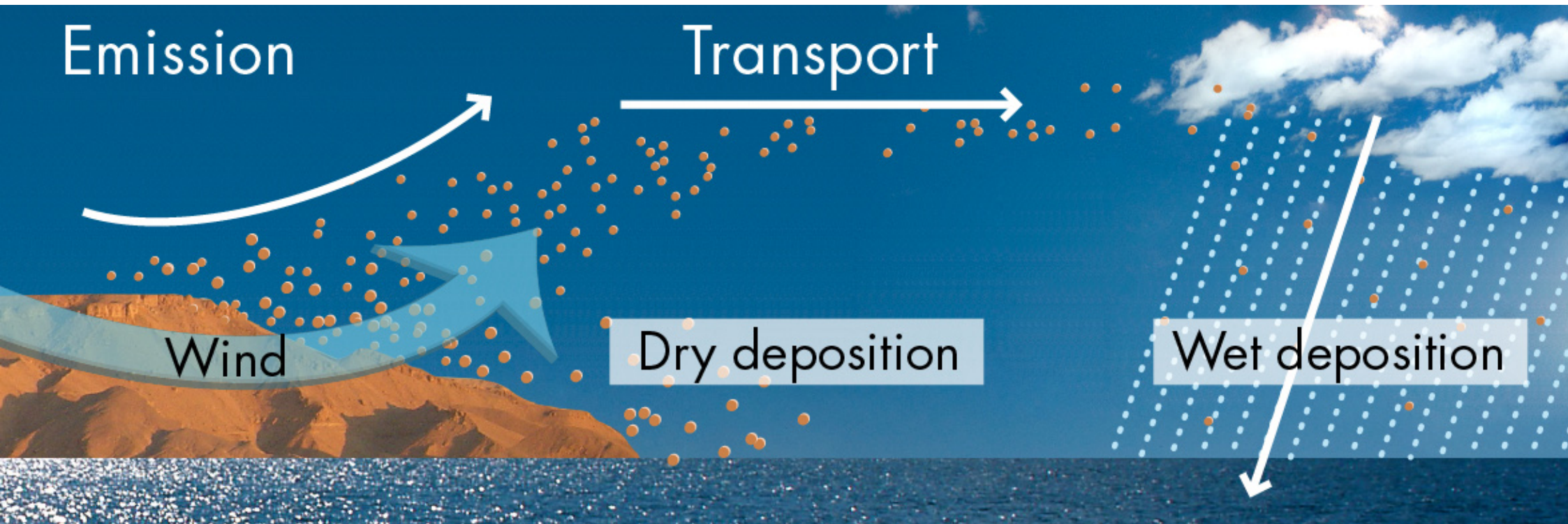
Fe
Mn
Co

Micronutrients
limited in the
Southern Ocean

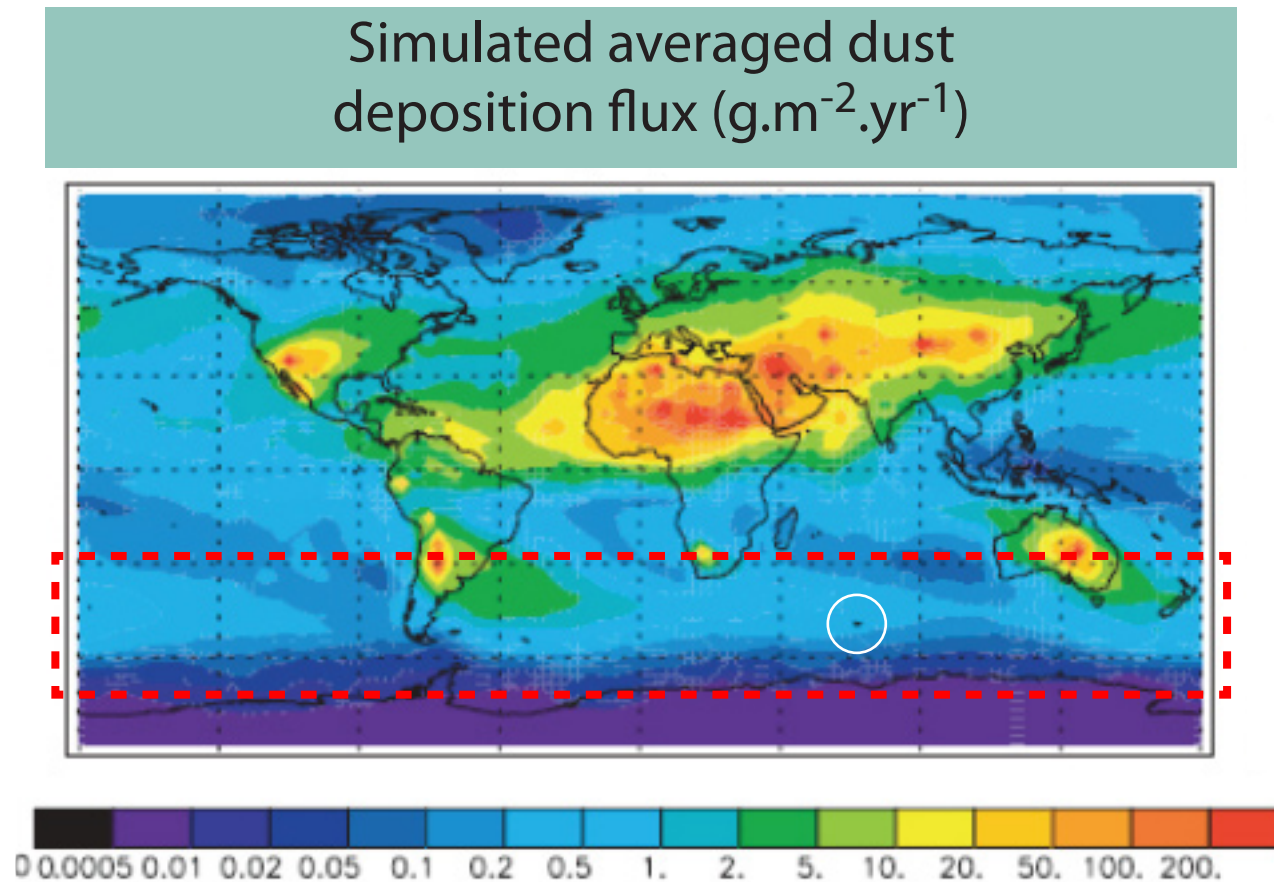
Micronutrient supplies



Dust cycle

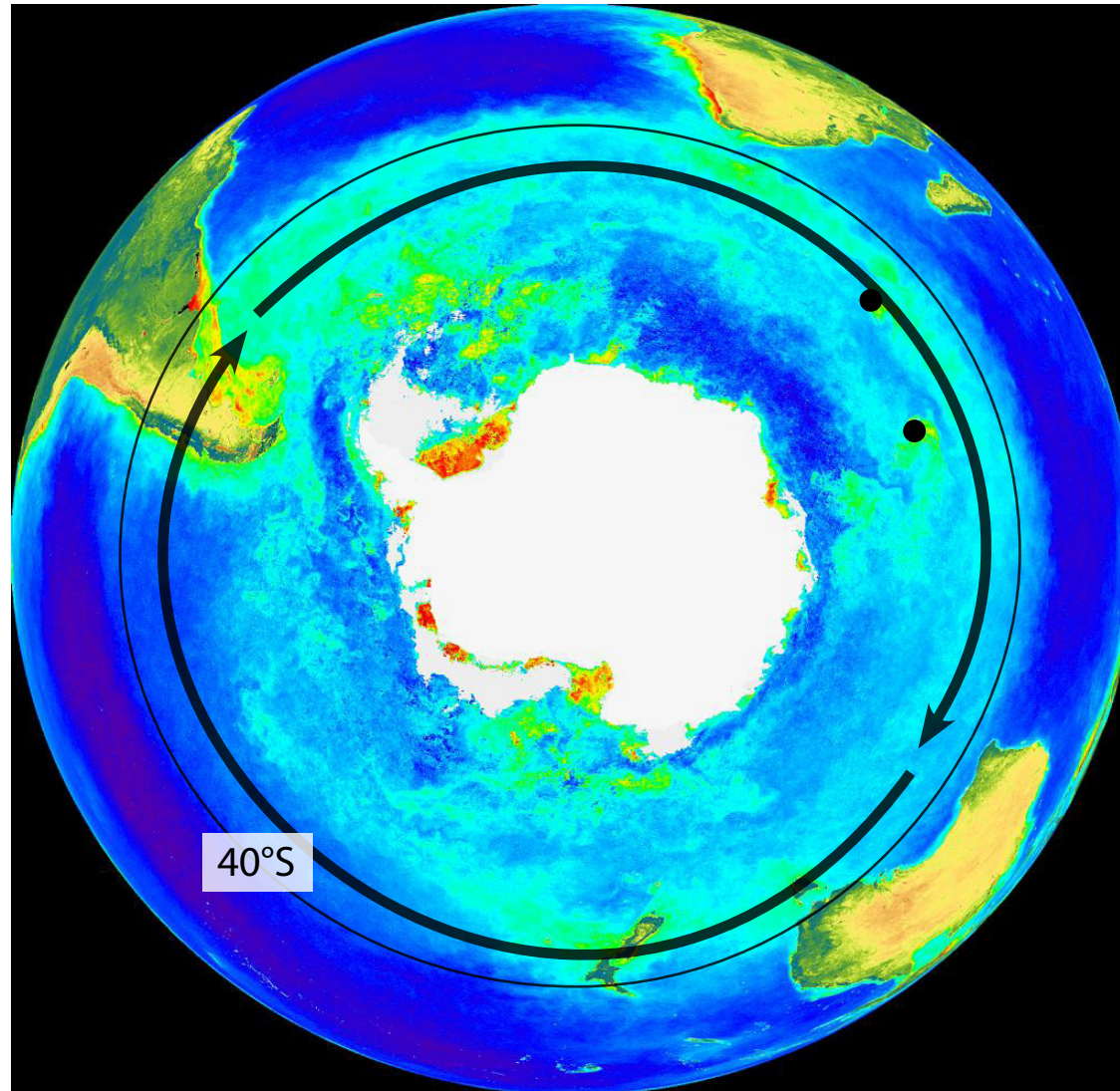


Atmospheric deposition to the open ocean



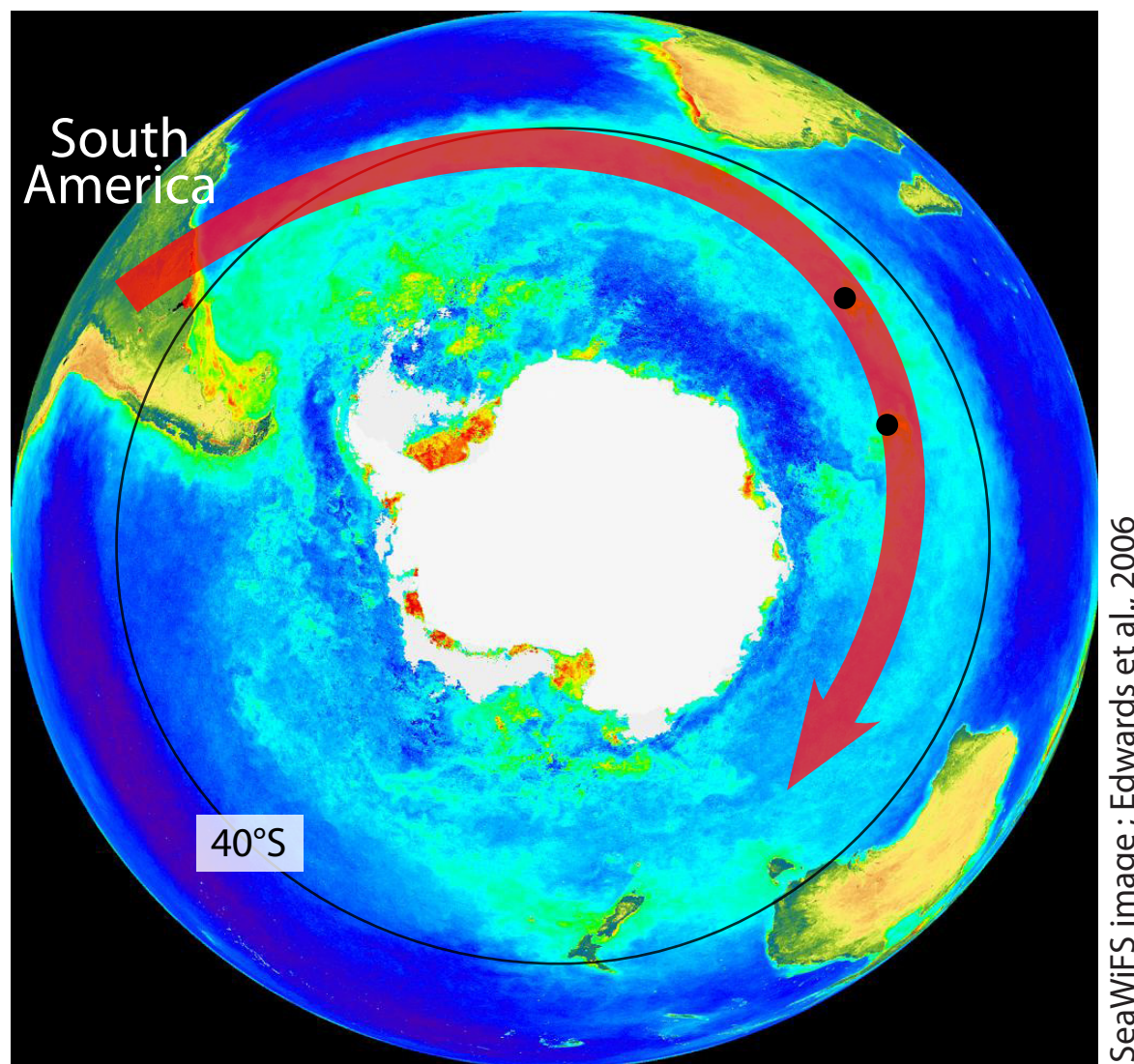
Low atmospheric deposition flux over the Southern Ocean

Dust sources in the South Hemisphere

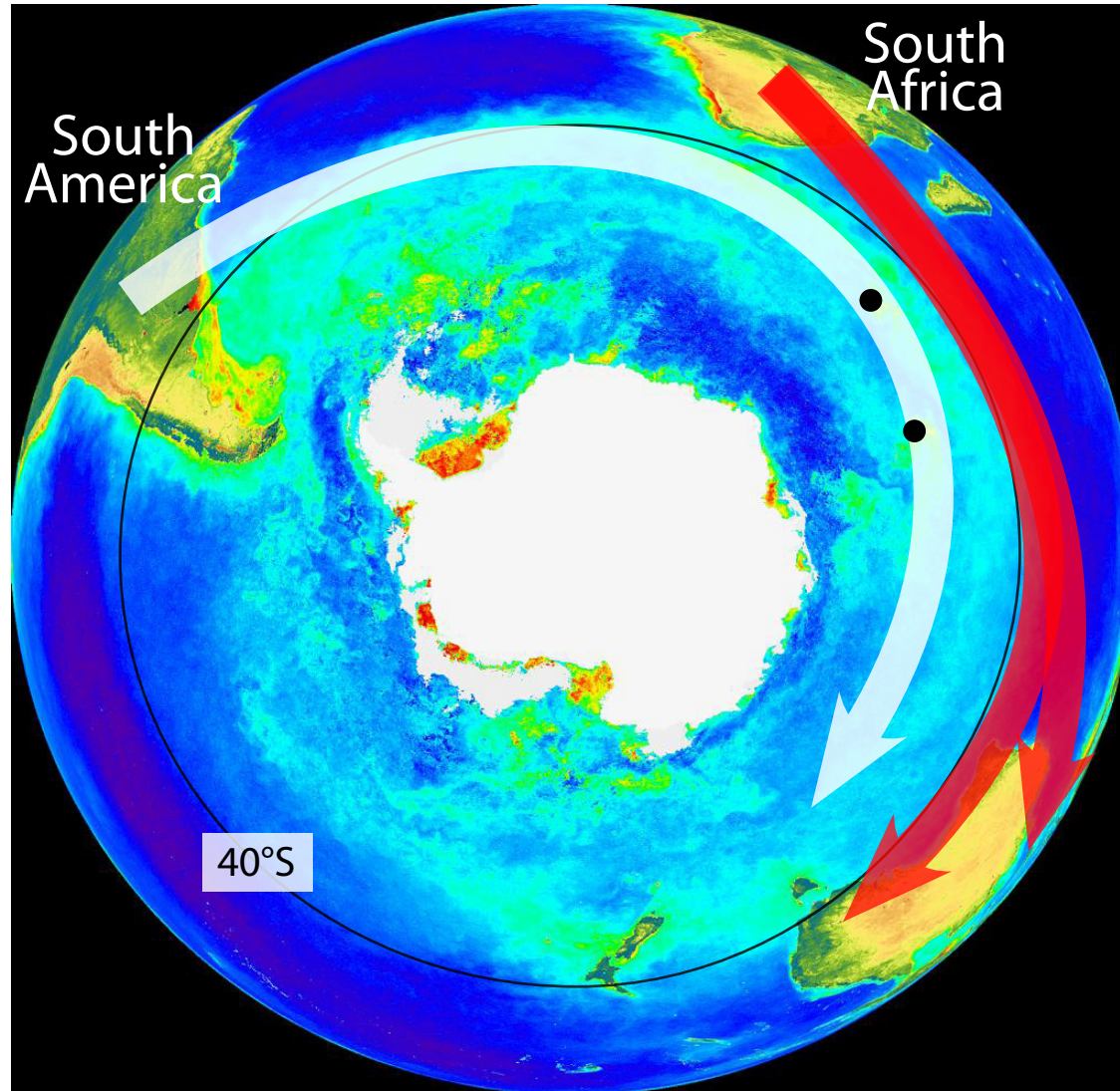


SeaWiFS image ; Edwards et al., 2006

Dust sources in the South Hemisphere

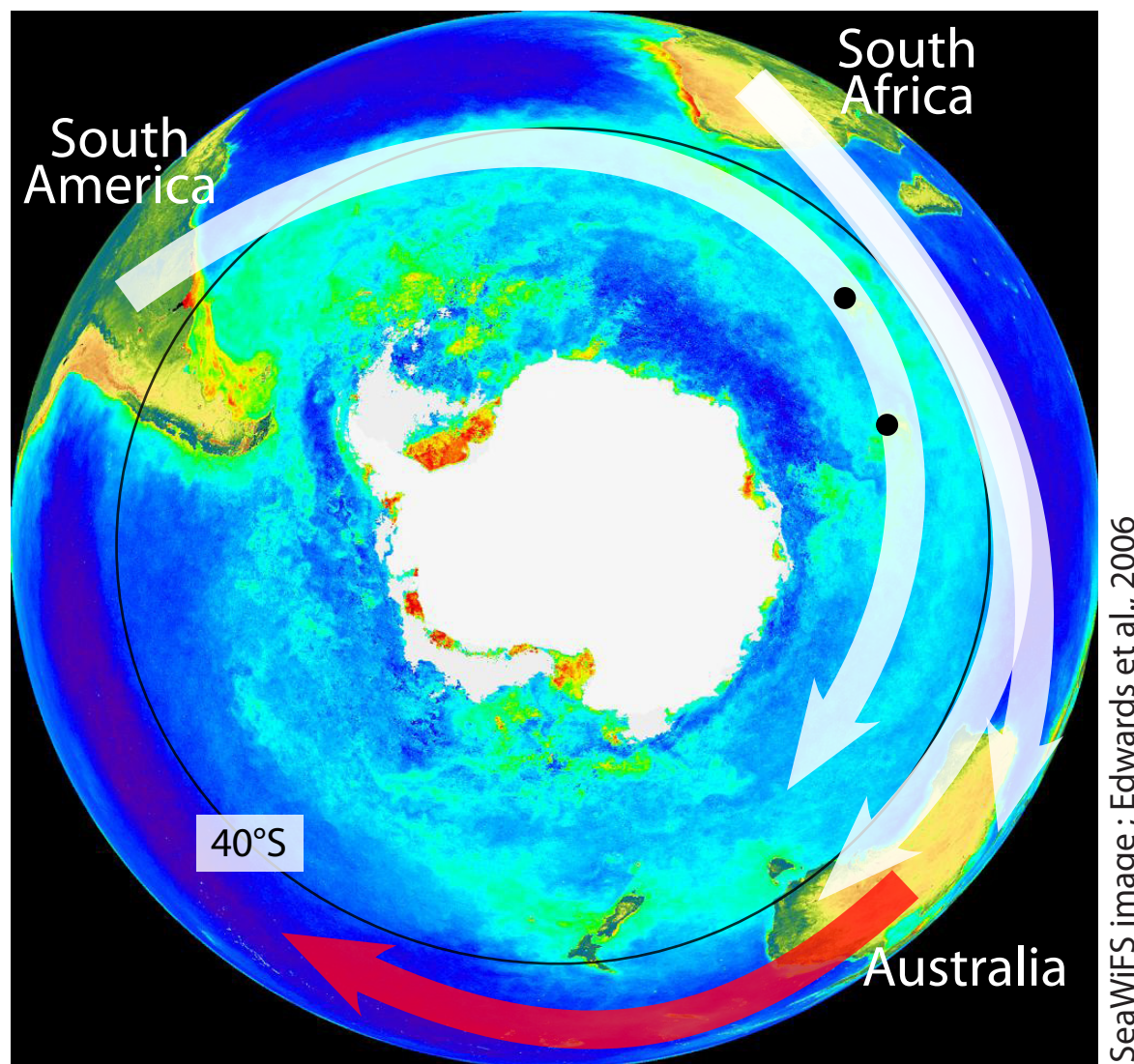


Dust sources in the South Hemisphere

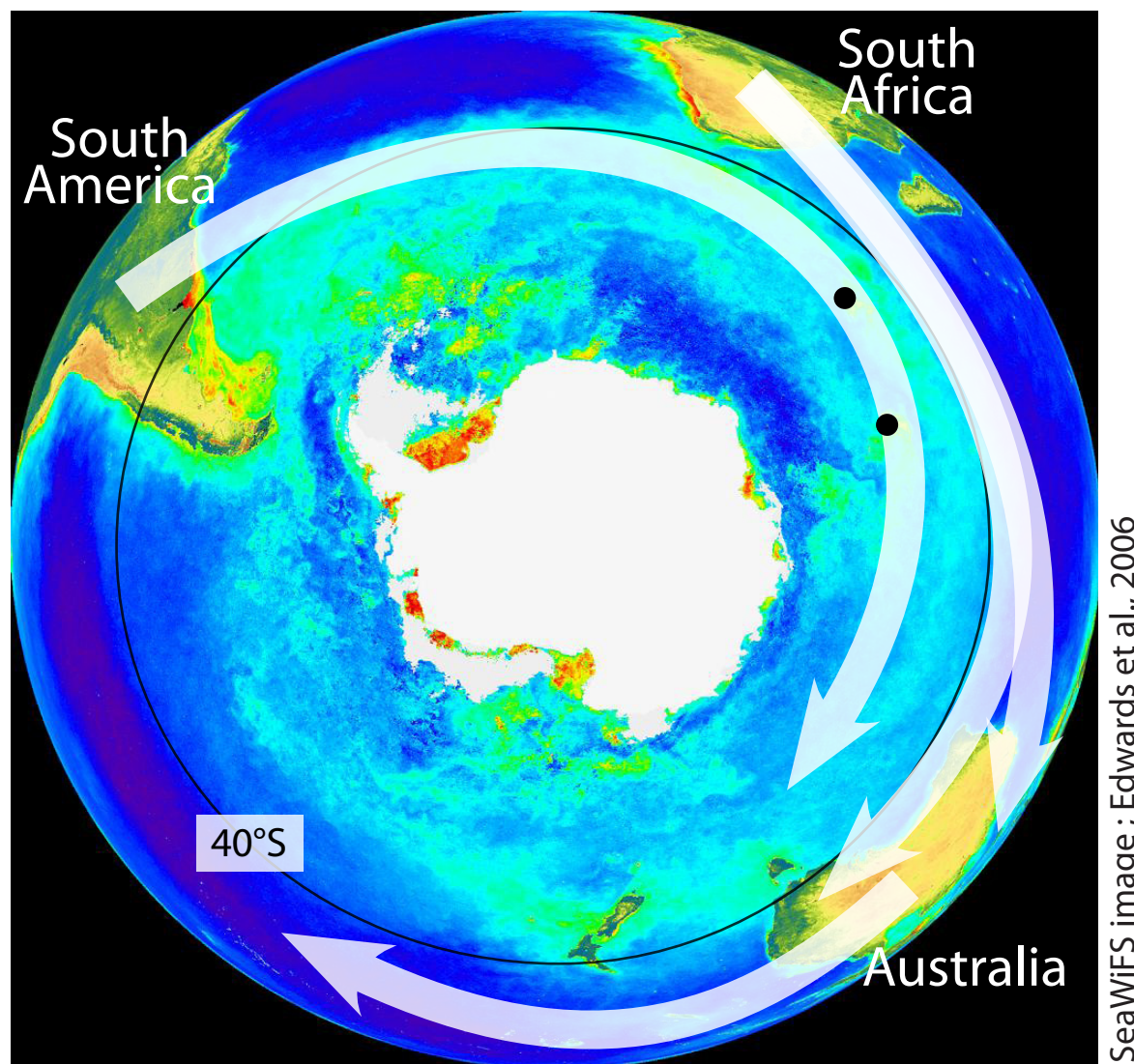


SeaWiFS image ; Edwards et al., 2006

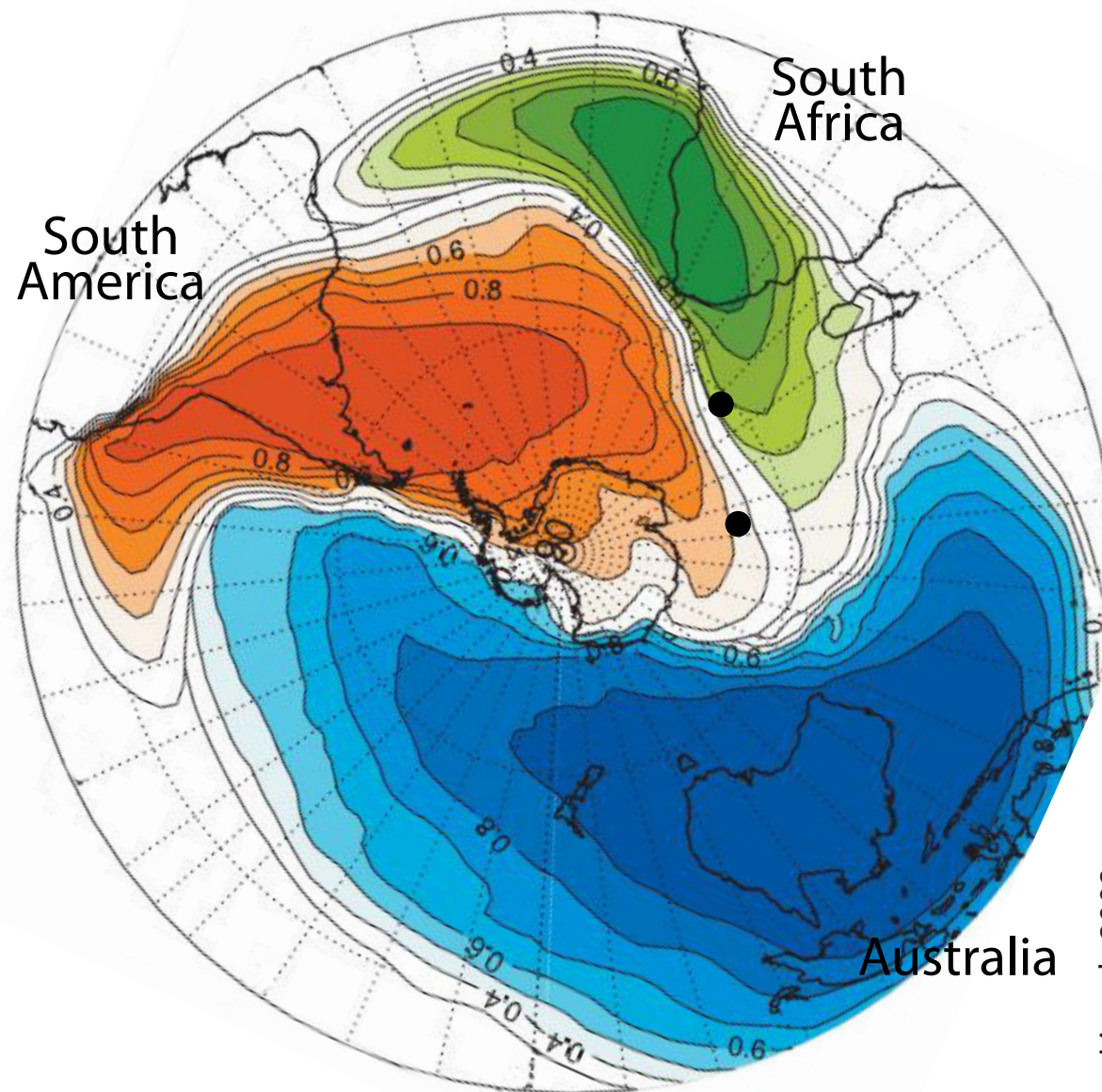
Dust sources in the South Hemisphere

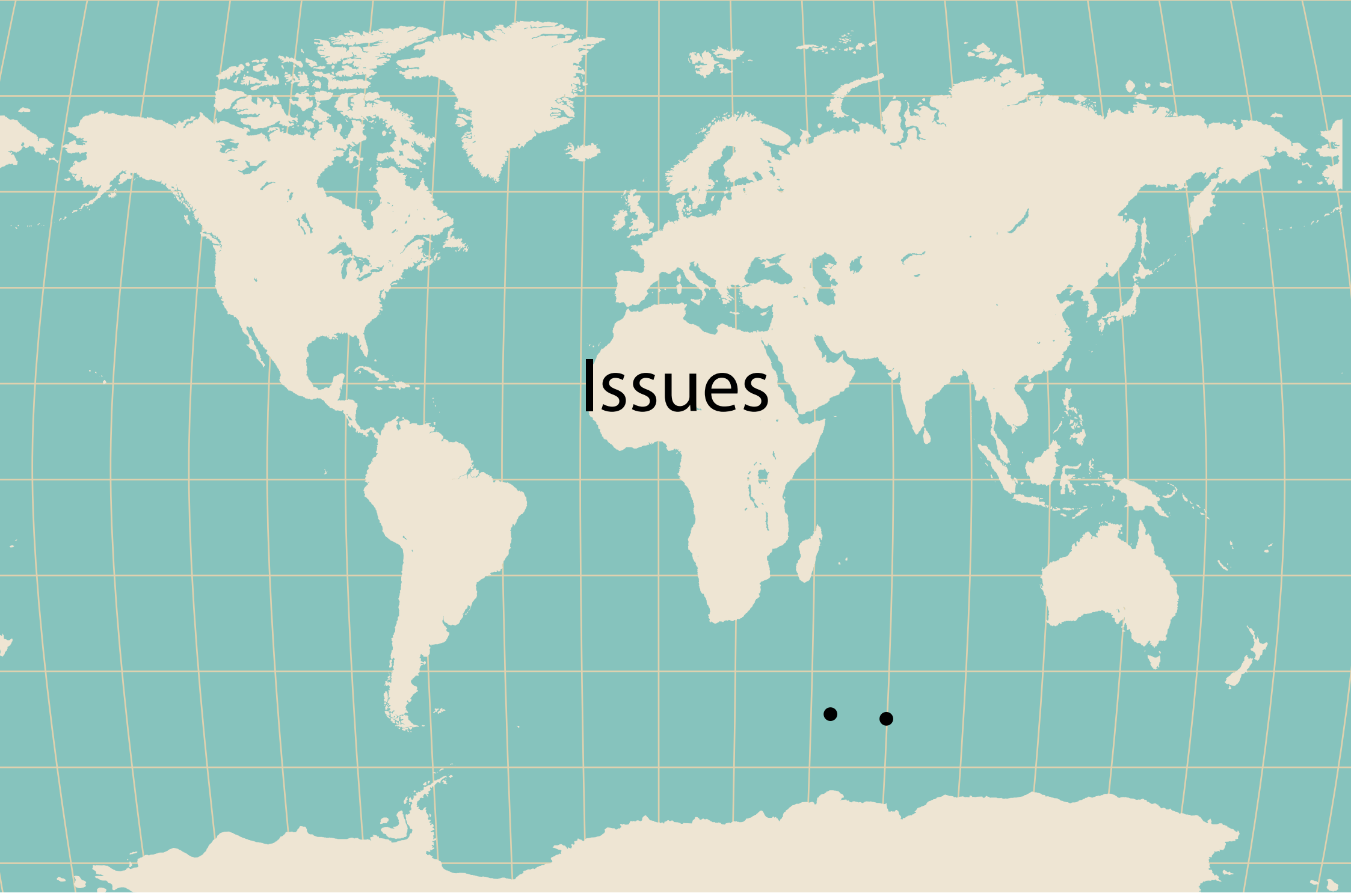


Dust sources in the South Hemisphere



Dust sources in the South Hemisphere





Issues

Scientific context

Issues

Methods

Results

What we know?

Atmospheric deposition is not well known
over the Southern Ocean

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Atmospheric deposition is not well known
over the Southern Ocean



Very low atmospheric concentrations:
measurements are very difficult to
performed, **adapted ultra clean work
conditions are essential!**

What we know?

Atmospheric deposition is not well known over the Southern Ocean

1. Only two studies based on observations available in literature before this work

→ Planquette et al. (2007) and Wagener et al. (2008):
sporadic offshore atmospheric measurements (aerosols, rainwater)
→ deposition fluxes (dust, iron, dissolved iron)

What we know?

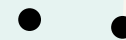
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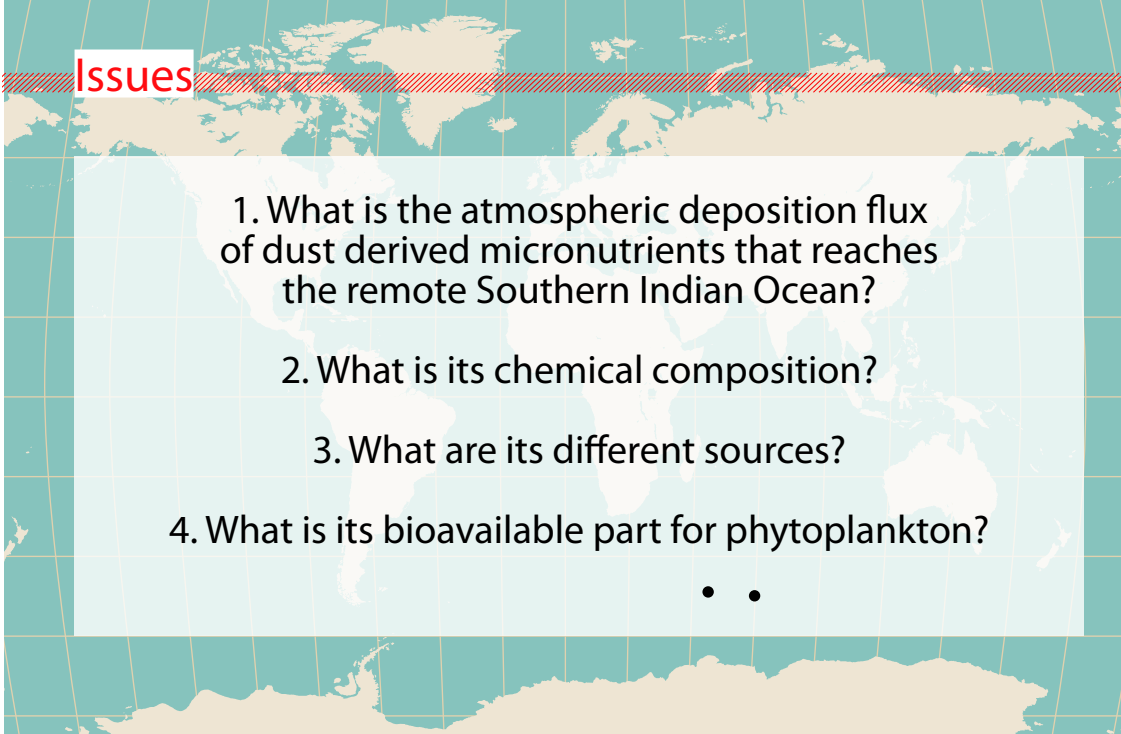
→ Planquette et al. (2007) and Wagener et al. (2008):
sporadic offshore atmospheric measurements (aerosols, rainwater)
→ deposition fluxes (dust, iron, dissolved iron)

2. Models need to be constrained and/or validated by atmospheric measurements over the Southern Ocean (deposition, chemical composition, solubility, dust sources)

1. What is the atmospheric deposition flux of dust derived micronutrients that reaches the remote Southern Indian Ocean?
2. What is its chemical composition?
3. What are its different sources?
4. What is its bioavailable part for phytoplankton?



Strategy



Issues

1. What is the atmospheric deposition flux of dust derived micronutrients that reaches the remote Southern Indian Ocean?
2. What is its chemical composition?
3. What are its different sources?
4. What is its bioavailable part for phytoplankton?

• •

Long term observations on atmospheric deposition over the South Indian Ocean

A world map with a light blue background and a white grid. The map shows the continents in a light beige color. Two small red dots are located in the southern ocean, south of Africa and India. The word "Methods" is written in a large, bold, black font in the center of the map.

Methods

Scientific context

Issues

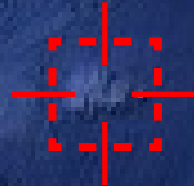
Methods

Results

FLATOCOA

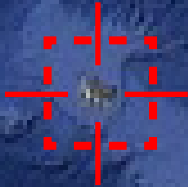
« FLux Atmosphérique d'Origine Continentale sur l'Océan Austral »

Crozet



46°S 51°E

Kerguelen Islands



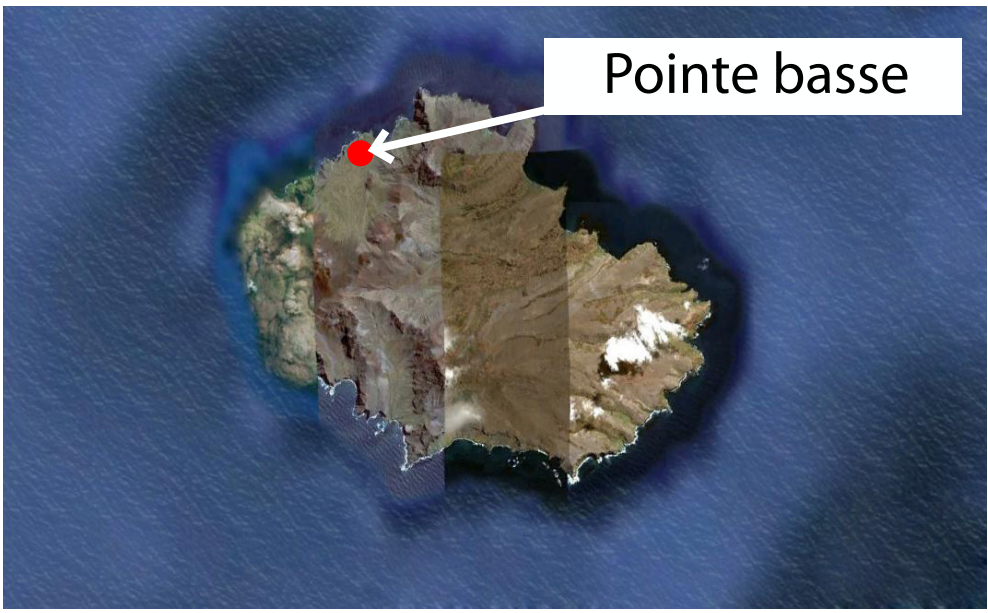
49°S 70°E

Trace metal measurements
on a continuous basis

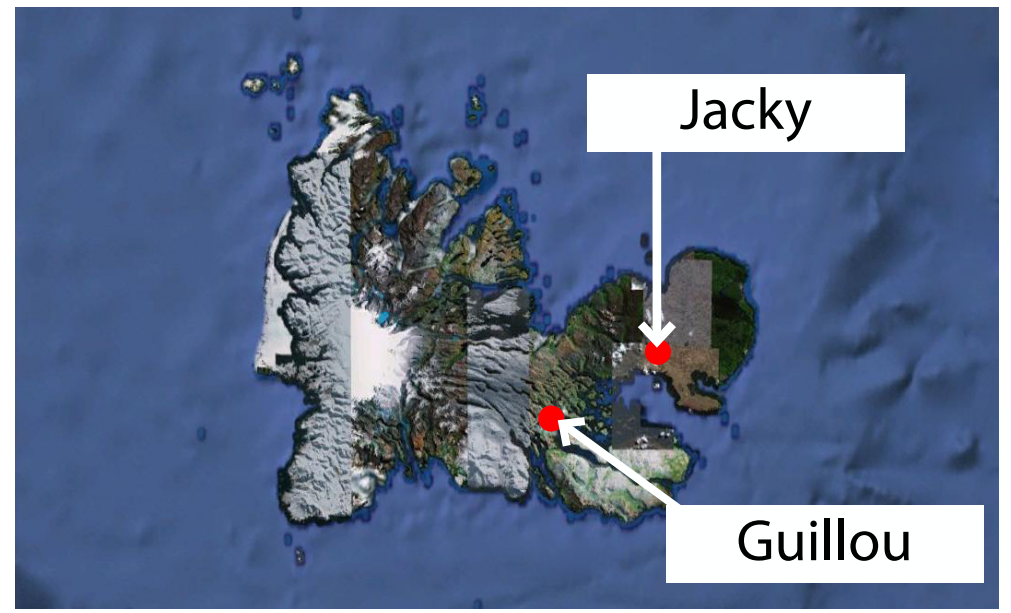
Two-year sampling
(end of 2008 - end of 2010)



Sampling sites



Crozet



Kerguelen

Material preparation

- Under ISO1 laminar flow bench in ISO5 clean room
- Air quality on Kerguelen Islands: ISO4 (analog)



Concentration (particle.m⁻³)

Class	0,1 μ	0,2 μ	0,3 μ	0,5 μ	1 μ	5 μ
ISO 1	10	2	0	0	0	0
ISO 4	10000	10000	1020	352	83	0

Total atmospheric deposition sampling

Duplicat systems (A, B)

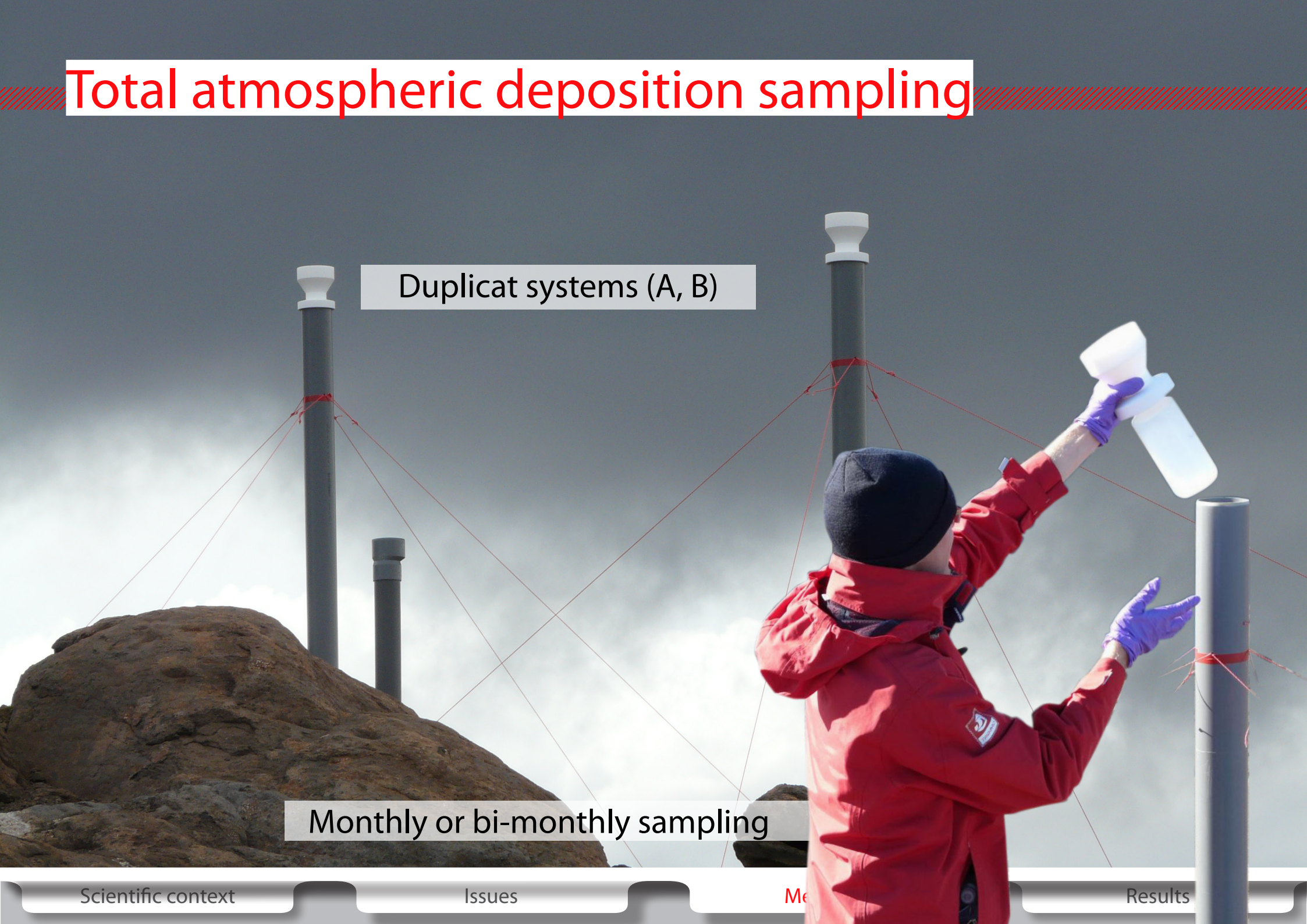
Monthly or bi-monthly sampling

Scientific context

Issues

Me

Results



37 analyzed and validated elements

ICP-AES and HR-ICP-MS
(clean room)

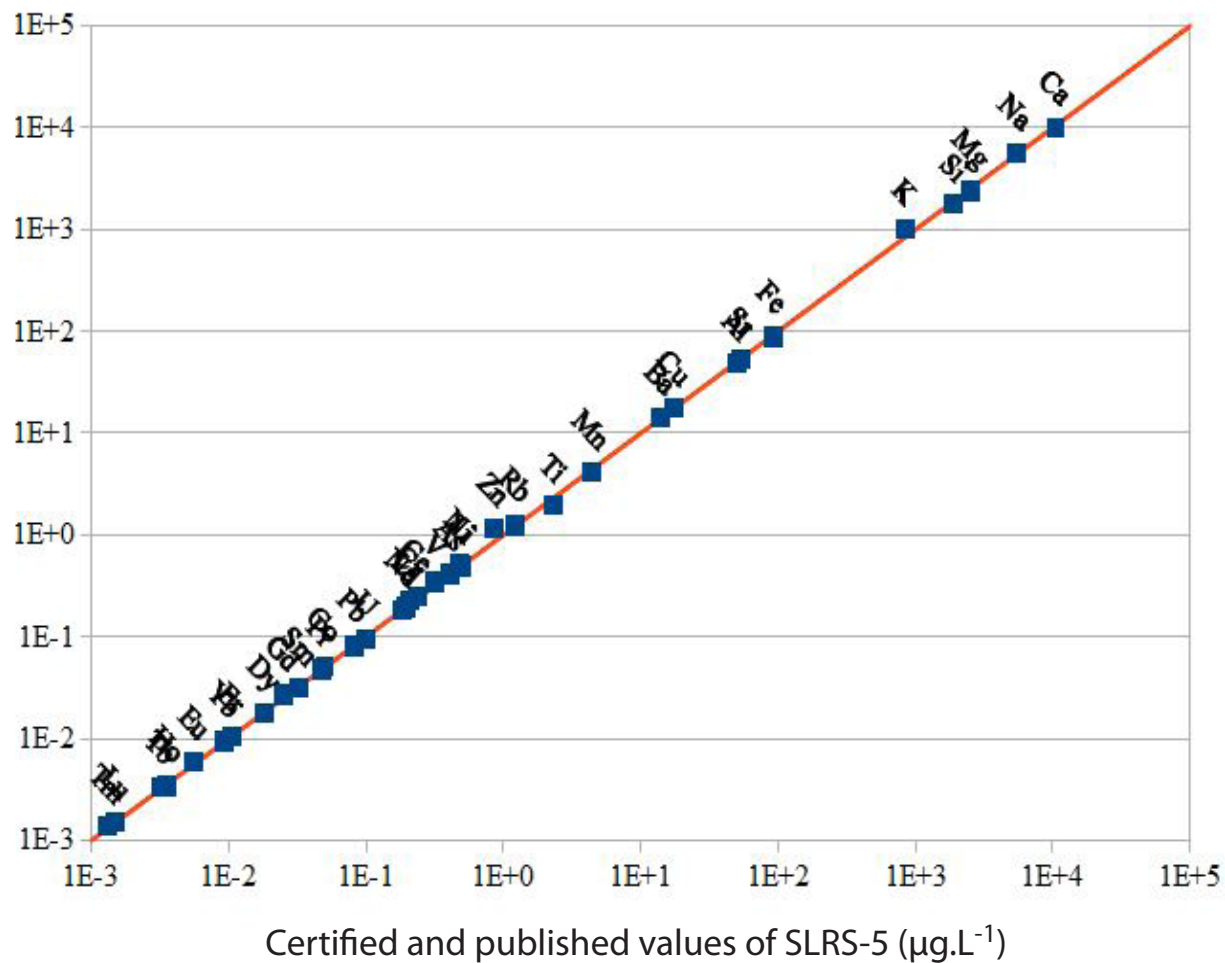


hydrogen 1 H 1.0079																	helium 2 He 4.0026						
lithium 3 Li 6.941	beryllium 4 Be 9.0122																	boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180
sodium 11 Na 22.990	magnesium 12 Mg 24.305																	aluminum 13 Al 26.982	silicon 14 Si 28.086	phosphorus 15 P 30.974	sulfur 16 S 32.065	chlorine 17 Cl 35.453	argon 18 Ar 39.948
potassium 19 K 39.098	calcium 20 Ca 40.078	scandium 21 Sc 44.956	titanium 22 Ti 47.867	vanadium 23 V 50.942	chromium 24 Cr 51.996	manganese 25 Mn 54.938	iron 26 Fe 55.845	cobalt 27 Co 58.933	nickel 28 Ni 58.693	copper 29 Cu 63.546	zinc 30 Zn 65.39	gallium 31 Ga 69.723	germanium 32 Ge 72.61	arsenic 33 As 74.922	selenium 34 Se 78.96	bromine 35 Br 79.904	krypton 36 Kr 83.80						
rubidium 37 Rb 85.468	strontium 38 Sr 87.62	yttrium 39 Y 88.906	zirconium 40 Zr 91.224	niobium 41 Nb 92.906	molybdenum 42 Mo 95.94	technetium 43 Tc [98]	ruthenium 44 Ru 101.07	rhodium 45 Rh 102.91	palladium 46 Pd 106.42	silver 47 Ag 107.87	cadmium 48 Cd 112.41	indium 49 In 114.82	tin 50 Sn 118.71	antimony 51 Sb 121.76	tellurium 52 Te 127.60	iodine 53 I 126.90	xenon 54 Xe 131.29						
caesium 55 Cs 132.91	barium 56 Ba 137.33	57-70 *	lutetium 71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	platinum 78 Pt 195.08	gold 79 Au 196.97	mercury 80 Hg 200.59	thallium 81 Tl 204.38	lead 82 Pb 207.2	bismuth 83 Bi 208.98	polonium 84 Po [209]	astatine 85 At [210]	radon 86 Rn [222]					
francium 87 Fr [223]	radium 88 Ra [226]	89-102 * *	lawrencium 103 Lr [262]	rutherfordium 104 Rf [261]	dubnium 105 Db [262]	seaborgium 106 Sg [266]	bohrium 107 Bh [264]	hassium 108 Hs [269]	meitnerium 109 Mt [268]	ununilium 110 Uun [271]	unununium 111 Uuu [272]	ununbium 112 Uub [277]	ununquadium 114 Uuq [289]										

lanthanum 57 La 138.91	cerium 58 Ce 140.12	praseodymium 59 Pr 140.91	neodymium 60 Nd 144.24	promethium 61 Pm [145]	samarium 62 Sm 150.36	europium 63 Eu 151.96	gadolinium 64 Gd 157.25	terbium 65 Tb 158.93	dysprosium 66 Dy 162.50	holmium 67 Ho 164.93	erbium 68 Er 167.26	thulium 69 Tm 168.93	ytterbium 70 Yb 173.04
actinium 89 Ac [227]	thorium 90 Th 232.04	protactinium 91 Pa 231.04	uranium 92 U 238.03	neptunium 93 Np [237]	plutonium 94 Pu [244]	americium 95 Am [243]	curium 96 Cm [247]	berkelium 97 Bk [247]	californium 98 Cf [251]	einsteinium 99 Es [252]	fermium 100 Fm [257]	mendelevium 101 Md [258]	nobelium 102 No [259]

Analytical validation

Measured values
of SLRS-5
($\mu\text{g}\cdot\text{L}^{-1}$)



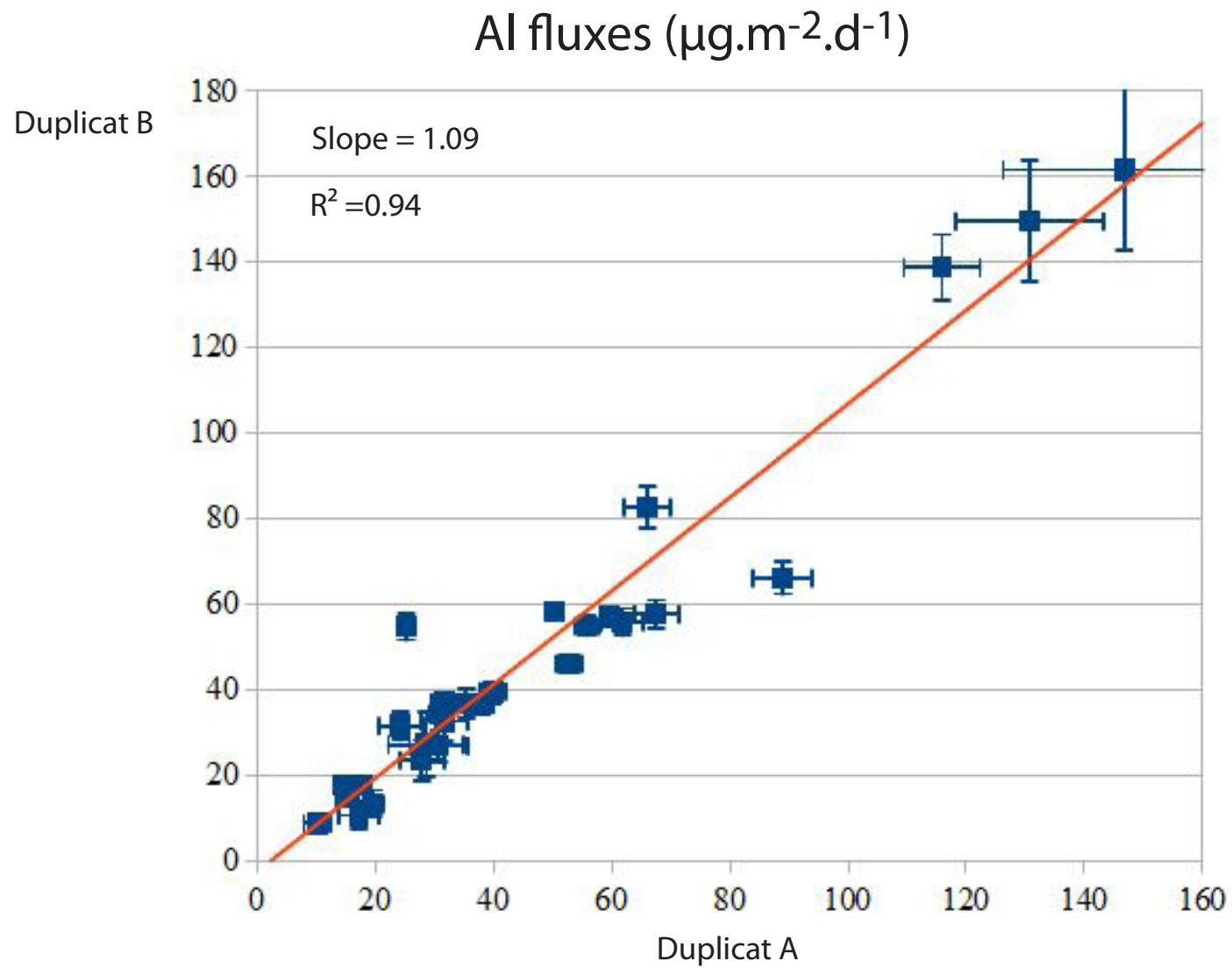
Accuracy on very low trace metal concentrations

Experimental validation

Quantities in field blanks $< 10\%$ of the ones in samples



Experimental validation



Lead isotopes: 206, 207, 208

HR-ICP-MS



hydrogen 1 H 1.0079																	helium 2 He 4.0026						
lithium 3 Li 6.941	beryllium 4 Be 9.0122																	boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180
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Aerosol sampling

A photograph of aerosol sampling equipment mounted on a grey metal mast against a blue sky with scattered white clouds. The equipment consists of several grey cylindrical components. One component on the left has a clear lens-like cover over a green tube. Two other components on the right have clear covers revealing white circular filters. Red and yellow safety ropes are tied around the mast and equipment.

At «Jacky» only (Kerguelen)

Filtration on Téflon[®] filters
2 m from ground level

Same sampling time than
atmospheric deposition

Analyzed using X ray
fluorescence spectrometry
(Al, Na)

Rainwater sampling (event basis)



On Kerguelen base «PAF»

Direct filtration on PC filters:
separation of the both
soluble and insoluble
fractions during the sampling

16 collected rains

Digestion using H_2O_2 and
 HF/HNO_3 (insoluble)

Analyses using HR-ICP-MS
(soluble and insoluble)

7 analyzed and validated elements in 5 rains

HR-ICP-MS



hydrogen 1 H 1.0079																helium 2 He 4.0026						
lithium 3 Li 6.941	beryllium 4 Be 9.0122																boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180
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A world map with a light blue background and a white grid. The map shows the continents in a light beige color. Two small red dots are located in the southern Indian Ocean, south of Africa and east of Madagascar. The word "Results" is written in a large, black, sans-serif font in the center of the map.

Results

Scientific context

Issues

Methods

Results

1. What is the atmospheric deposition flux of dust derived micronutrients that reaches the remote Southern Indian Ocean?
2. What is its chemical composition?
3. What are its different sources?
4. What is its bioavailable part for phytoplankton?

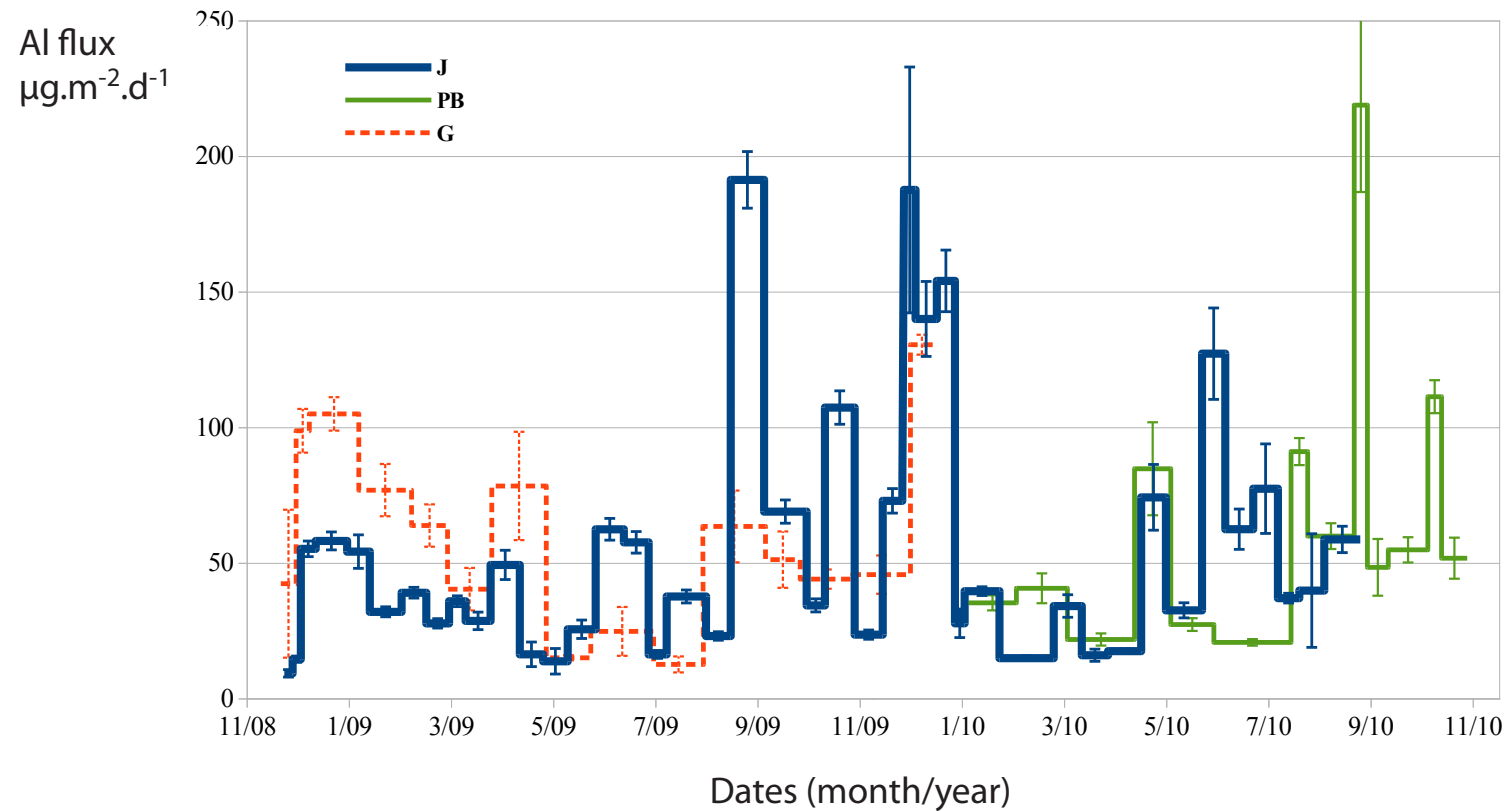


Total dust deposition flux

$$F_{\text{dust}} = \frac{F_{\text{Al}}}{\text{Abundance}_{\text{Al}}}$$



Total Al deposition flux (dust proxy)



$654 \pm 70 \mu\text{gDust}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$



Total dust deposition flux in literature (1)

Observations

$38 \pm 14 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Wagener et al., 2008)

$654 \pm 70 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Heimburger et al., 2012)



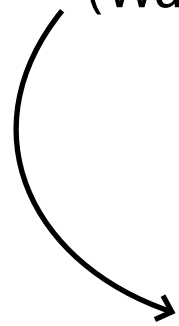
Total dust deposition flux in literature (1)

Observations

$38 \pm 14 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Wagener et al., 2008)

$654 \pm 70 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Heimbürger et al., 2012)

$F_{\text{total}} = \text{direct measurements}$



Indirect measurements of total deposition calculation:

$$F_{\text{total}} = F_{\text{dry}} + F_{\text{wet}}$$

$$F_{\text{dry}} = C_{\text{aerosol}} \cdot V_{\text{dry deposition}}$$

$$F_{\text{wet}} = C_{\text{aerosol}} \cdot \text{SR} \cdot \text{rainfall}$$



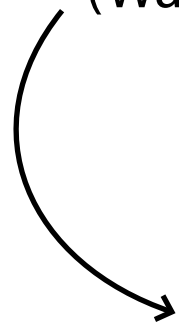
Total dust deposition flux in literature (1)

Observations

$38 \pm 14 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Wagener et al., 2008)

$654 \pm 70 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
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Indirect measurements of total deposition calculation:

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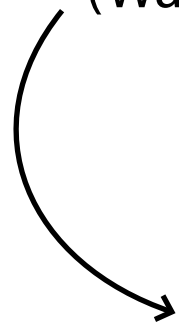
Total dust deposition flux in literature (1)

Observations

$38 \pm 14 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
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$654 \pm 70 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Heimbürger et al., 2012)

$F_{\text{total}} = \text{direct measurements}$



Indirect measurements of total deposition calculation:

$$F_{\text{total}} = F_{\text{dry}} + F_{\text{wet}}$$

$$F_{\text{dry}} = C_{\text{aerosol}} \cdot V_{\text{dry deposition}}$$

$$F_{\text{wet}} = C_{\text{aerosol}} \cdot \text{SR} \cdot \text{rainfall}$$

SR = 200 (constant)
(Wagener et al., 2008)

SR = measured
(Heimbürger et al., 2012)



Scavenging ratio

$$SR = \frac{C_{\text{rain}}}{C_{\text{air}}}$$



Scavenging ratio

$$SR = \frac{C_{\text{rain}}}{C_{\text{air}}}$$

aerosol sampling





Scavenging ratio

$$SR = \frac{C_{\text{rain}}}{C_{\text{air}}}$$

concentration in
total deposition
wet deposition > 90%

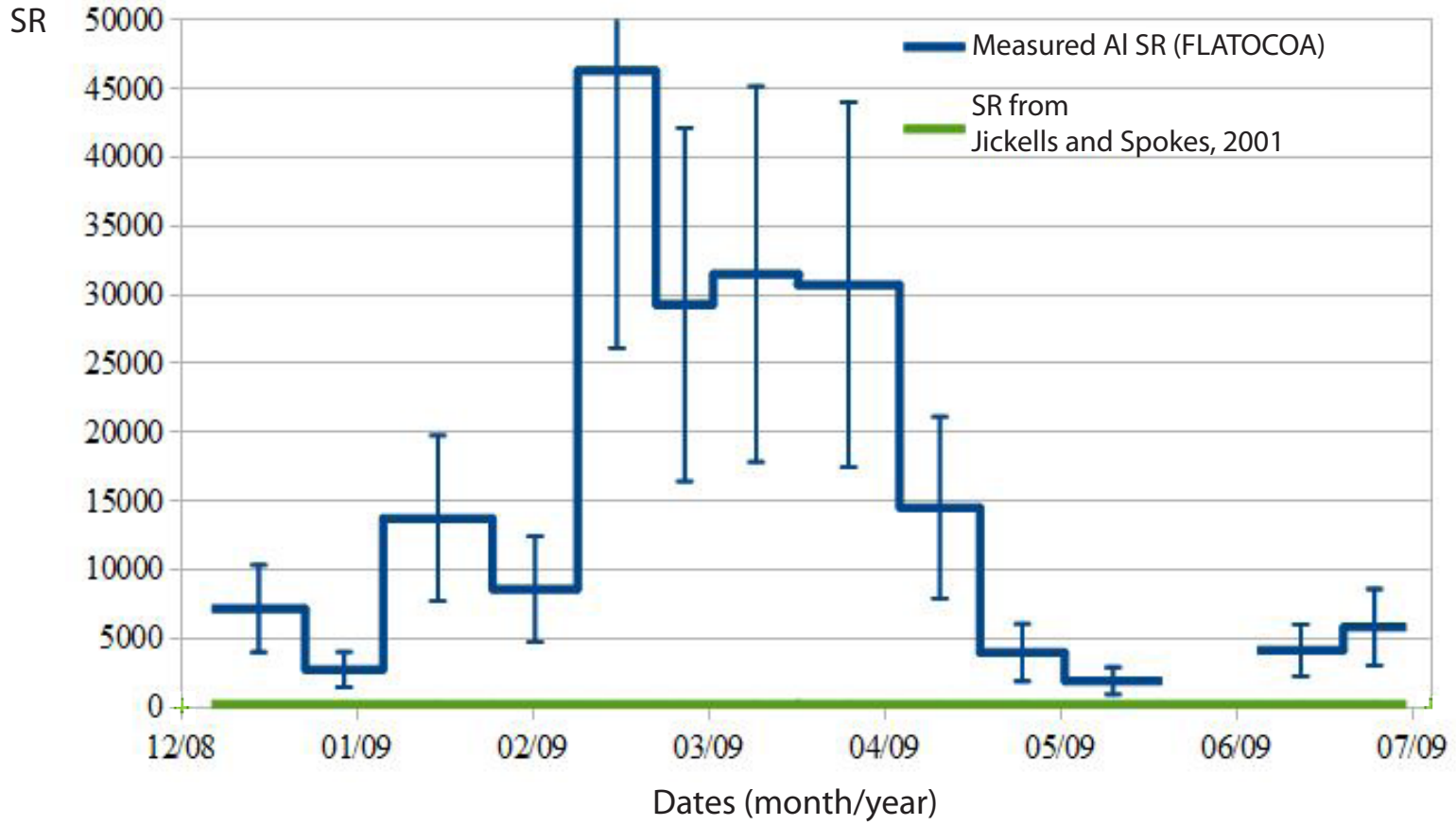


aerosol sampling



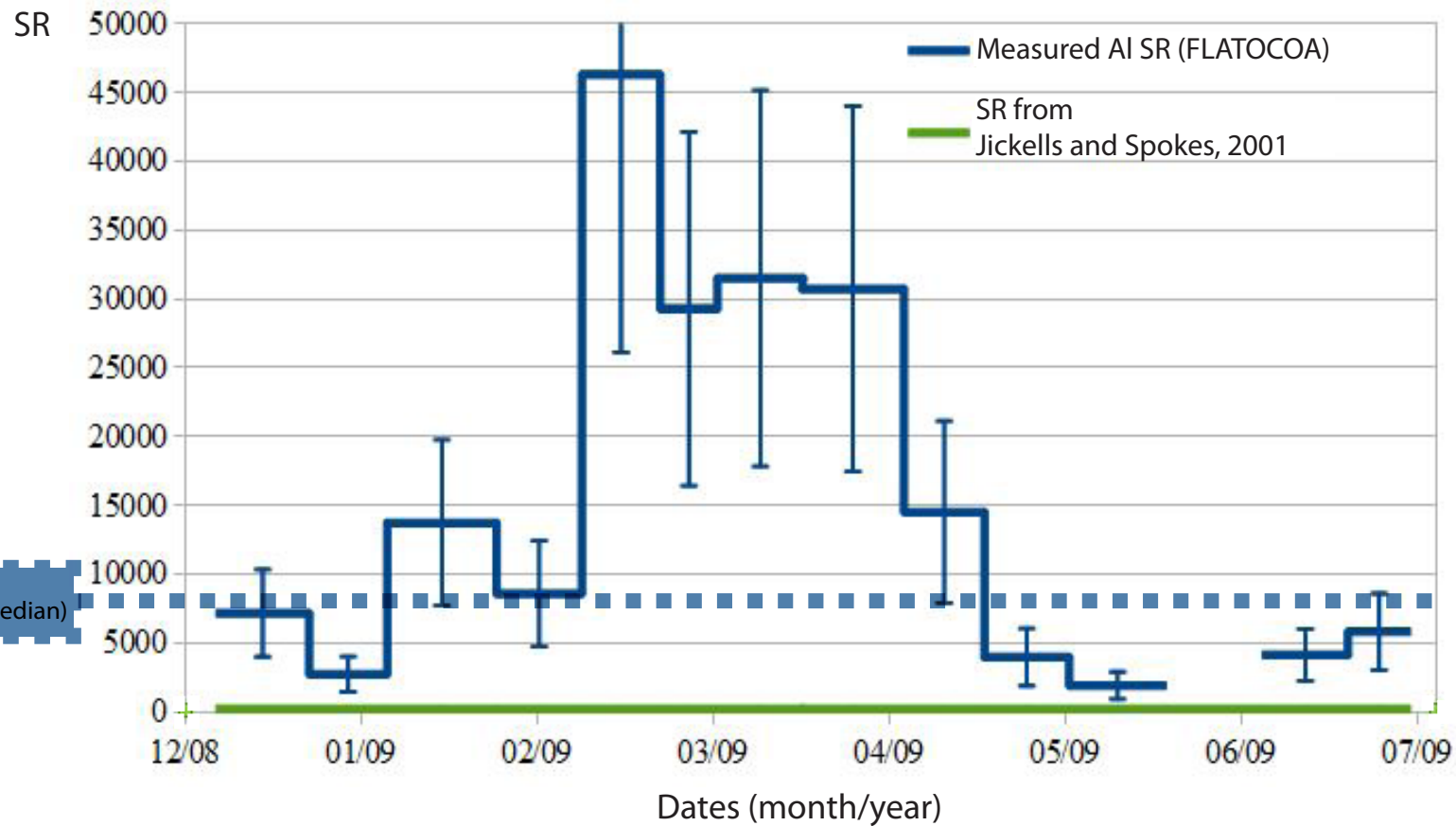


Scavenging ratio



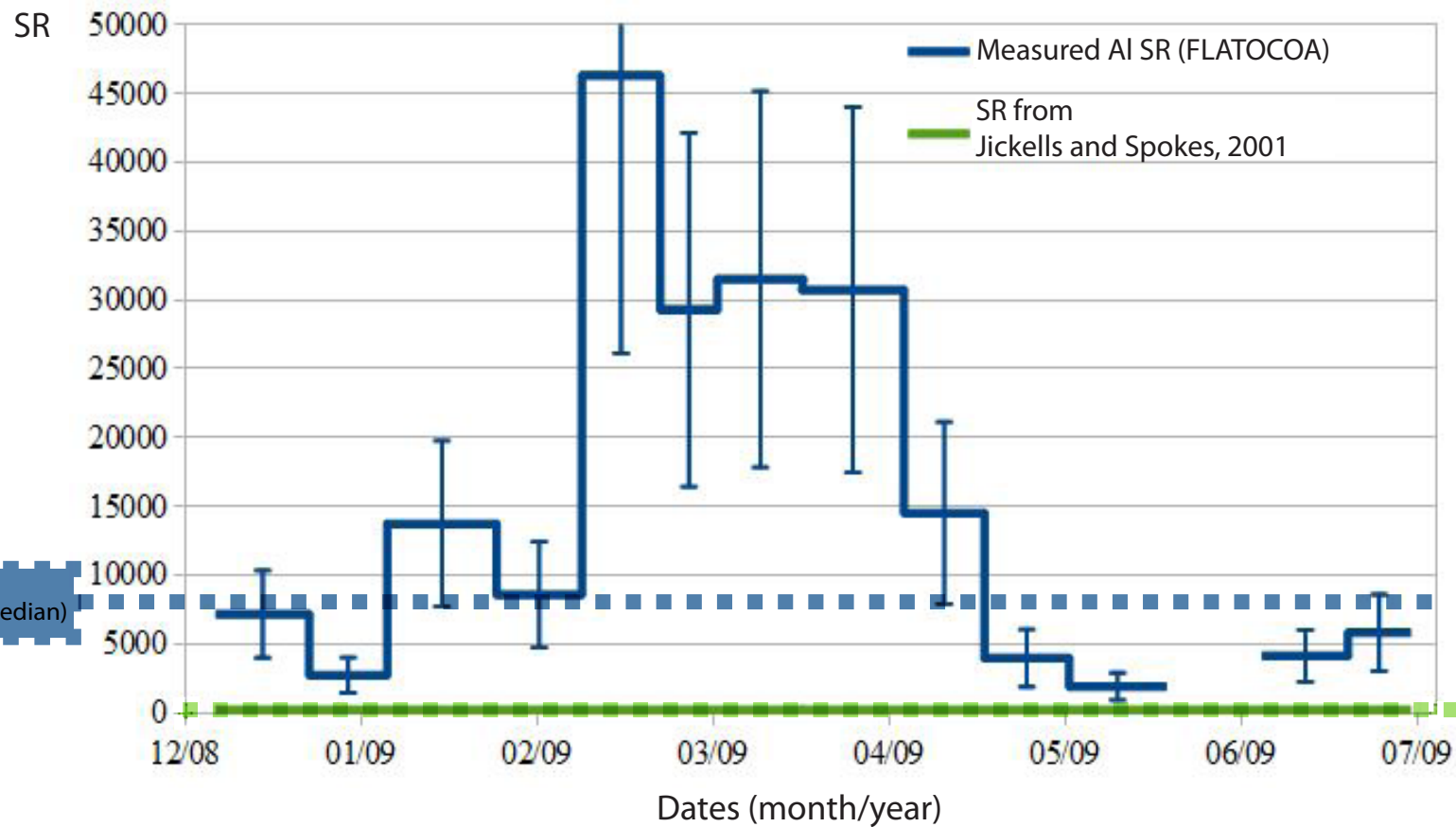


Scavenging ratio





Scavenging ratio

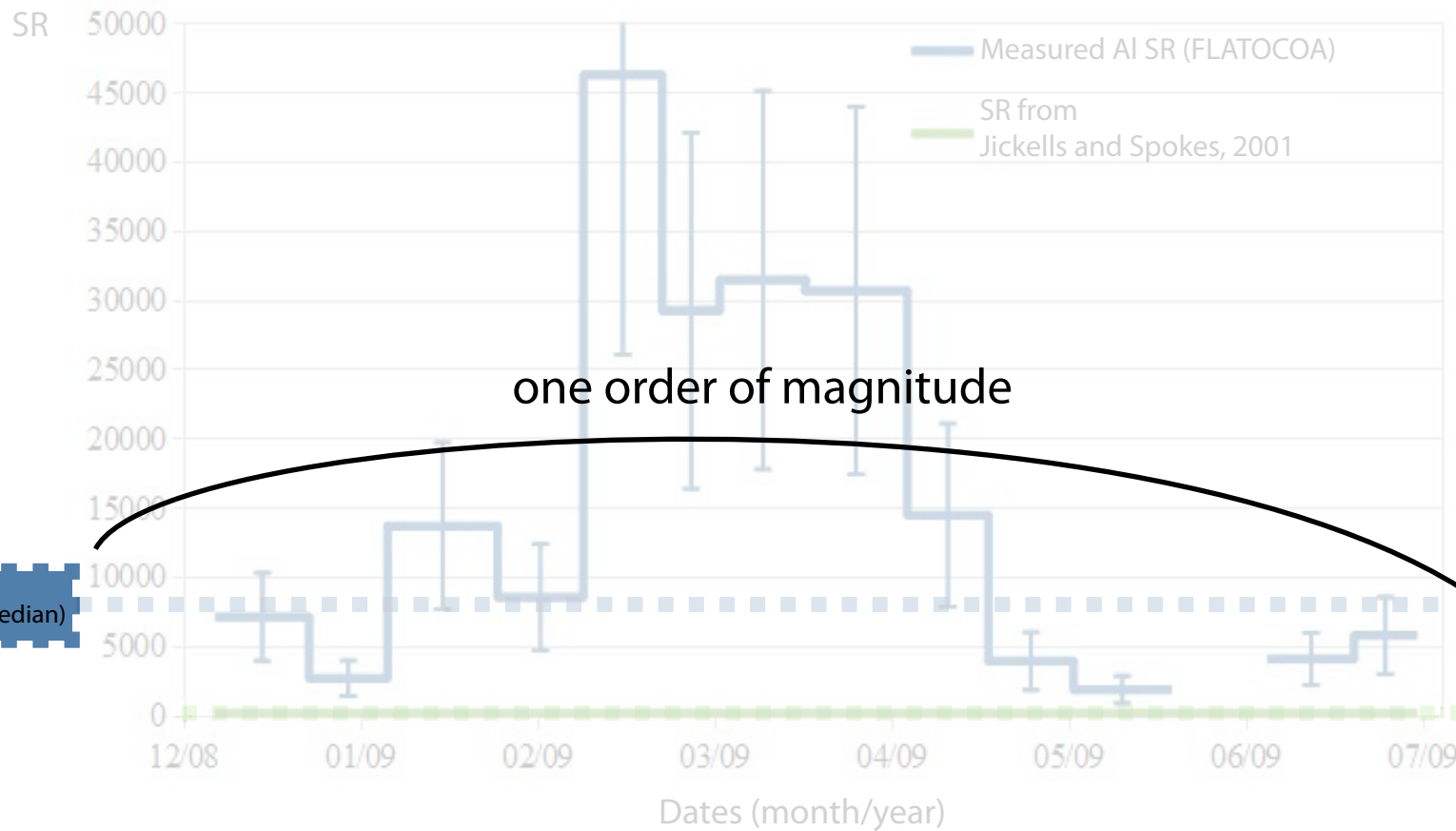


7000 (median)

200



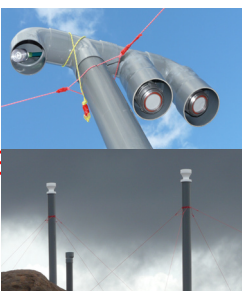
Scavenging ratio



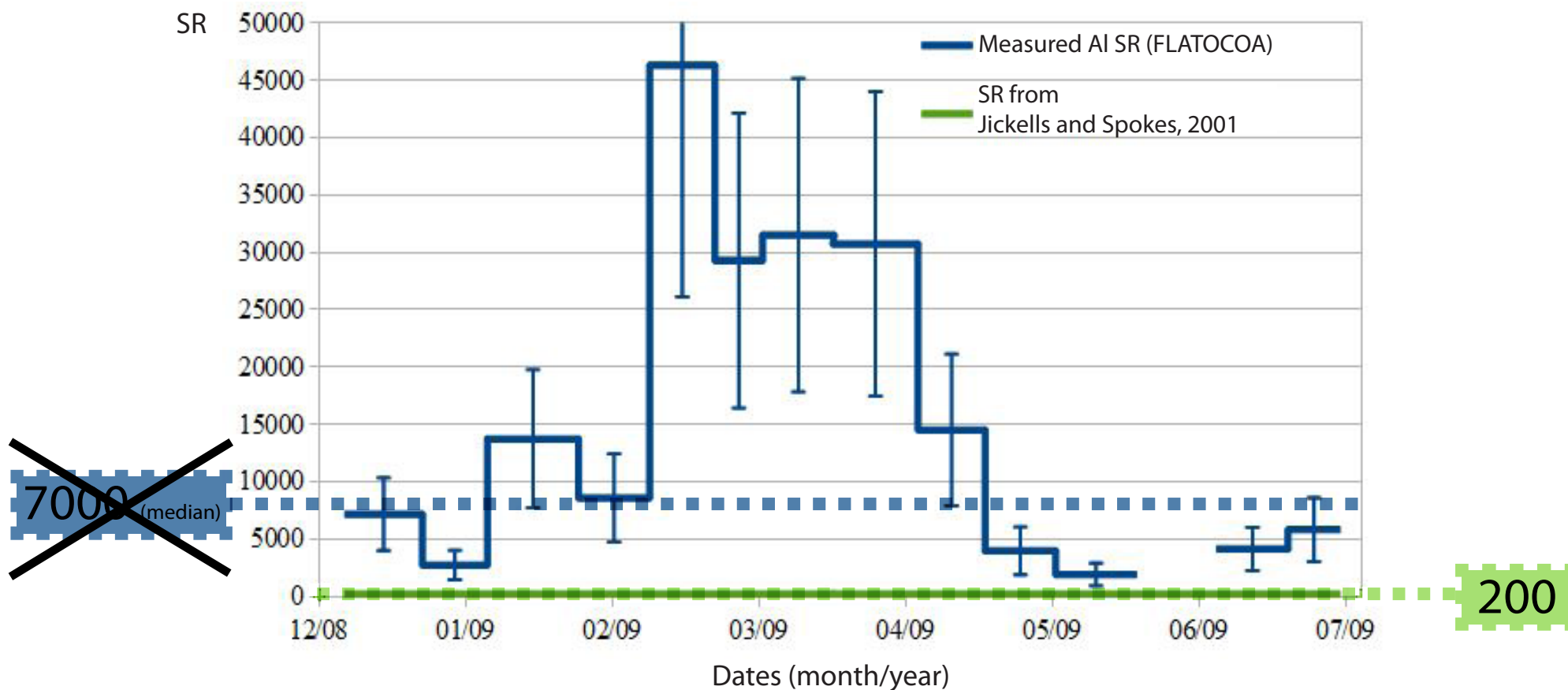
$654 \pm 70 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Heimburger et al., 2012)

$38 \pm 14 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(Wagener et al., 2008)

one order of magnitude



Scavenging ratio



Hypothesis:

aerosols scavenged by rain are in altitude and not at surface level



Total dust deposition flux in literature (1)

Observations

Dust deposition is one order of magnitude higher than the one previously observed: atmospheric deposition is more important than expected for micronutrient supply in the South Indian Ocean

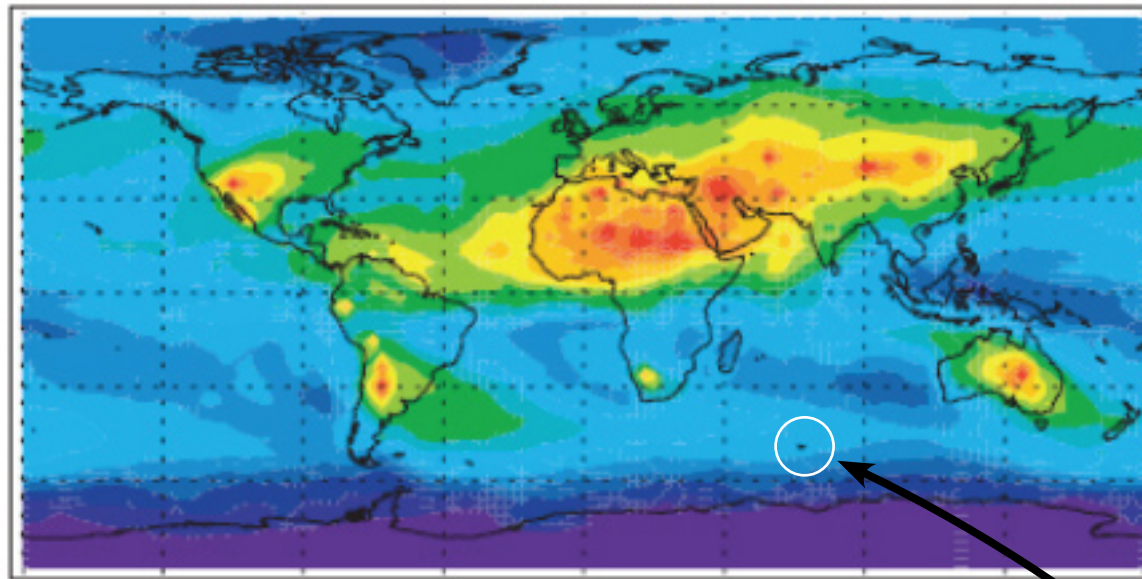
Measurements of C_{aerosols} at surface level only do not allow atmospheric deposition flux to be well calculated over remote oceanic area



Total dust deposition in literature (2)

Models

Simulated averaged dust
deposition flux ($\text{g.m}^{-2}.\text{yr}^{-1}$)

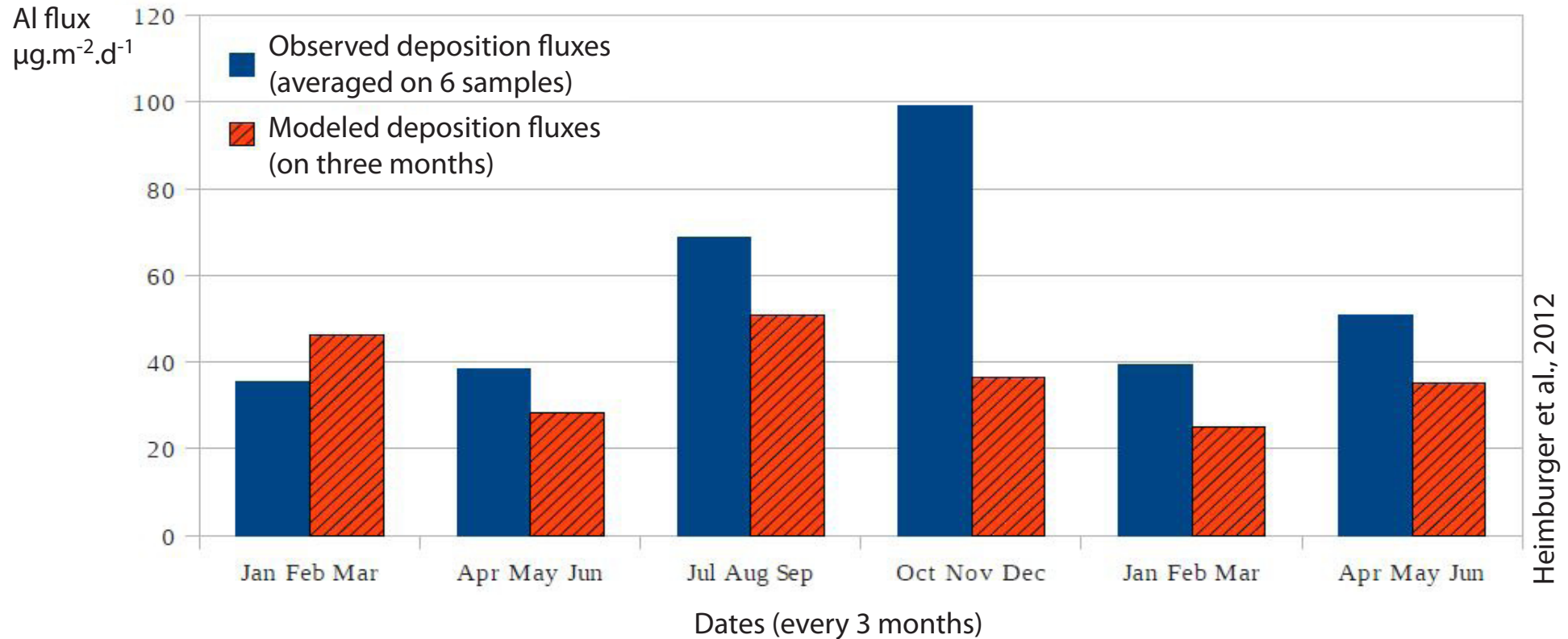


Estimated dust deposition flux:
between 550 and $1400 \mu\text{g.m}^{-2}.\text{d}^{-1}$
(Mahowald et al., 2007)

$654 \pm 70 \mu\text{g.m}^{-2}.\text{d}^{-1}$
(Heimburger et al., 2012)



Total dust deposition vs. models



Case study:
the model does not reflect observed seasonality

Issues

1. What is the atmospheric deposition flux of dust derived micronutrients that reaches the remote Southern Indian Ocean?
2. What is its chemical composition?
3. What are its different sources?
4. What is its bioavailable part for phytoplankton?



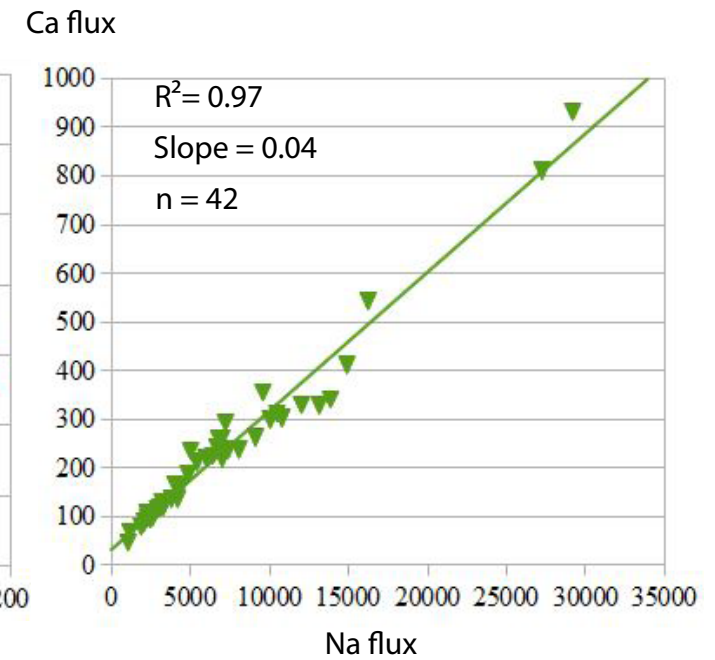
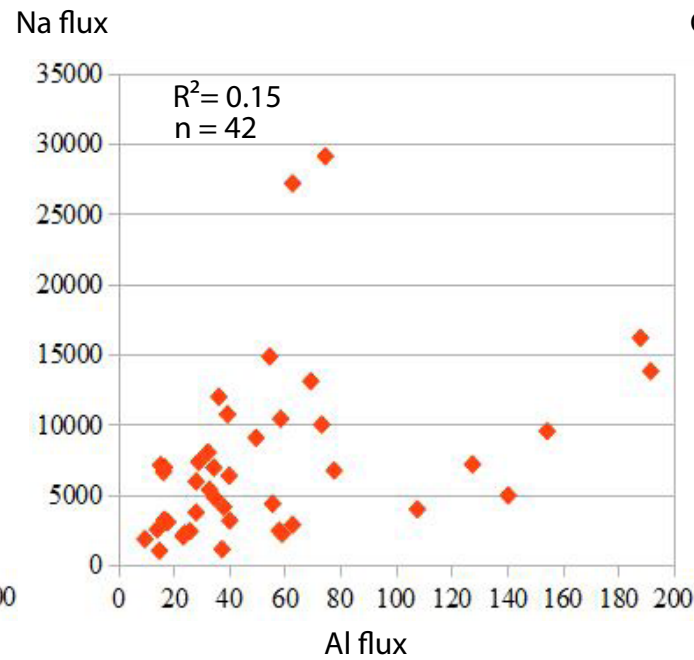
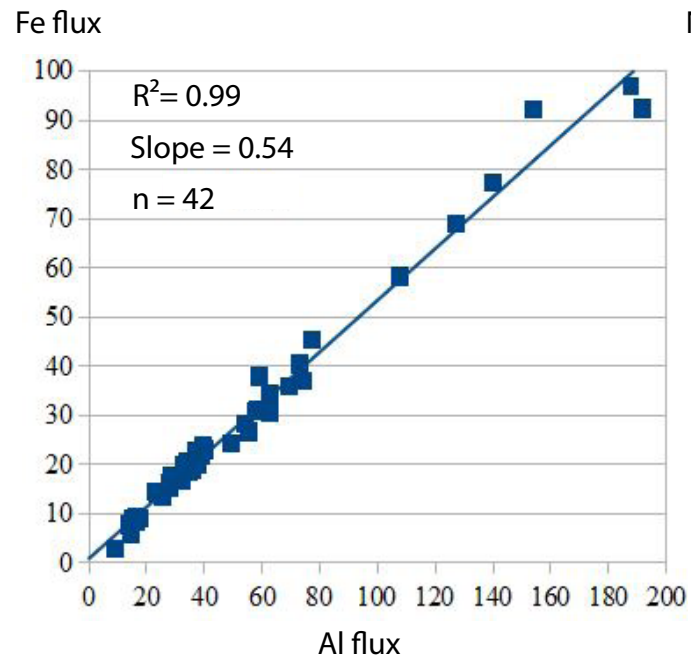
Total deposition fluxes

hydrogen 1 H 1.0079																		helium 2 He 4.0026
lithium 3 Li 6.941	beryllium 4 Be 9.0122											boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180	
sodium 11 Na 22.990	magnesium 12 Mg 24.305											aluminium 13 Al 26.982	silicon 14 Si 28.086	phosphorus 15 P 30.974	sulfur 16 S 32.065	chlorine 17 Cl 35.453	argon 18 Ar 39.948	
potassium 19 K 39.098	calcium 20 Ca 40.078	scandium 21 Sc 44.956	titanium 22 Ti 47.867	vanadium 23 V 50.942	chromium 24 Cr 51.996	manganese 25 Mn 54.938	iron 26 Fe 55.845	cobalt 27 Co 58.933	nickel 28 Ni 58.693	copper 29 Cu 63.546	zinc 30 Zn 65.39	gallium 31 Ga 69.723	germanium 32 Ge 72.61	arsenic 33 As 74.922	selenium 34 Se 78.96	bromine 35 Br 79.904	krypton 36 Kr 83.80	
rubidium 37 Rb 85.468	strontium 38 Sr 87.62	yttrium 39 Y 88.906	zirconium 40 Zr 91.224	niobium 41 Nb 92.906	molybdenum 42 Mo 95.94	technetium 43 Tc [98]	ruthenium 44 Ru 101.07	rhodium 45 Rh 102.91	palladium 46 Pd 106.42	silver 47 Ag 107.87	cadmium 48 Cd 112.41	indium 49 In 114.82	tin 50 Sn 118.71	antimony 51 Sb 121.76	tellurium 52 Te 127.60	iodine 53 I 126.90	xenon 54 Xe 131.29	
caesium 55 Cs 132.91	barium 56 Ba 137.33	57-70 *	lutetium 71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	platinum 78 Pt 195.08	gold 79 Au 196.97	mercury 80 Hg 200.59	thallium 81 Tl 204.38	lead 82 Pb 207.2	bismuth 83 Bi 208.98	polonium 84 Po [209]	astatine 85 At [210]	radon 86 Rn [222]
francium 87 Fr [223]	radium 88 Ra [226]	89-102 * *	lawrencium 103 Lr [262]	rutherfordium 104 Rf [261]	dubnium 105 Db [262]	seaborgium 106 Sg [266]	bohrium 107 Bh [264]	hassium 108 Hs [269]	meitnerium 109 Mt [268]	ununnitium 110 Uun [271]	ununium 111 Uuu [272]	ununbium 112 Uub [277]		ununquadium 114 Uuq [289]				

lanthanum 57 La 138.91	cerium 58 Ce 140.12	praseodymium 59 Pr 140.91	neodymium 60 Nd 144.24	promethium 61 Pm [145]	samarium 62 Sm 150.36	europium 63 Eu 151.96	gadolinium 64 Gd 157.25	terbium 65 Tb 158.93	dysprosium 66 Dy 162.50	holmium 67 Ho 164.93	erbium 68 Er 167.26	thulium 69 Tm 168.93	ytterbium 70 Yb 173.04
actinium 89 Ac [227]	thorium 90 Th 232.04	protactinium 91 Pa 231.04	uranium 92 U 238.03	neptunium 93 Np [237]	plutonium 94 Pu [244]	americium 95 Am [243]	curium 96 Cm [247]	berkelium 97 Bk [247]	californium 98 Cf [251]	einsteinium 99 Es [252]	fermium 100 Fm [257]	mendelevium 101 Md [258]	nobelium 102 No [259]

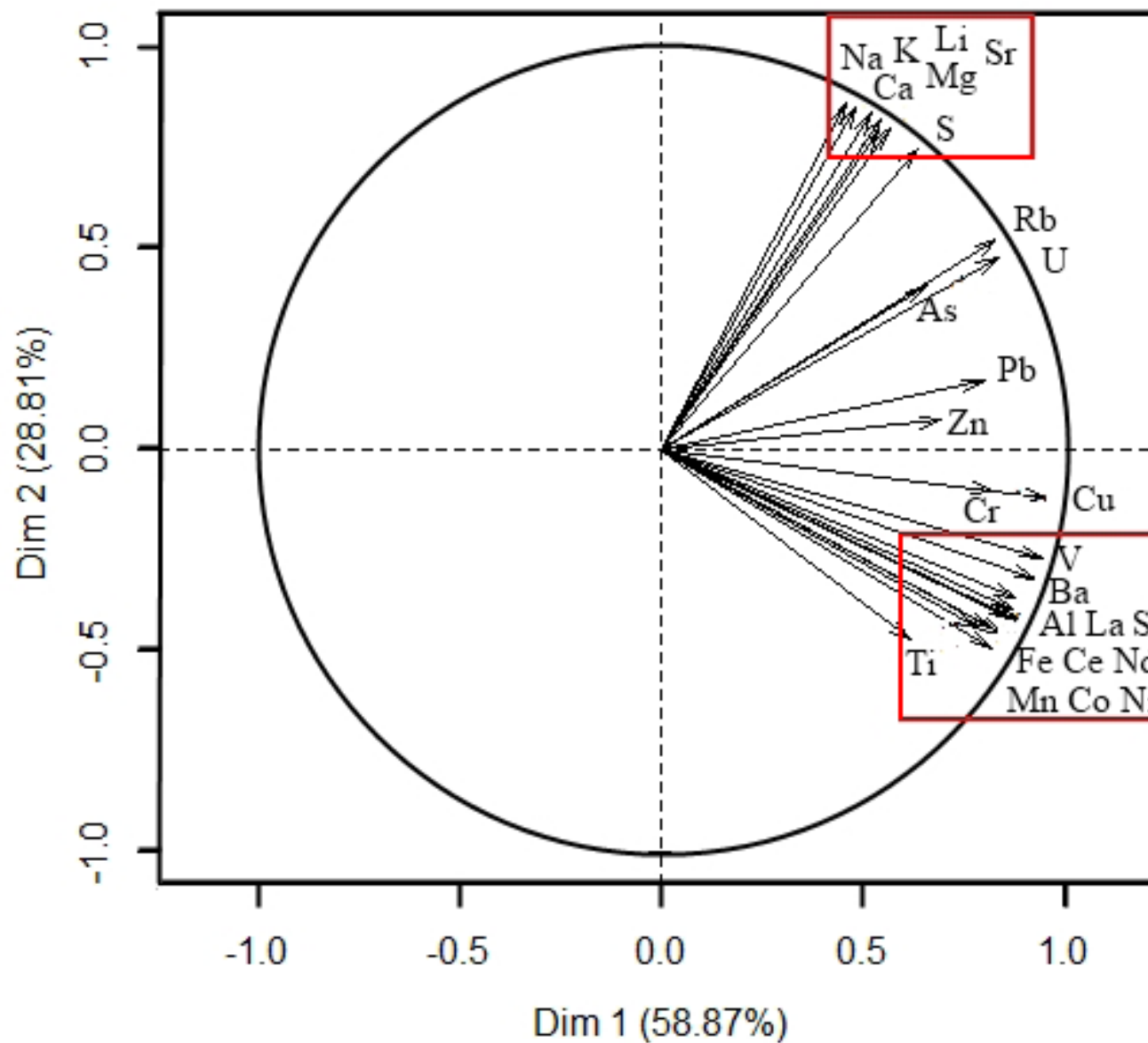


Correlation coefficients (Jacky)





Principal Component Analysis





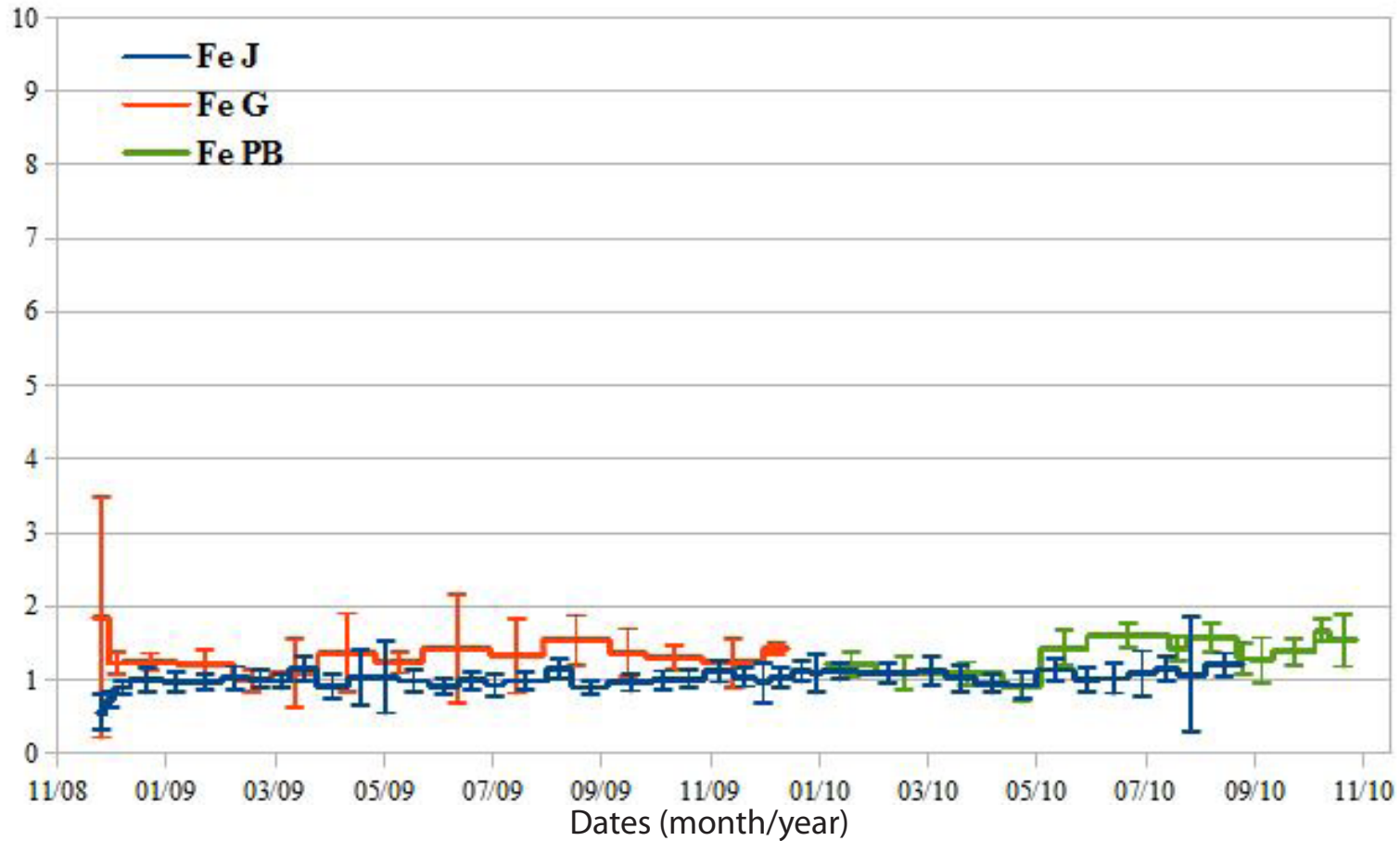
Enrichment factors

$$EF = \frac{\frac{[X]}{[AI]}}{\frac{[X]_{ref}}{[AI]_{ref}}}$$



Enrichment factors

Enrichment factors

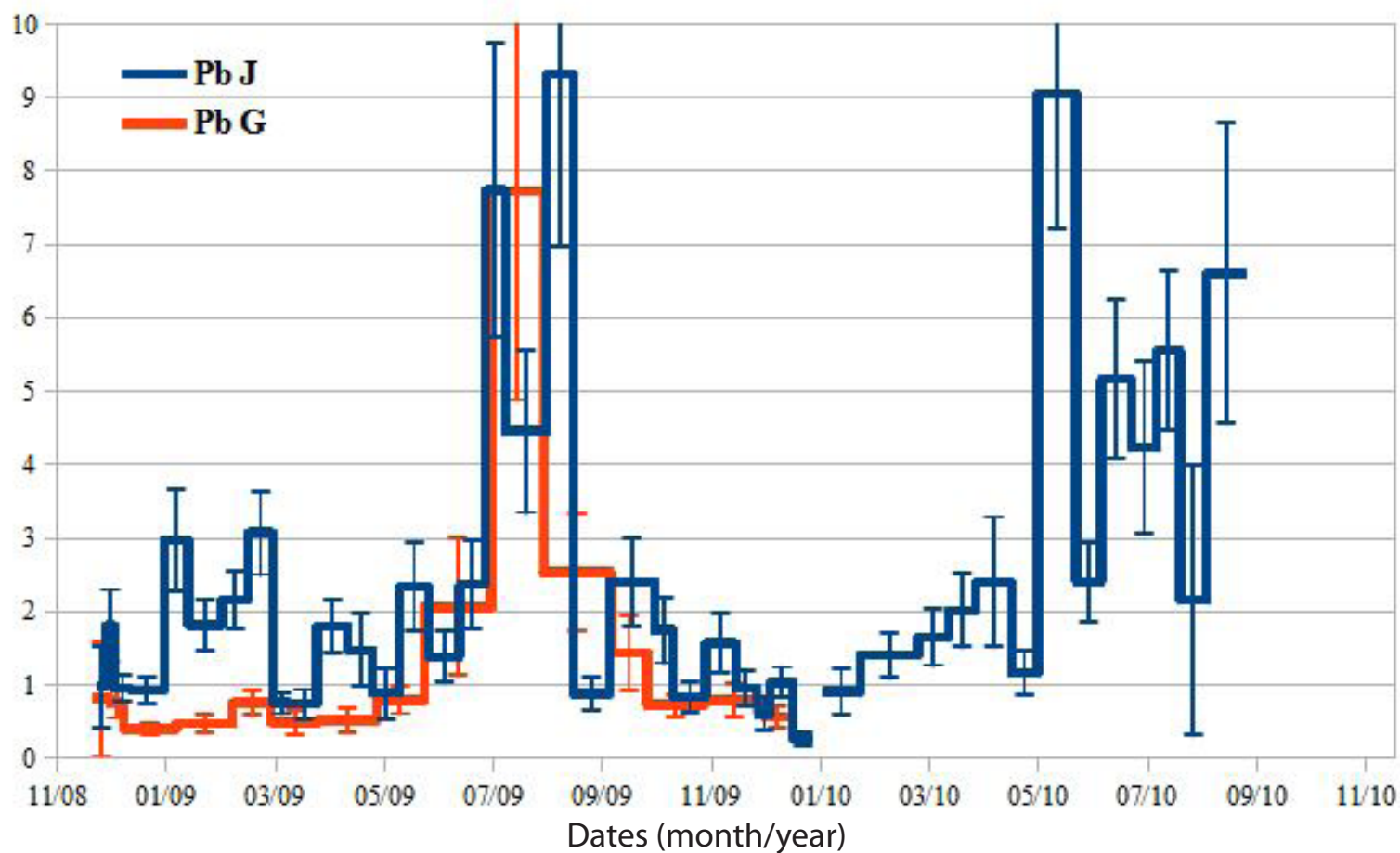


Constant EF for elements identified as coming from crustal origin...

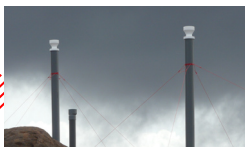


Enrichment factors

Enrichment factors

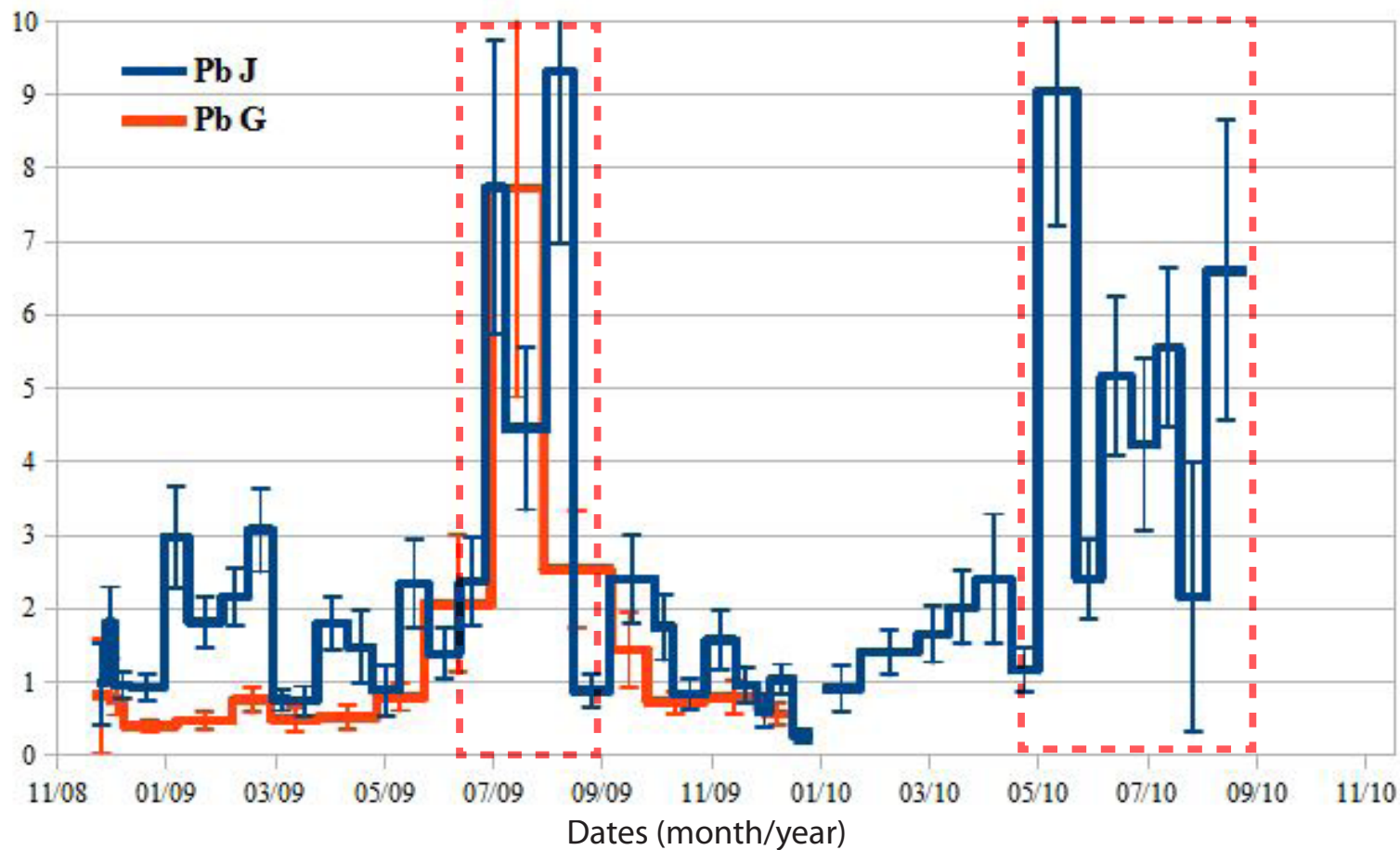


... in comparison with others elements
such as Pb, As, Cr, Cu and V...



Enrichment factors

Enrichment factors

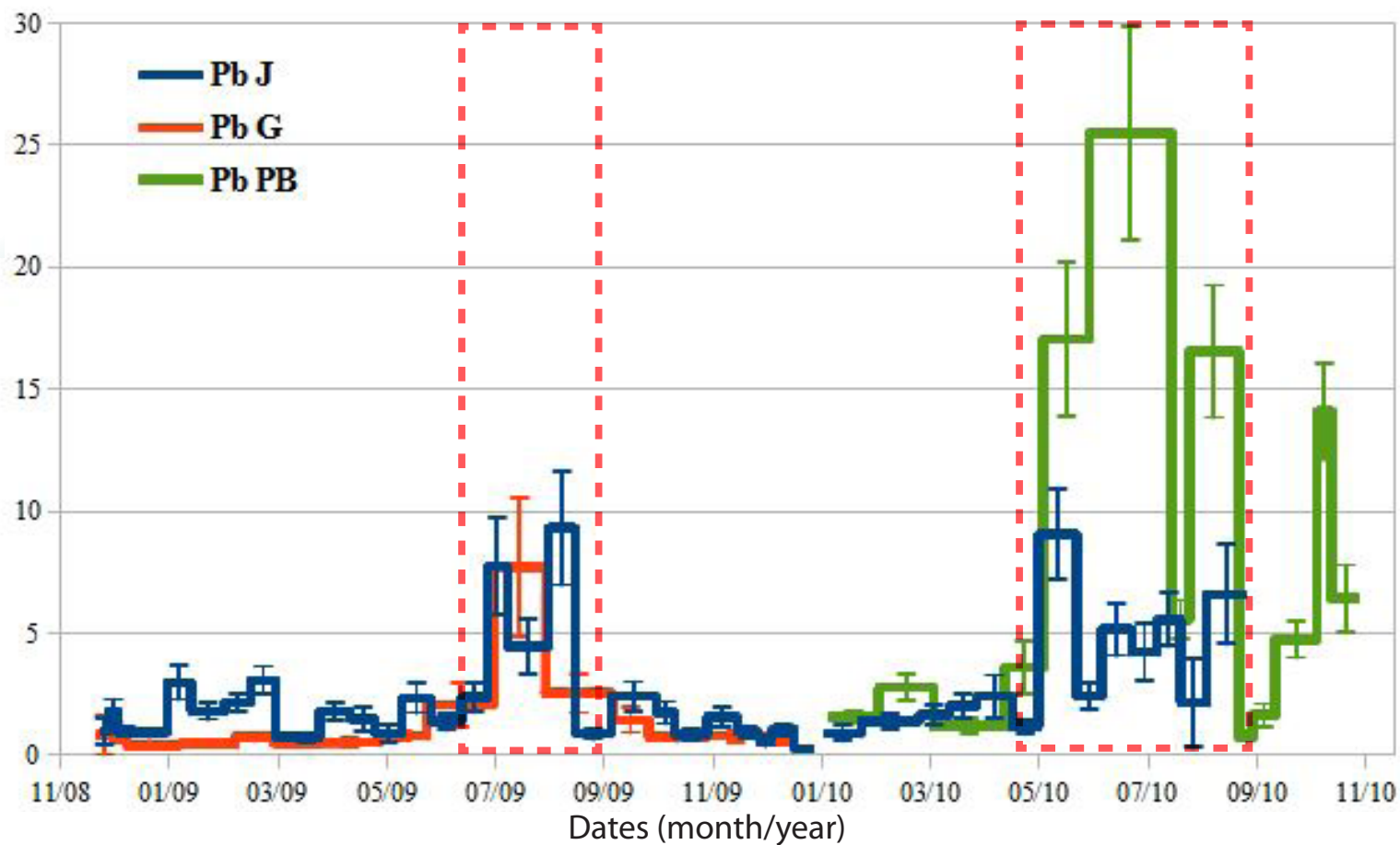


... for which an anthropogenic contribution in deposition is observed during the austral winter.



Enrichment factors

Enrichment factors



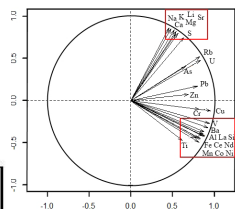
Gradient from Crozet to Kerguelen Islands
due to transport and/or emission?



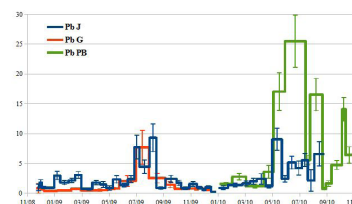
Total deposition fluxes

PCA, correlation coefficients and enrichment factors

hydrogen 1 H 1.0079	beryllium 4 Be 9.0122
lithium 3 Li 6.941	magnesium 12 Mg 24.305
sodium 11 Na 22.990	calcium 20 Ca 40.078
potassium 19 K 39.098	strontium 38 Sr 87.62
rubidium 37 Rb 85.468	barium 56 Ba 137.33
caesium 55 Cs 132.91	*
francium 87 Fr [223]	89-102 * *



Al	As	Ba	Ca	Co	Cr	Cs	Fe	Fr	La	Li	Mg	Mn	Ni	Pb	Pb	Si	Ti	U	V	Zn
1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00



boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	helium 2 He 4.0026
aluminum 13 Al 26.982	silicon 14 Si 28.086	phosphorus 15 P 30.974	sulfur 16 S 32.065	chlorine 17 Cl 35.453	neon 10 Ne 20.180
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thallium 81 Tl 204.38	lead 82 Pb 207.2	bismuth 83 Bi 208.98	polonium 84 Po [209]	astatine 85 At [210]	radon 86 Rn [222]
	ununquadium 114 Uuq [289]				

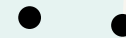
scandium 21 Sc 44.956	titanium 22 Ti 47.867	vanadium 23 V 50.942	chromium 24 Cr 51.996	manganese 25 Mn 54.938	iron 26 Fe 55.845	cobalt 27 Co 58.933	nickel 28 Ni 58.693	copper 29 Cu 63.546	zinc 30 Zn 65.39
yttrium 39 Y 88.906	zirconium 40 Zr 91.224	niobium 41 Nb 92.906	molybdenum 42 Mo 95.94	technetium 43 Tc [98]	ruthenium 44 Ru 101.07	rhodium 45 Rh 102.91	palladium 46 Pd 106.42	silver 47 Ag 107.87	cadmium 48 Cd 112.41
	lutetium 71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	gold 79 Au 196.97	mercury 80 Hg 200.59
	lawrencium 103 Lr [262]	rutherfordium 104 Rf [261]	dubnium 105 Db [262]	seaborgium 106 Sg [266]	bohrium 107 Bh [264]	hassium 108 Hs [269]	meitnerium 109 Mt [268]	ununilium 110 Uun [271]	unununium 111 Uuu [272]

- Sea-salt
- Crustal
- Anthropogenic in winter

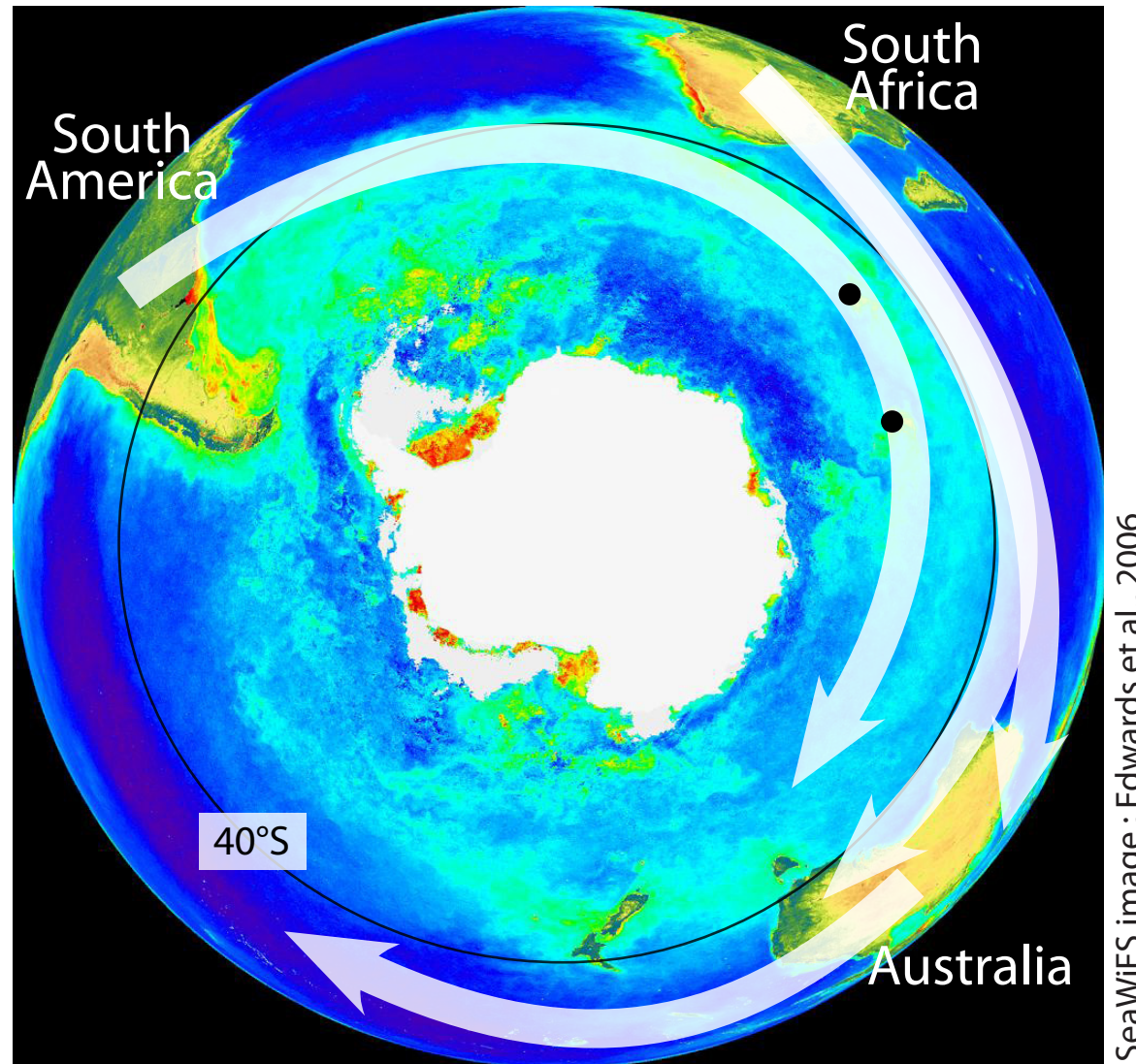
lanthanum 57 La 138.91	cerium 58 Ce 140.12	praseodymium 59 Pr 140.91	neodymium 60 Nd 144.24	promethium 61 Pm [145]	samarium 62 Sm 150.36	europium 63 Eu 151.96	gadolinium 64 Gd 157.25	terbium 65 Tb 158.93	dysprosium 66 Dy 162.50	holmium 67 Ho 164.93	erbium 68 Er 167.26	thulium 69 Tm 168.93	ytterbium 70 Yb 173.04
actinium 89 Ac [227]	thorium 90 Th 232.04	protactinium 91 Pa 231.04	uranium 92 U 238.03	neptunium 93 Np [237]	plutonium 94 Pu [244]	americium 95 Am [243]	curium 96 Cm [247]	berkelium 97 Bk [247]	californium 98 Cf [251]	einsteinium 99 Es [252]	fermium 100 Fm [257]	mendelevium 101 Md [258]	nobelium 102 No [259]

Issues

1. What is the atmospheric deposition flux of dust derived micronutrients that reaches the remote Southern Indian Ocean?
2. What is its chemical composition?
- 3. What are its different sources?**
4. What is its bioavailable part for phytoplankton?

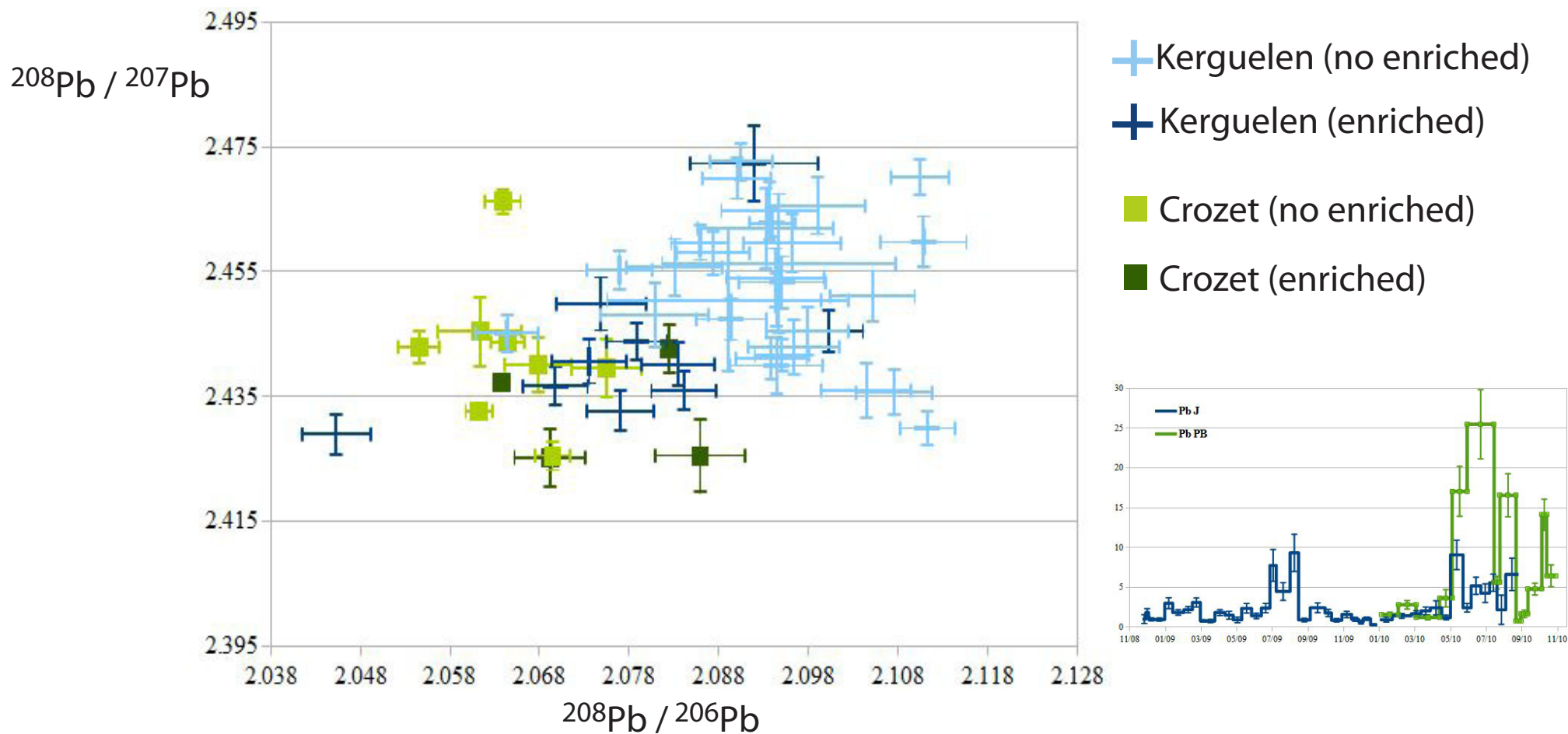


Dust sources in the South Hemisphere



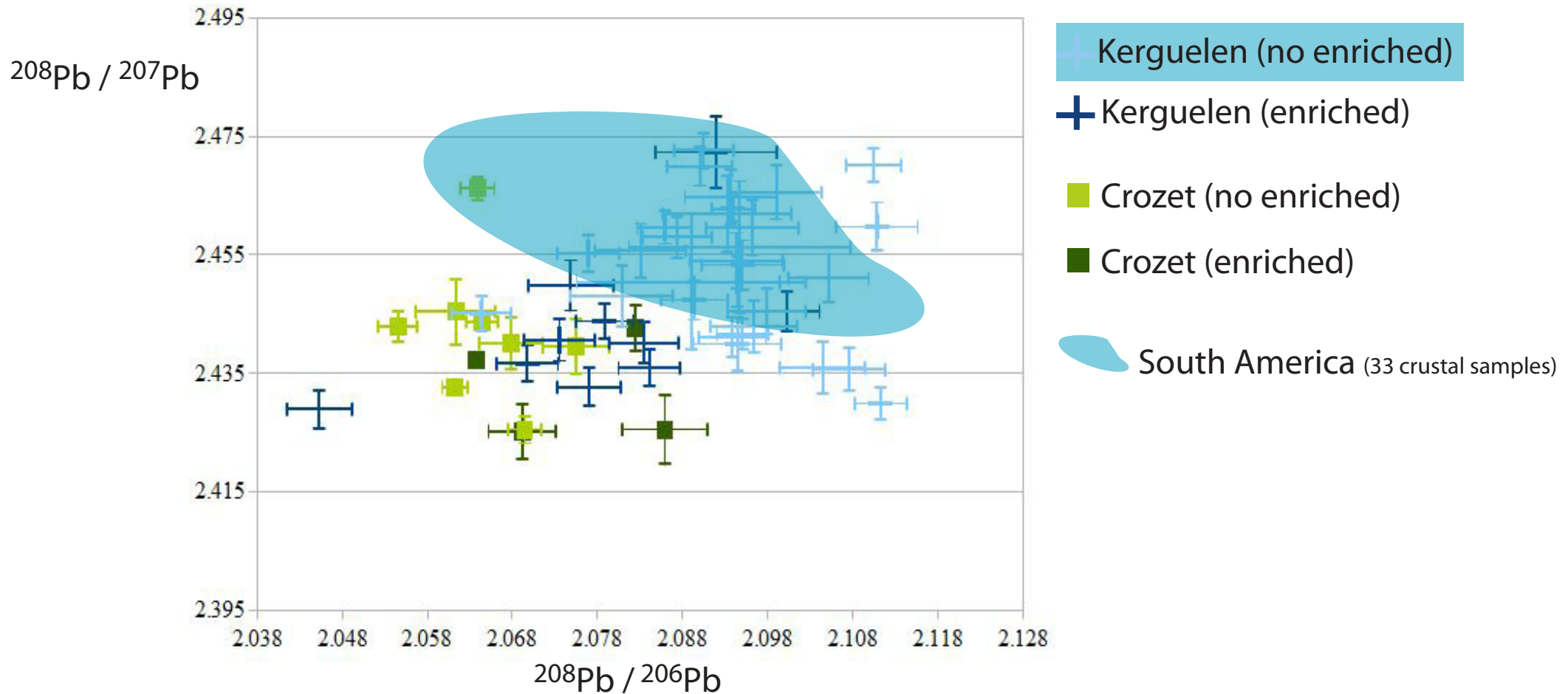


Pb isotopes





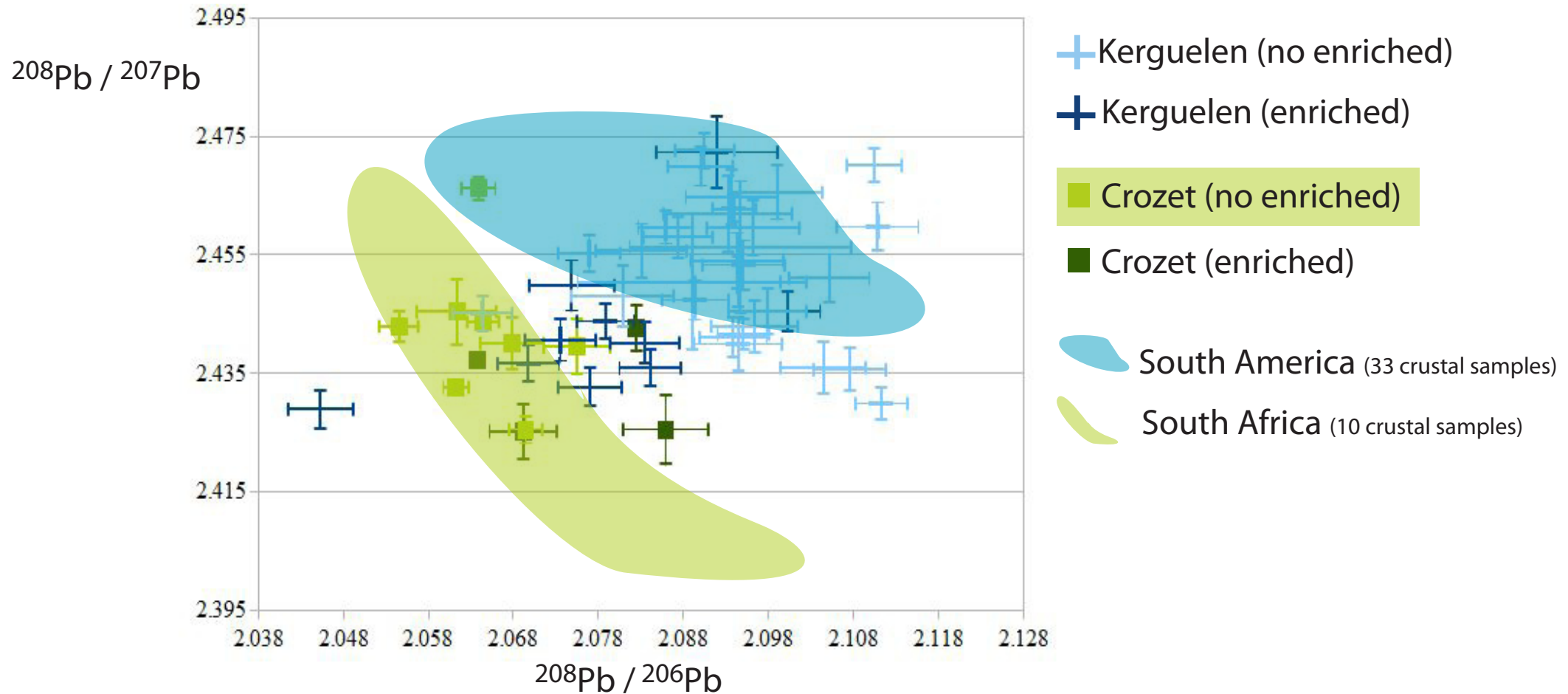
Pb isotopes



Correlation between Kerguelen (no enriched) and South America samples



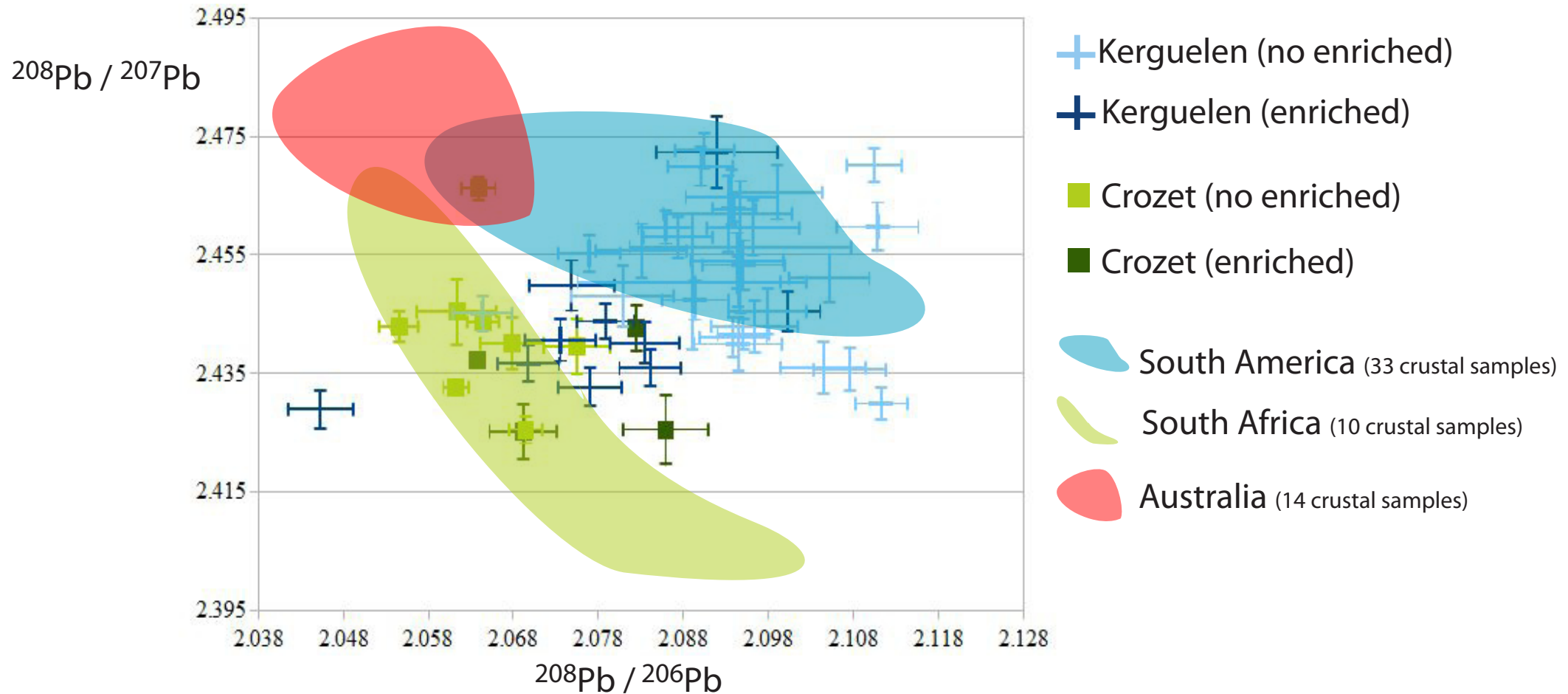
Pb isotopes



Correlation between Crozet (no enriched) and South Africa samples



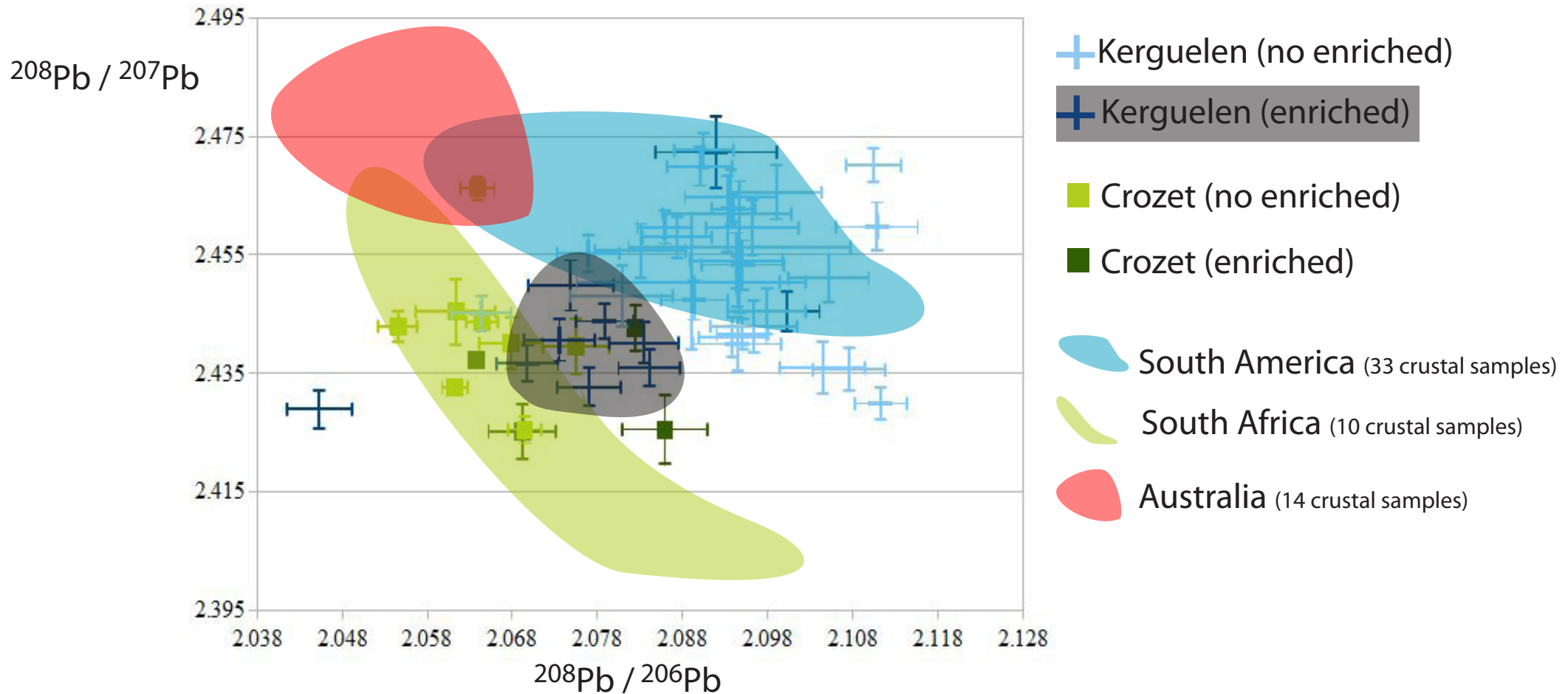
Pb isotopes



But no correlation with samples from Australia



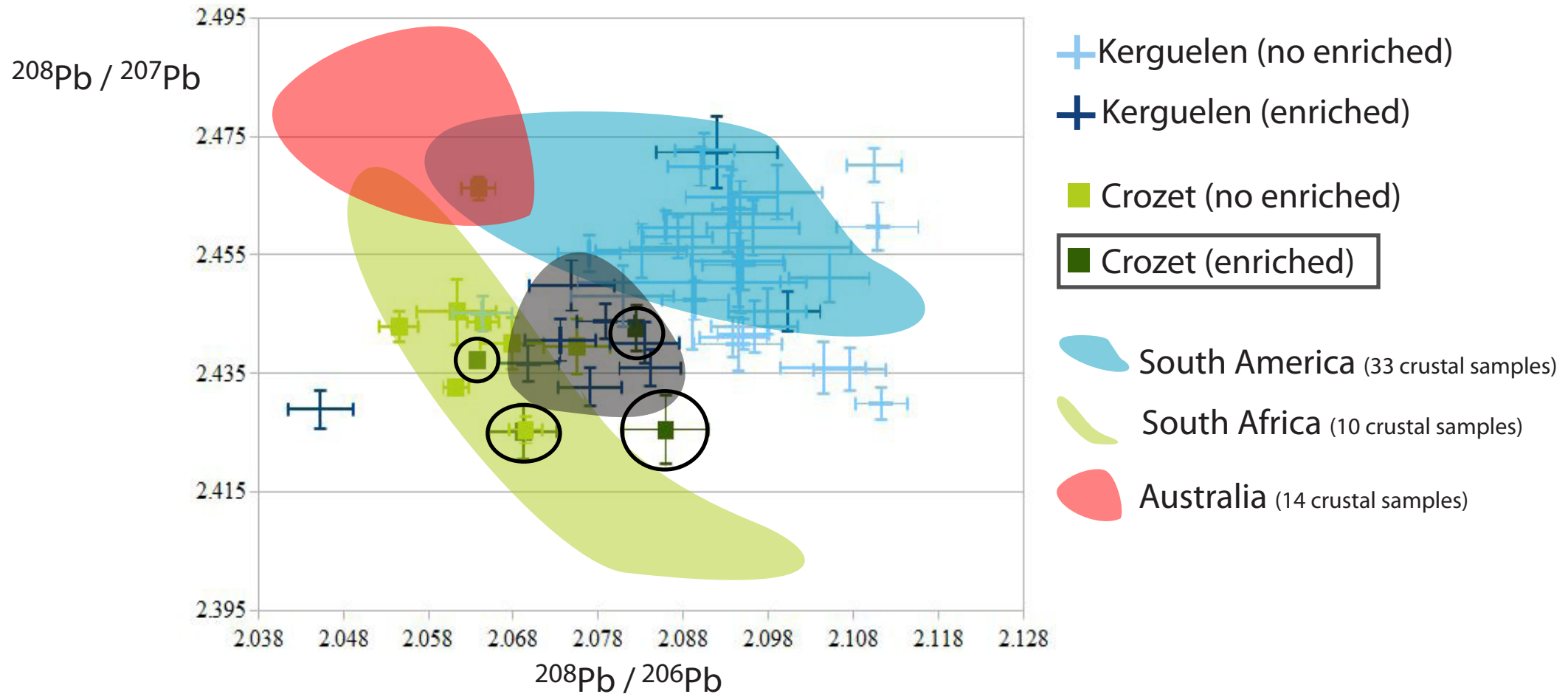
Pb isotopes



Kerguelen (enriched) influenced by both South America and South Africa during the austral winter



Pb isotopes



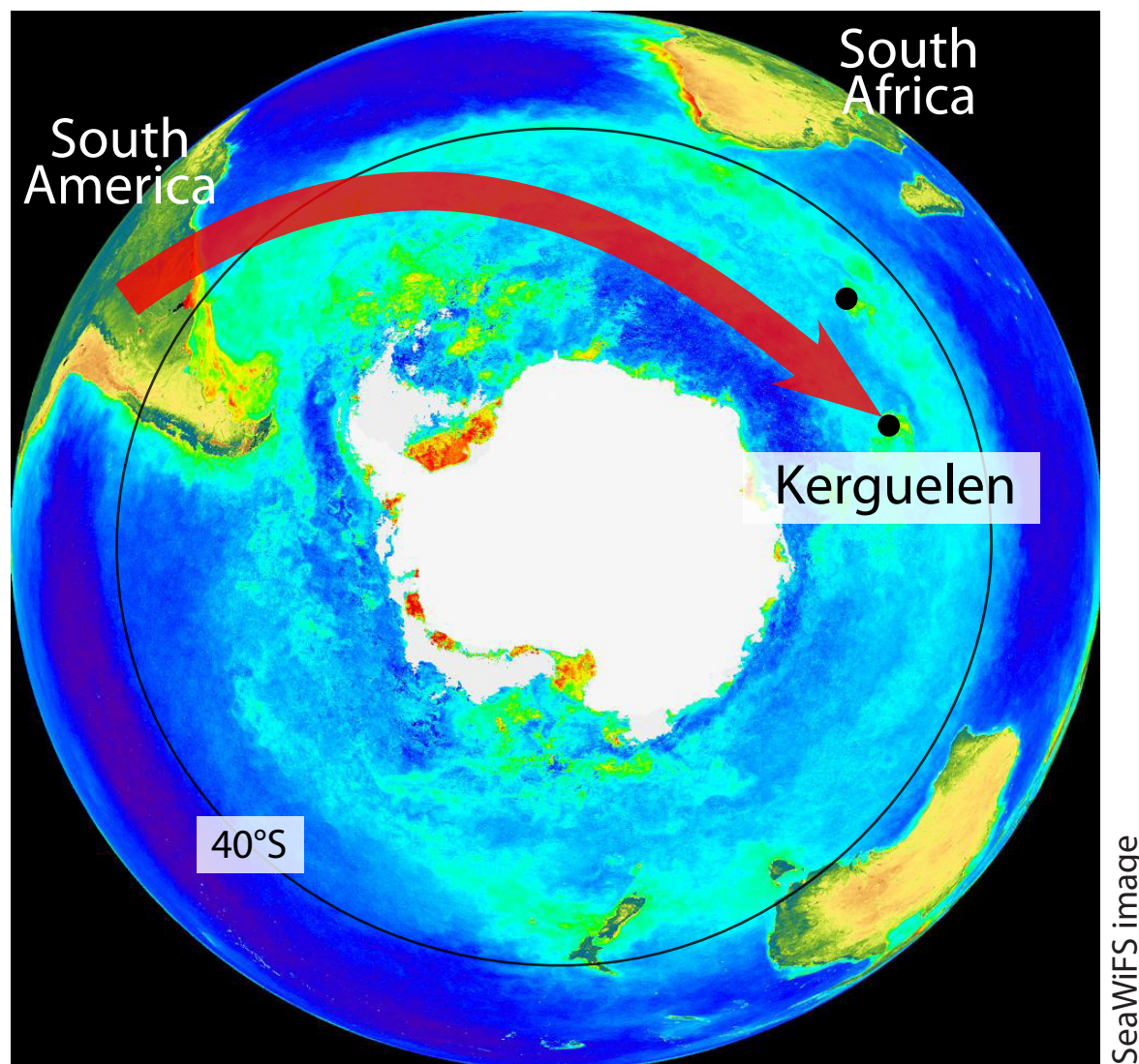
Crozet (enriched)?



Sources of deposition

Kerguelen:

Influenced
by dust coming
from South
America



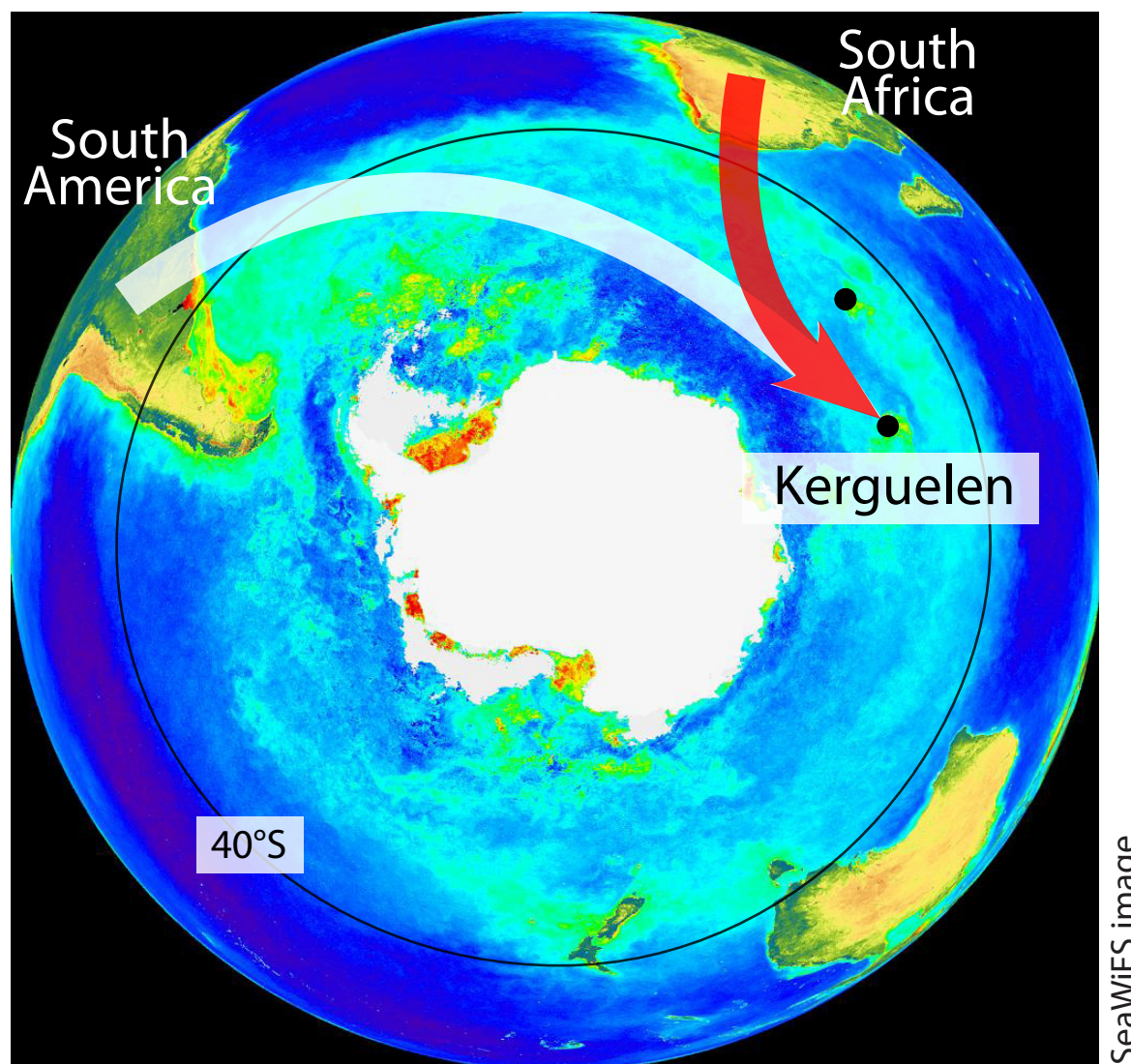


Sources of deposition

Kerguelen:

Influenced
by dust coming
from South
America

Deposition
influenced by
South Africa
during the
austral winter



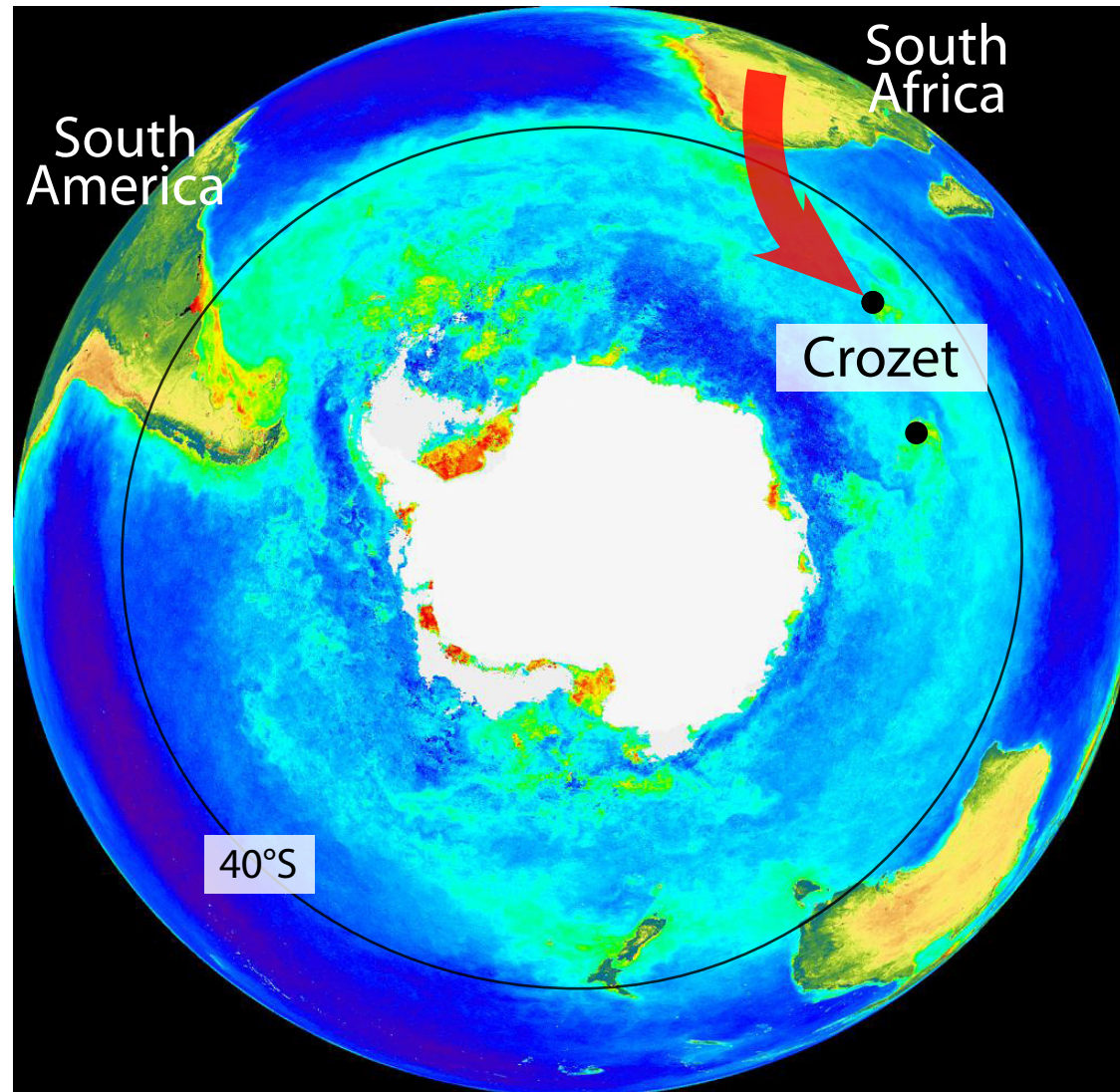


Sources of deposition

Kerguelen:

Influenced
by dust coming
from South
America

Deposition
influenced by
South Africa
during the
austral winter



Crozet:

Influenced
by dust coming
from South
Africa only

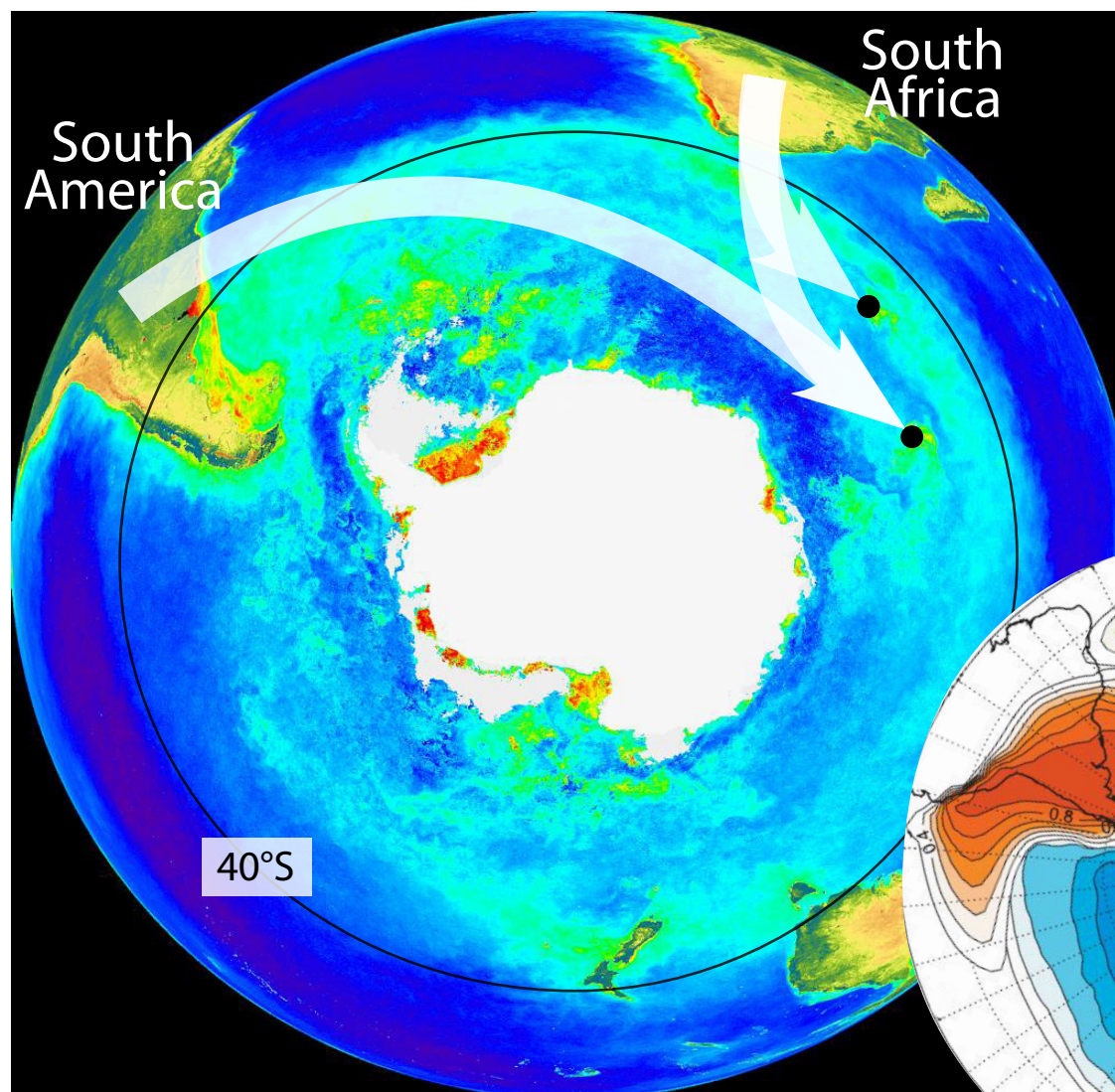


Sources of deposition

Kerguelen:

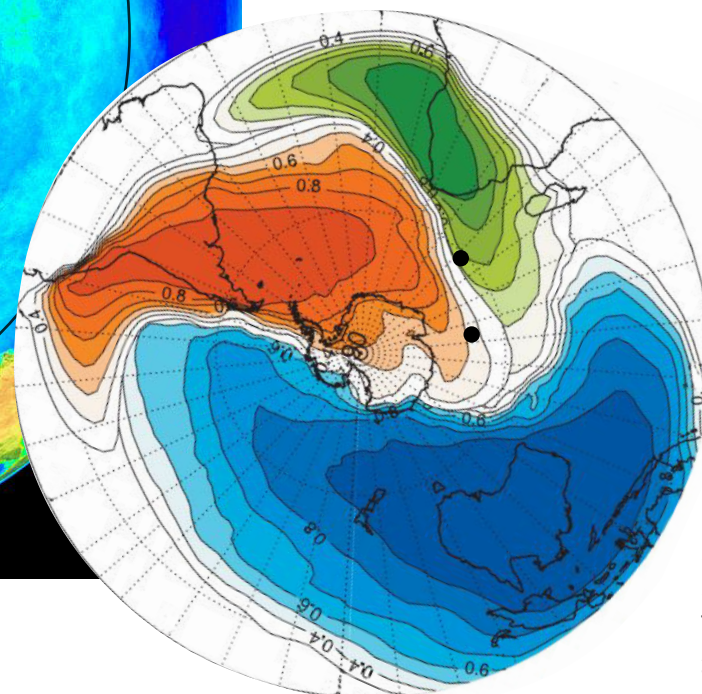
Influenced
by dust coming
from South
America

Deposition
influenced by
South Africa
during the
austral winter



Crozet:

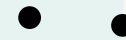
Influenced
by dust coming
from South
Africa only



Li et al., 2008

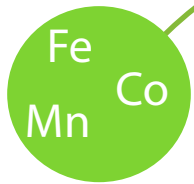
Issues

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2. What is its chemical composition?
3. What are its different sources?
4. What is its bioavailable part for phytoplankton?





Solubility



Bioavailability = Solubility



Solubility

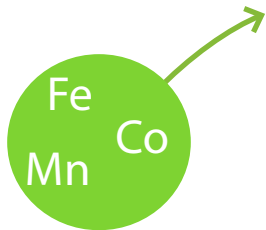


Bioavailability = Solubility

$$S\% = \frac{[X]_{\text{soluble}}}{[X]_{\text{soluble}} + [X]_{\text{insoluble}}}$$



Solubility



Bioavailability = Solubility

$$S\% = \frac{[X]_{\text{soluble}}}{[X]_{\text{soluble}} + [X]_{\text{insoluble}}}$$





Solubility

Element	Range (5 rains)
Al	67% - 96%
Ce	68% - 96%
Fe	51% - 91%
La	70% - 96%
Mn	66% - 94%
Nd	70% - 98%
Ti	33% - 83%

High solubilities (> 60%),
high solubility for other not measured elements (Ni, Cu, Co, Zn...)



Fe solubility in rainwater

● 23% ± 5%
Zhuang et al., 1992

● 21% ± 3%
Hawai
Zhuang et al., 1992

● 16 - 48%
Bermuda
Kieber et al., 2003

● 20 - 40%
Buck et al., 2010

● 18% ± 23%
Finokalia
Theodosi et al., 2010

● 51% - 91%
Kerguelen islands
this study



Bioavailable (dissolved) Fe flux

Calculation from:

1) Median of Fe solubility = 82%

Element	Range (5 rains)
Fe	51% - 91%

2) Fe flux on Kerguelen islands of $28 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Heimburger et al., 2012),

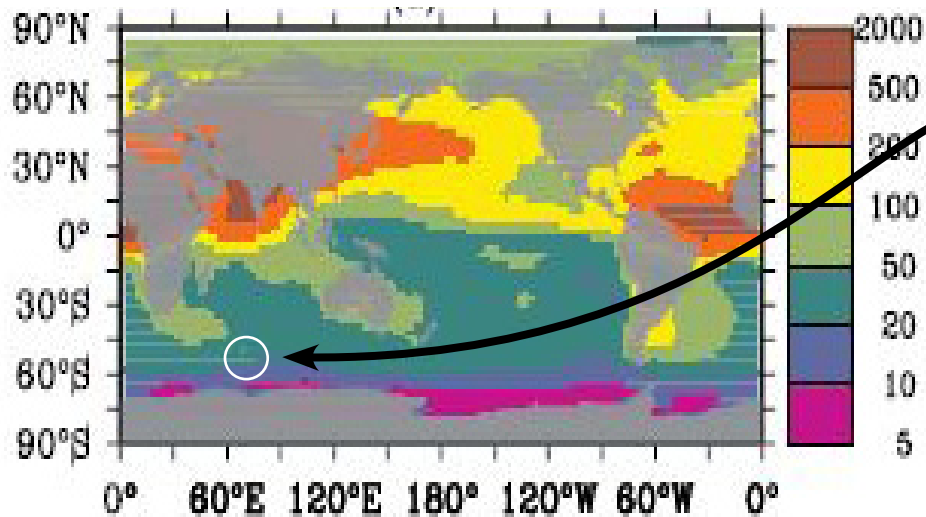
Bioavailable Fe flux:
 $23 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ($412 \text{ nmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)



Dissolved Fe flux in literature

Models

$23 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ($412 \text{ nmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)
with a solubility of 82%
(Heimburger et al., in review)



Estimated dissolved Fe deposition flux
with a solubility of 17%:
between 3 and $8 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
(between 54 and $143 \text{ nmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)
(Fan et al., 2006)



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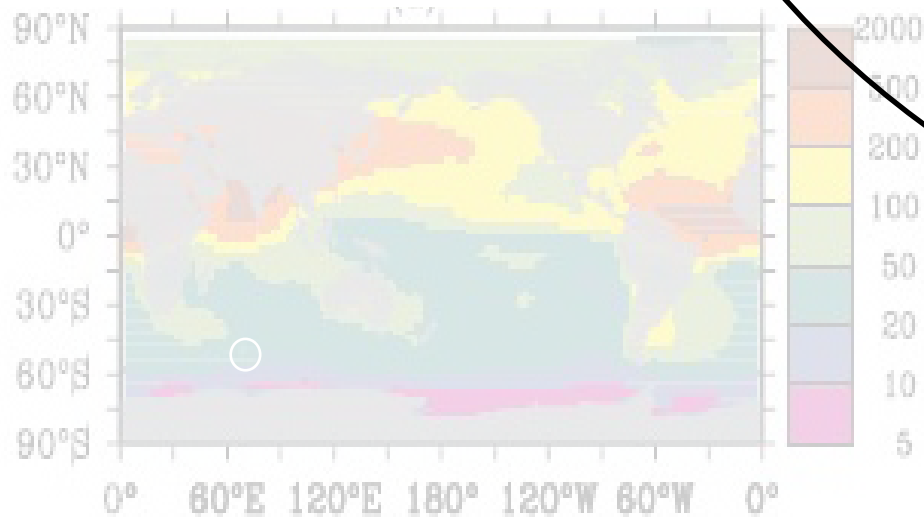
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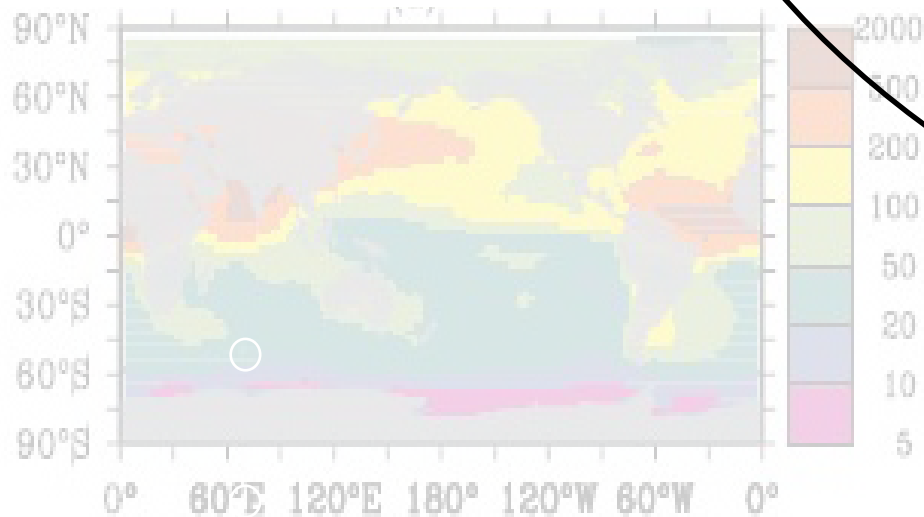
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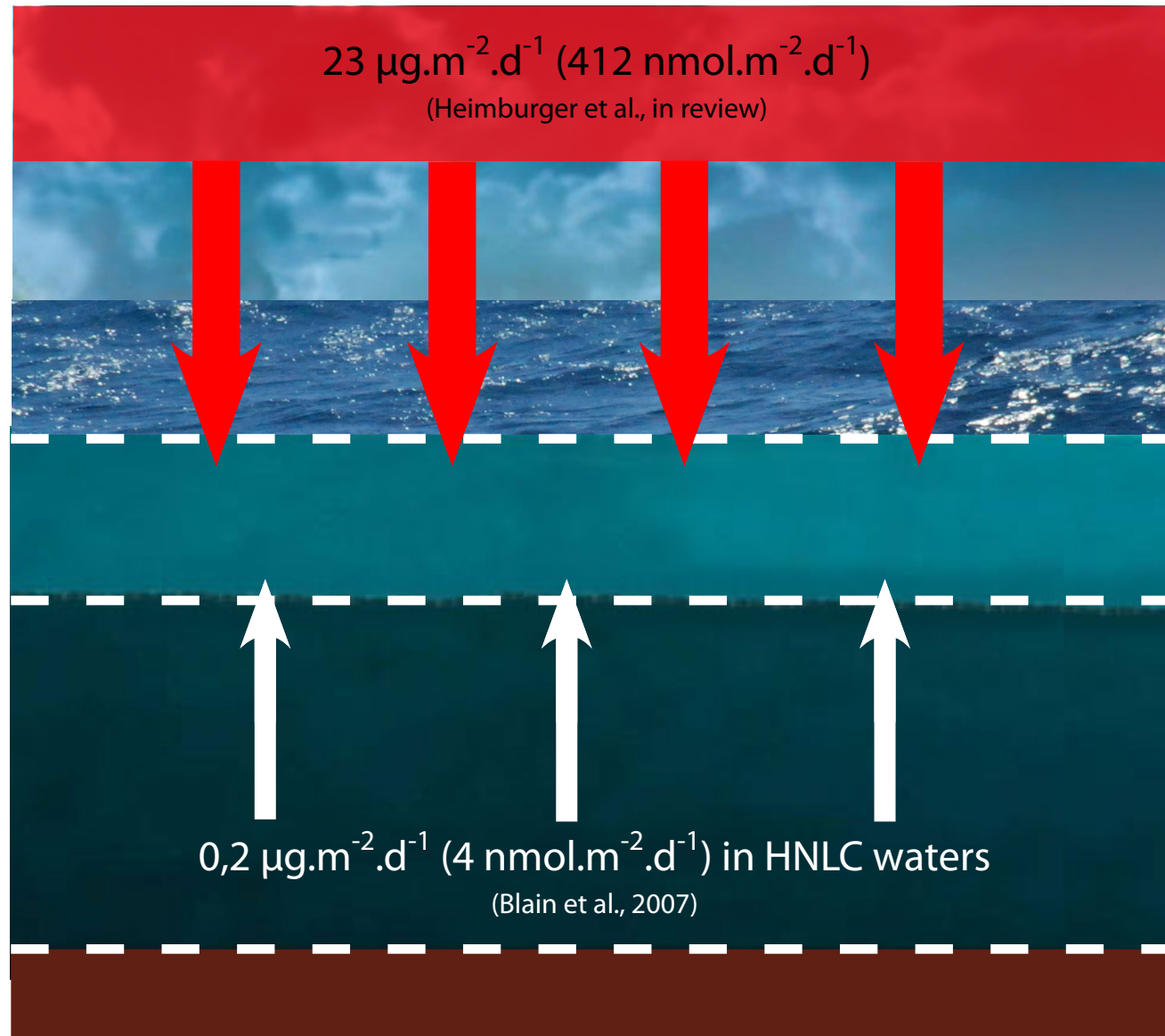
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Atmospheric vs. oceanic inputs





Bioavailability

High dissolved micronutrient fluxes
over the South Indian Ocean due to high solubility

Micronutrient inputs coming from atmospheric deposition
and bioavailable for phytoplankton are more important
in the micronutrient oceanic budget than expected.

Conclusion

1. First direct measurement of dust flux over the South Indian Ocean

$$F_{\text{dust}} = 650 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$$

In line with atmospheric models but not with previous observation:

Atmospheric deposition flux more important than expected for micronutrient supply.

Using C_{aerosol} to find total deposition flux over remote oceanic areas seems not appropriate because aerosols collected at surface level are not representative of aerosols scavenged by precipitations.

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Anthropogenic contribution observed during the Austral winter

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4. First measurement of solubility in rainwater events in the South Indian Ocean
High solubility ($> 60\%$)
Bioavailable Fe flux = $23 \mu\text{g.m}^{-2}.\text{d}^{-1}$, i.e. 5 times higher than the ones previously predicted and observed. This result suggests that atmospheric supply is the major source of micronutrients for phytoplankton in the studied area.

Perspectives and remaining questions

1. In order to confirm the hypothesis that aerosols collected at surface level do not allow total atmospheric deposition flux to be well calculated observed remote oceanic area, vertical aerosol measurements need to be performed.
2. In order to confirm deposition sources highlighted by Pb isotopes, new tracers are needed such as REE profiles.
3. It is necessary to confirm solubility values found in rainwater samples by new field experiments on solubility in the South Indian Ocean.

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- What are the causes responsible for seasonality observed in atmospheric dust deposition during the austral spring? (intensity of dust emissions from sources, transport efficiency, ...)
 - What is the connection between higher dust deposition fluxes and phytoplankton bloom, both observed during the austral spring?
 - Why aerosol solubility observed in rainwater and collected on Kerguelen Islands is so high? Which processes lead to such values?

Thanks for your attention



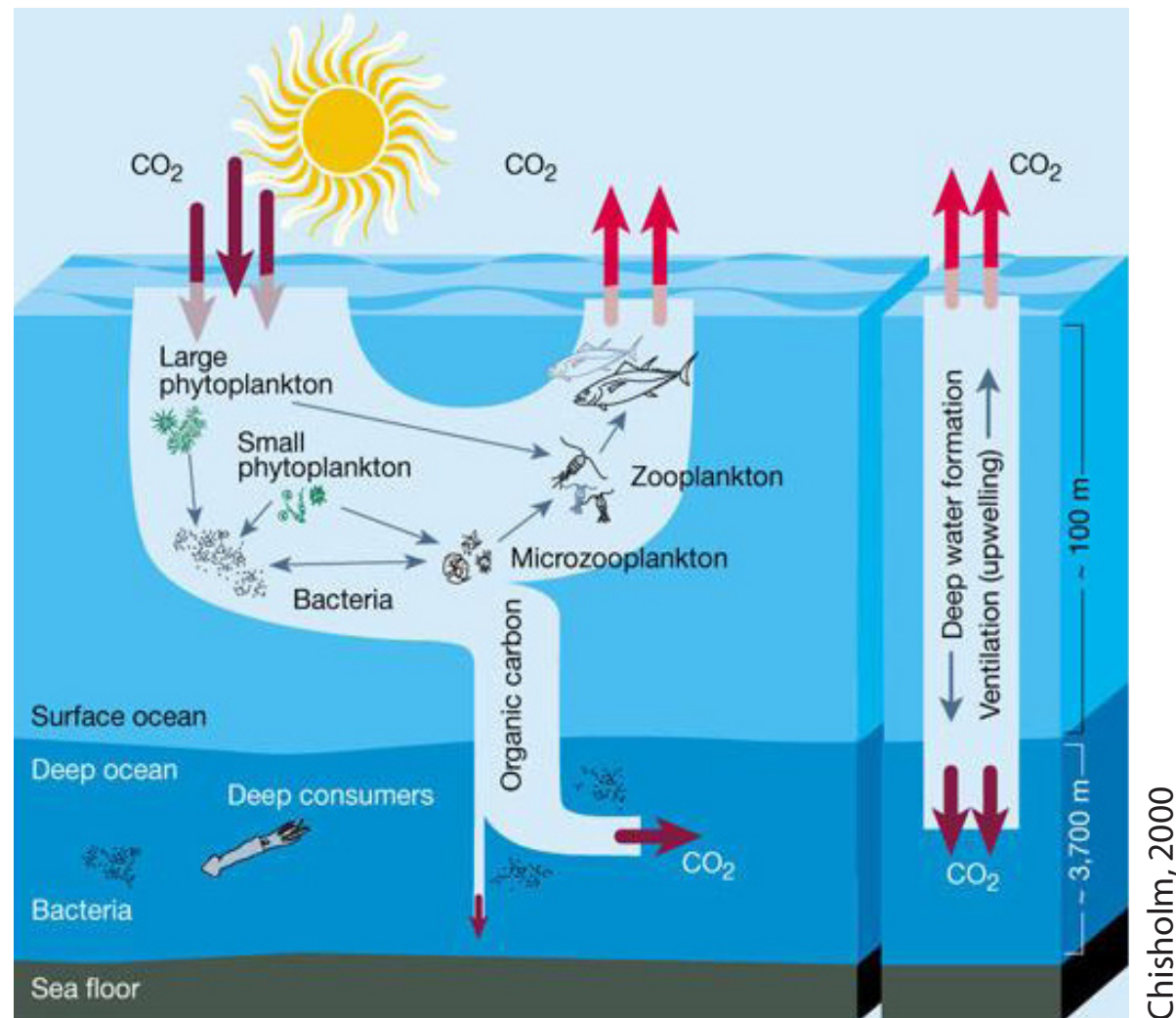
Scientific context

Issues

Methods

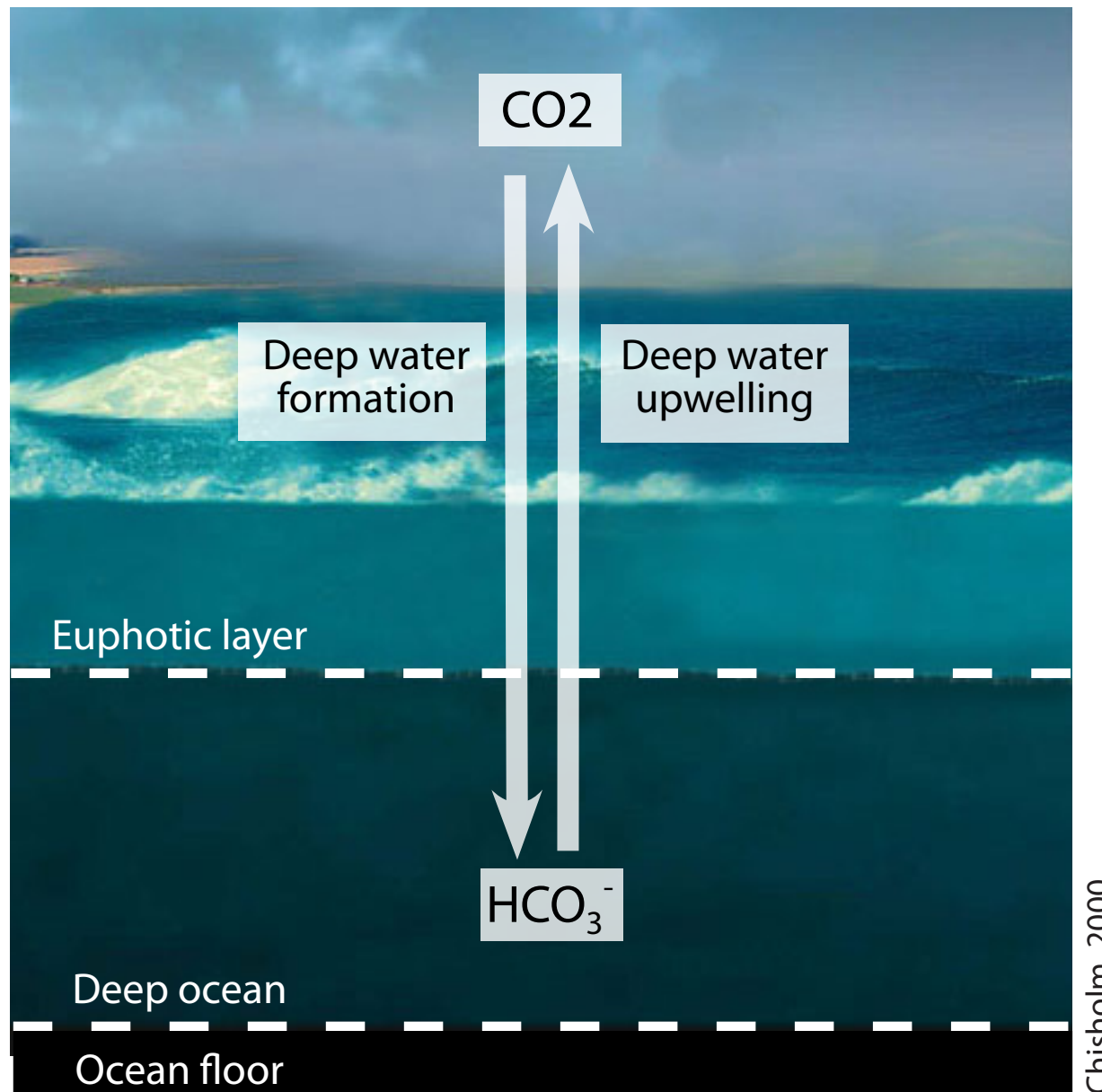
Results

Oceanic CO₂ pump

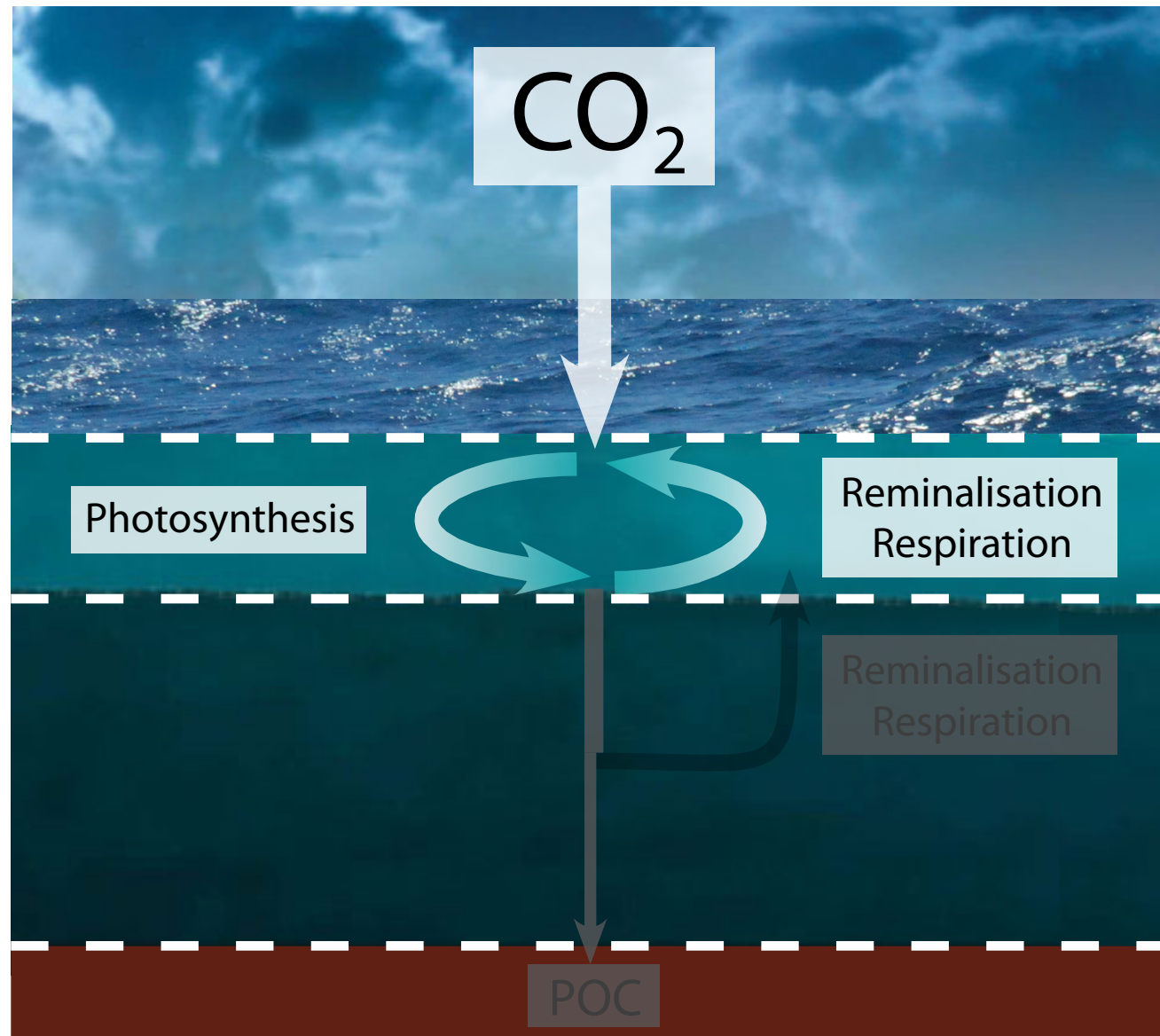


The open ocean is an important sink of carbon
 Strong contribution of high southern latitudes

Oceanic CO₂ physical pump

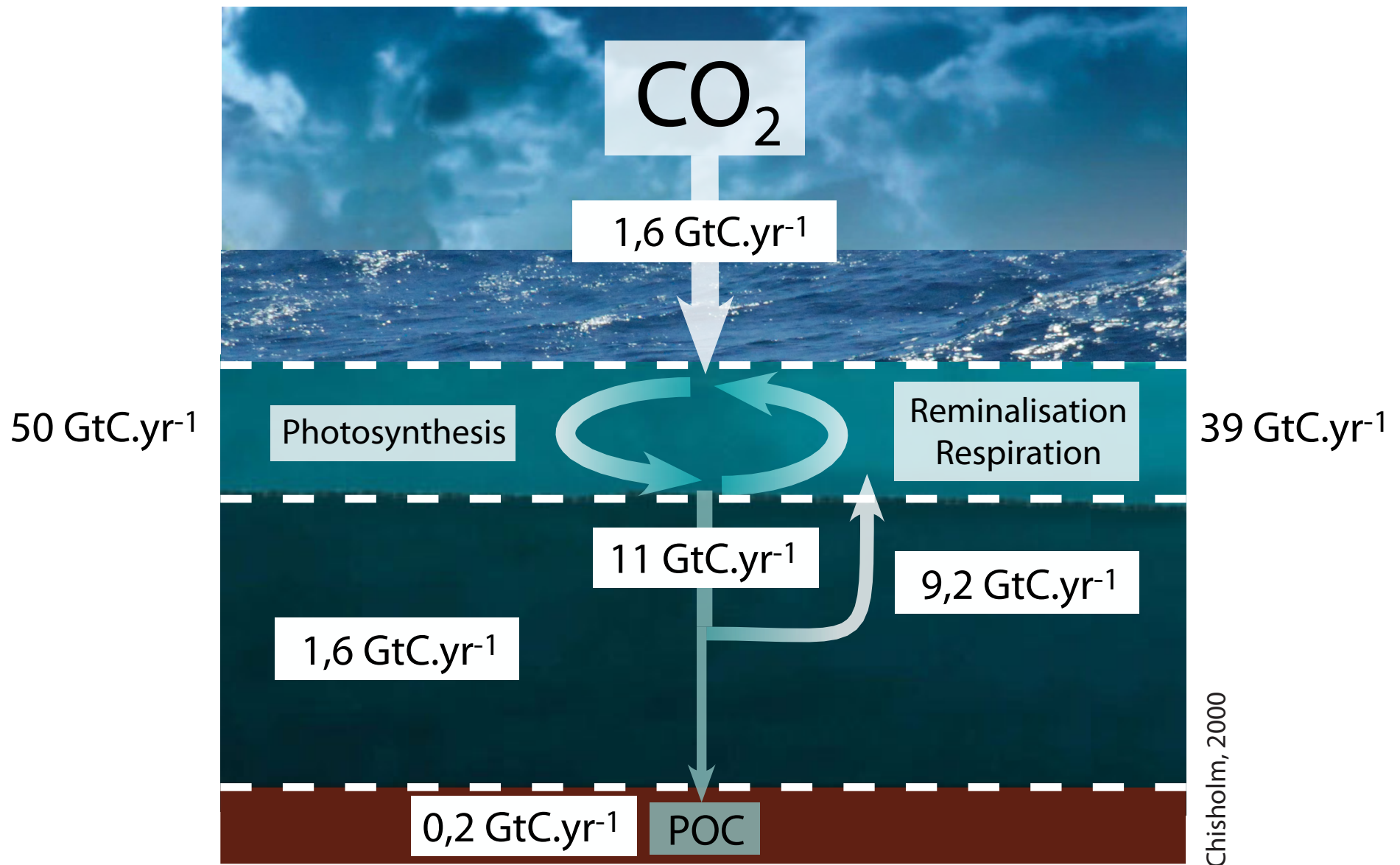


Oceanic CO₂ biological pump

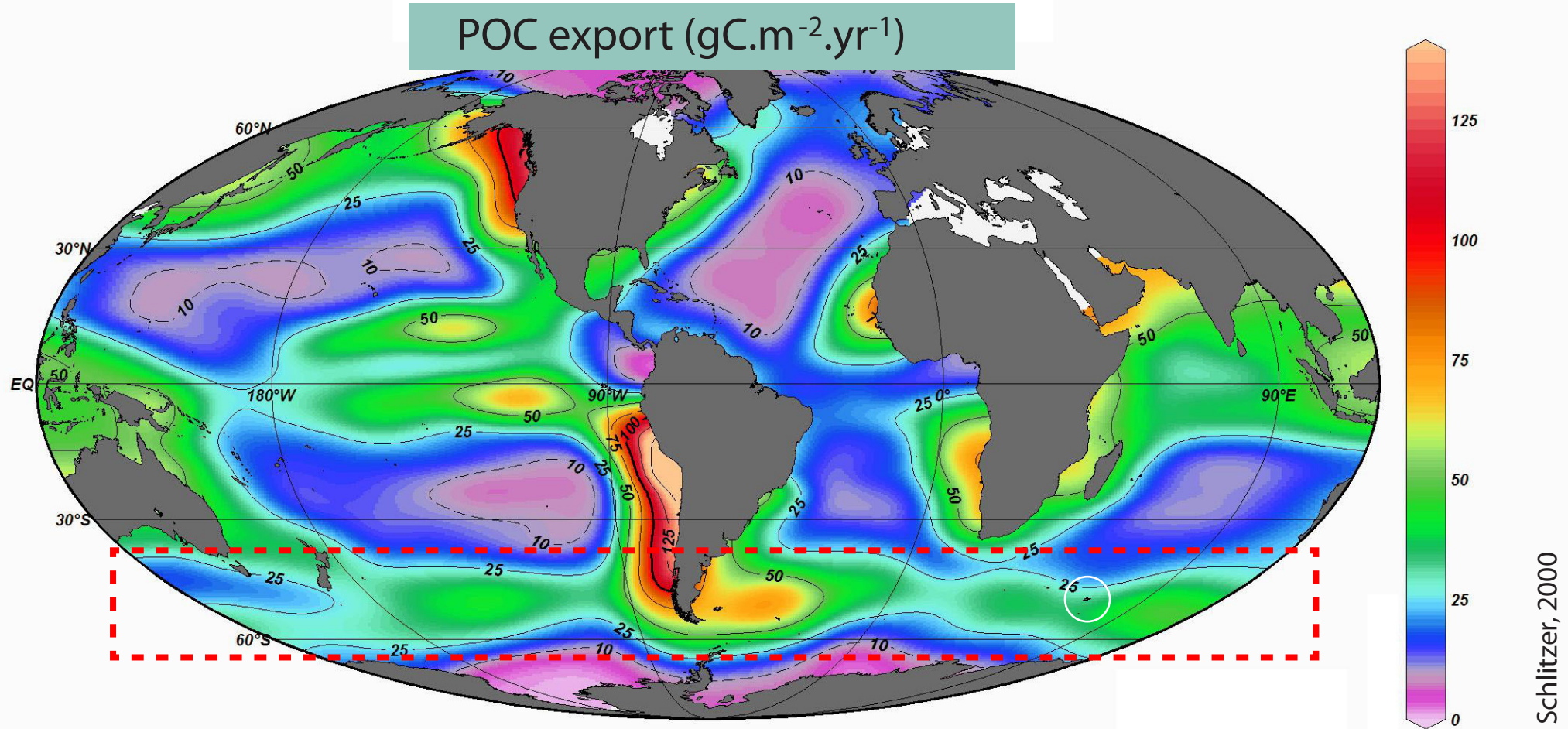


Chisholm, 2000

Oceanic CO₂ biological pump

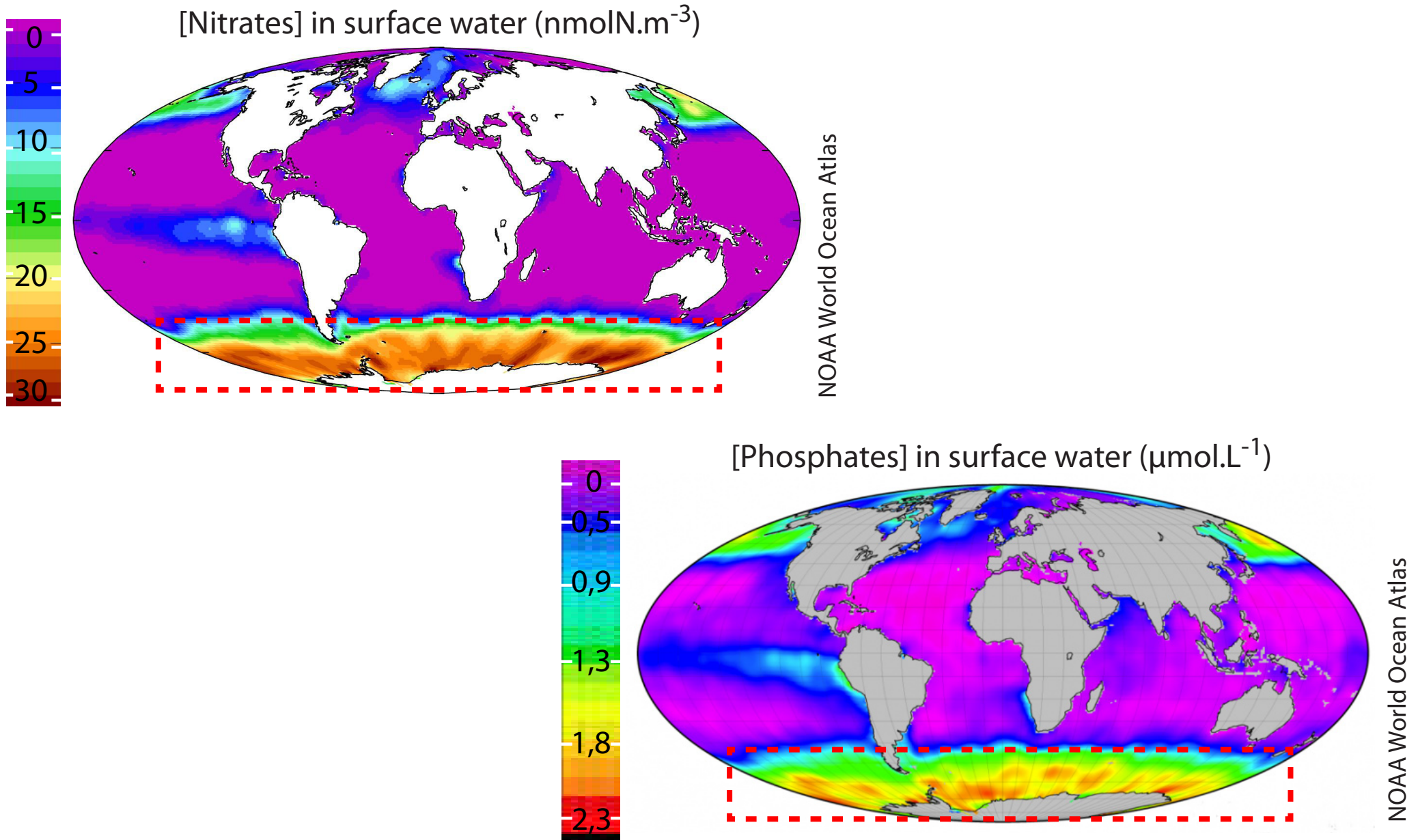


Carbon export in the open ocean



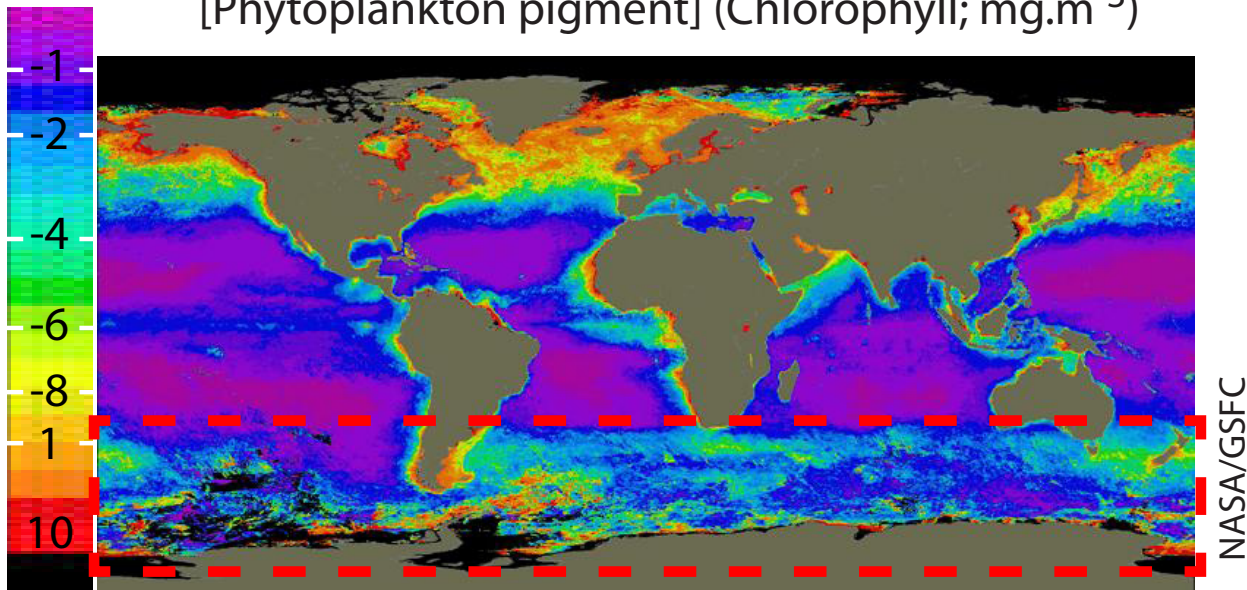
Strong contribution
of high southern latitudes

The Southern Ocean: HNLC area

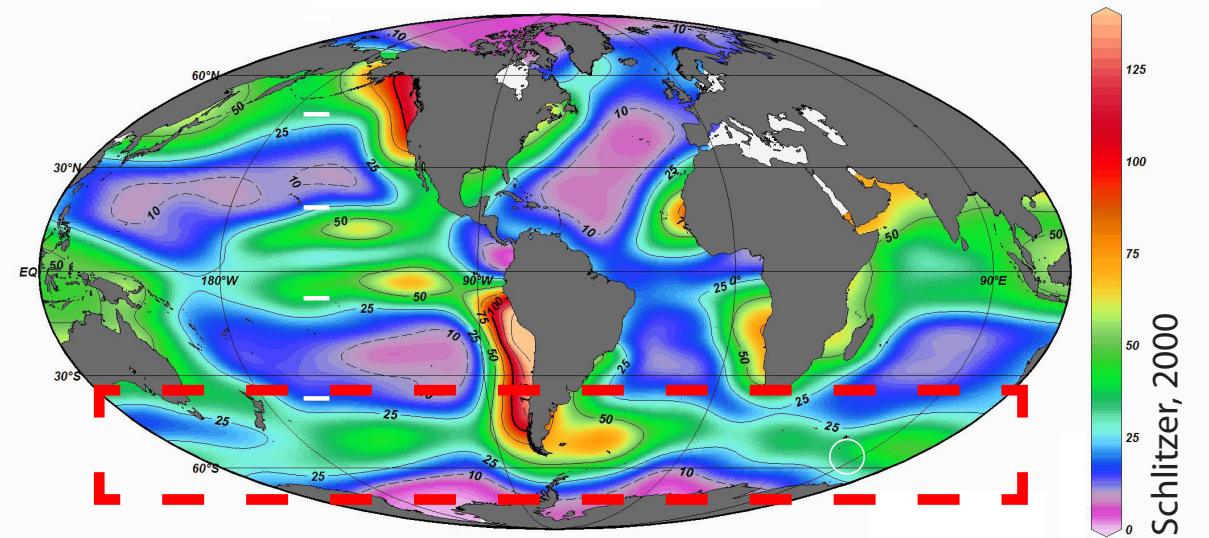


POC vs phytoplankton

[Phytoplankton pigment] (Chlorophyll; $\text{mg}\cdot\text{m}^{-3}$)



POC export ($\text{gC}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$)



37 analyzed and validated elements

ICP-AES and HR-ICP-MS
(clean room)



hydrogen 1 H 1.0079																		helium 2 He 4.0026
lithium 3 Li 6.941	beryllium 4 Be 9.0122											boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180	
sodium 11 Na 22.990	magnesium 12 Mg 24.305											aluminum 13 Al 26.982	silicon 14 Si 28.086	phosphorus 15 P 30.974	sulfur 16 S 32.065	chlorine 17 Cl 35.453	argon 18 Ar 39.948	
potassium 19 K 39.098	calcium 20 Ca 40.078	scandium 21 Sc 44.956	titanium 22 Ti 47.867	vanadium 23 V 50.942	chromium 24 Cr 51.996	manganese 25 Mn 54.938	iron 26 Fe 55.845	cobalt 27 Co 58.933	nickel 28 Ni 58.693	copper 29 Cu 63.546	zinc 30 Zn 65.39	gallium 31 Ga 69.723	germanium 32 Ge 72.61	arsenic 33 As 74.922	selenium 34 Se 78.96	bromine 35 Br 79.904	krypton 36 Kr 83.80	
rubidium 37 Rb 85.468	strontium 38 Sr 87.62	yttrium 39 Y 88.906	zirconium 40 Zr 91.224	niobium 41 Nb 92.906	molybdenum 42 Mo 95.94	technetium 43 Tc [98]	ruthenium 44 Ru 101.07	rhodium 45 Rh 102.91	palladium 46 Pd 106.42	silver 47 Ag 107.87	cadmium 48 Cd 112.41	indium 49 In 114.82	tin 50 Sn 118.71	antimony 51 Sb 121.76	tellurium 52 Te 127.60	iodine 53 I 126.90	xenon 54 Xe 131.29	
caesium 55 Cs 132.91	barium 56 Ba 137.33	57-70 *	lutetium 71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	platinum 78 Pt 195.08	gold 79 Au 196.97	mercury 80 Hg 200.59	thallium 81 Tl 204.38	lead 82 Pb 207.2	bismuth 83 Bi 208.98	polonium 84 Po [209]	astatine 85 At [210]	radon 86 Rn [222]
francium 87 Fr [223]	radium 88 Ra [226]	89-102 * *	lanthanum 103 La [227]	rutherfordium 104 Rf [261]	dubnium 105 Db [262]	seaborgium 106 Sg [266]	bohrium 107 Bh [264]	hassium 108 Hs [269]	meitnerium 109 Mt [268]	ununilium 110 Uun [271]	unununium 111 Uuu [272]	ununbium 112 Uub [277]		ununquadium 114 Uuq [289]				

lanthanum 57 La 138.91	cerium 58 Ce 140.12	praseodymium 59 Pr 140.91	neodymium 60 Nd 144.24	promethium 61 Pm [145]	samarium 62 Sm 150.36	europium 63 Eu 151.96	gadolinium 64 Gd 157.25	terbium 65 Tb 158.93	dysprosium 66 Dy 162.50	holmium 67 Ho 164.93	erbium 68 Er 167.26	thulium 69 Tm 168.93	ytterbium 70 Yb 173.04
actinium 89 Ac [227]	thorium 90 Th 232.04	protactinium 91 Pa 231.04	uranium 92 U 238.03	neptunium 93 Np [237]	plutonium 94 Pu [244]	americium 95 Am [243]	curium 96 Cm [247]	berkelium 97 Bk [247]	californium 98 Cf [251]	einsteinium 99 Es [252]	fermium 100 Fm [257]	mendelevium 101 Md [258]	nobelium 102 No [259]

Soil sampling

Goals:

Field prospecting N/N-W of «Jacky» and «Guillou» sites: sampling of soils potentially erodable

Local emissions of dust?

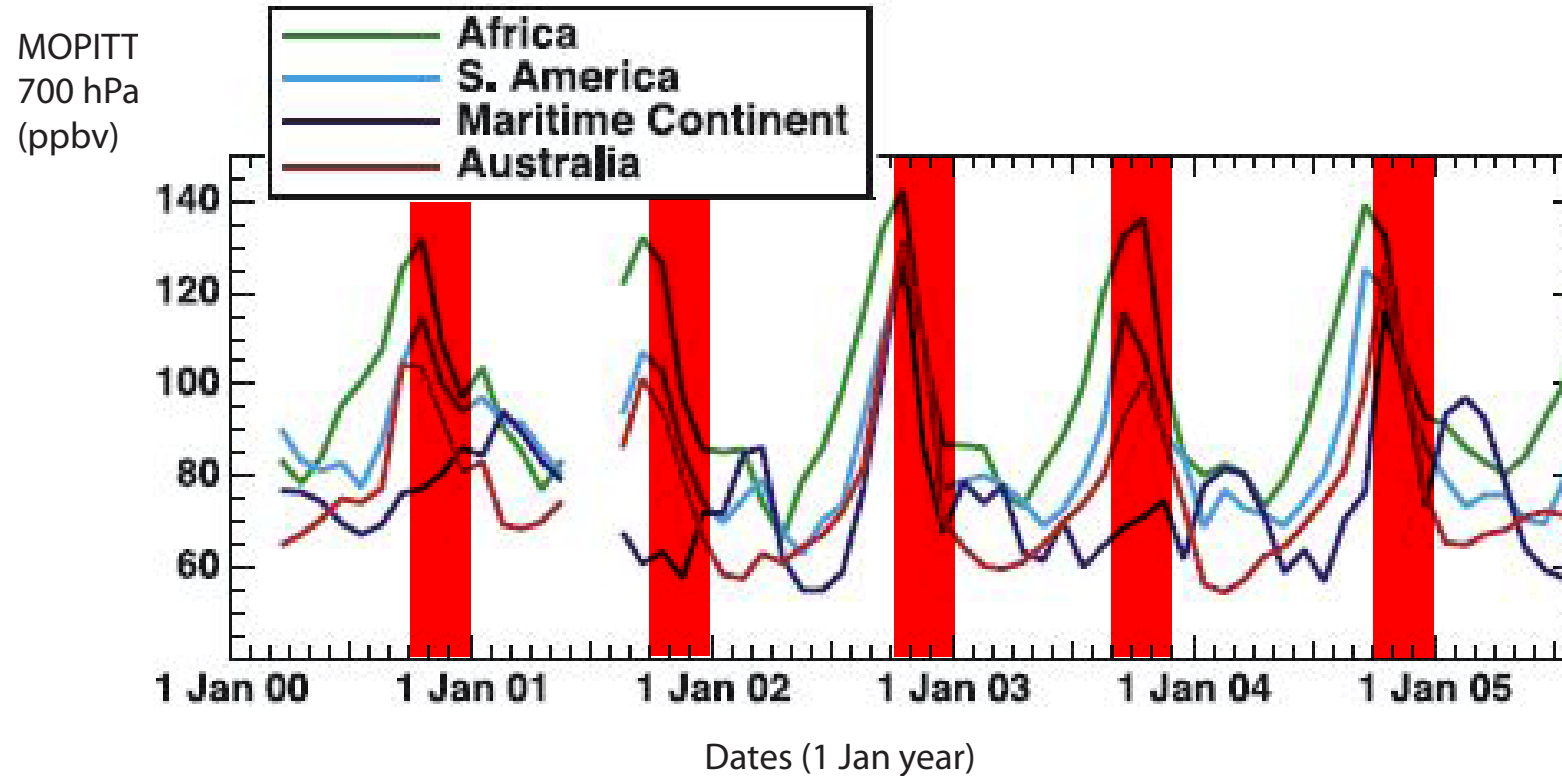
32 collected soils:

- over 500 km²
- analyses using X ray fluorescence spectrometry (Ti, Al)





Total deposition flux: variability



Biomass burning?



Total dust deposition flux in literature

$$F_{\text{dry}} = C_{\text{aerosol}} \cdot V_{\text{dry deposition}}$$



1-3 cm.s⁻¹ over the South Indian Ocean

(Ezat and Dulac, 1995; Wagener et al, 2008)

	FLATOCOA	Wagener et al., 2008
Al concentrations in aerosols (ng.m ⁻³) median ± SD	1,86 ± 1,75	1,00 ± 0,49
Al dry deposition flux (µg.m ⁻² .d ⁻¹)	20 - 60	31 ± 11



Total dust deposition flux in literature

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Al concentration in aerosols (ng.m ⁻³) median ± SD	1,86 ± 1,75	1,00 ± 0,49
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of the total deposition



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$$3,01 \pm 0,3 \text{ mm.d}^{-1} \text{ (Wagener et al., 2008)}$$

$$2 - 3 \text{ mm.d}^{-1} \text{ (FLATOCOA)}$$



Total dust deposition flux in literature

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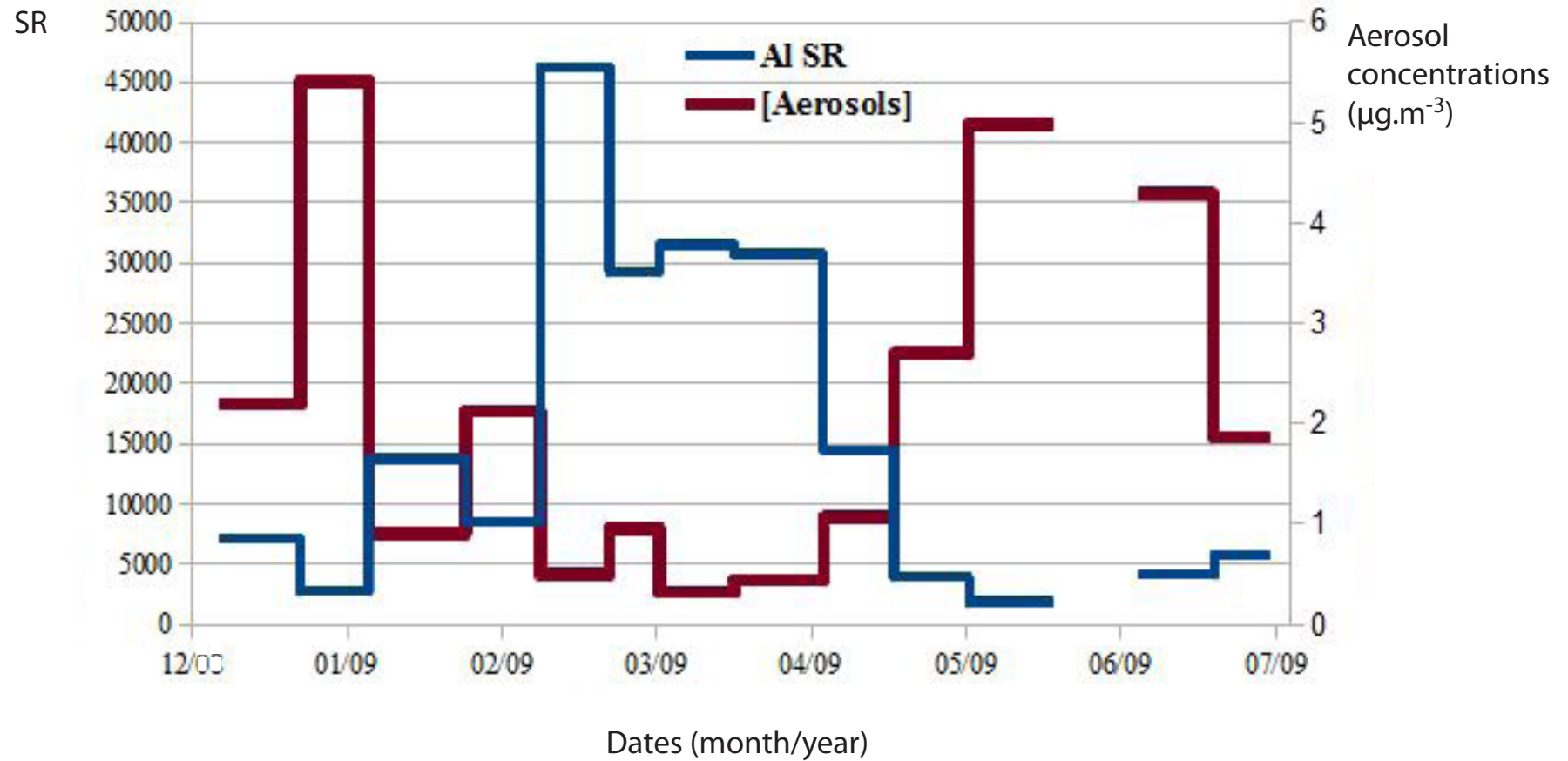
Difference between fluxes is due to wet deposition calculation

$$F_{\text{wet}} = C_{\text{aerosol}} \cdot \text{SR} \cdot \text{rainfall}$$

$$\text{SR} = \frac{C_{i, \text{rain}} \cdot \rho_{\text{air}}}{C_{i, \text{aerosol}}}$$

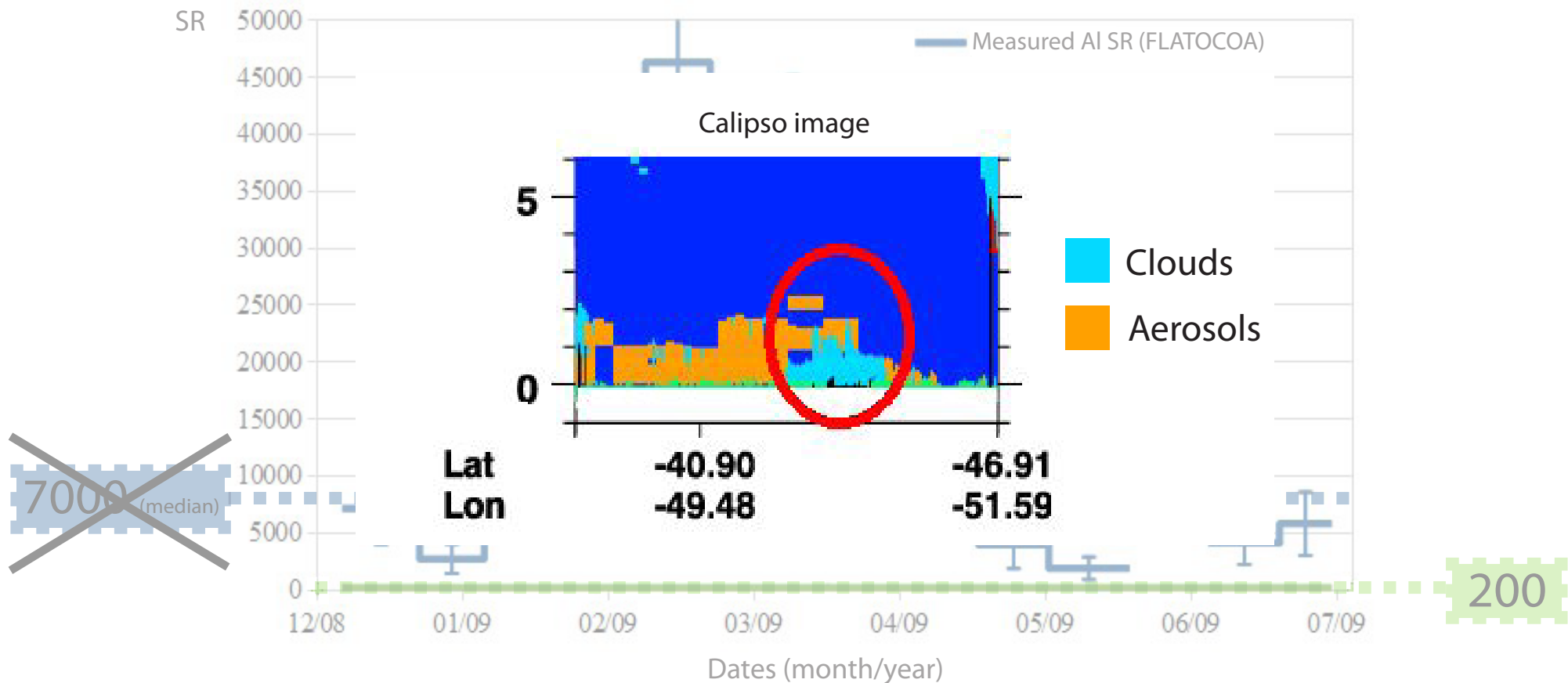


Scavenging ratio





Scavenging ratio



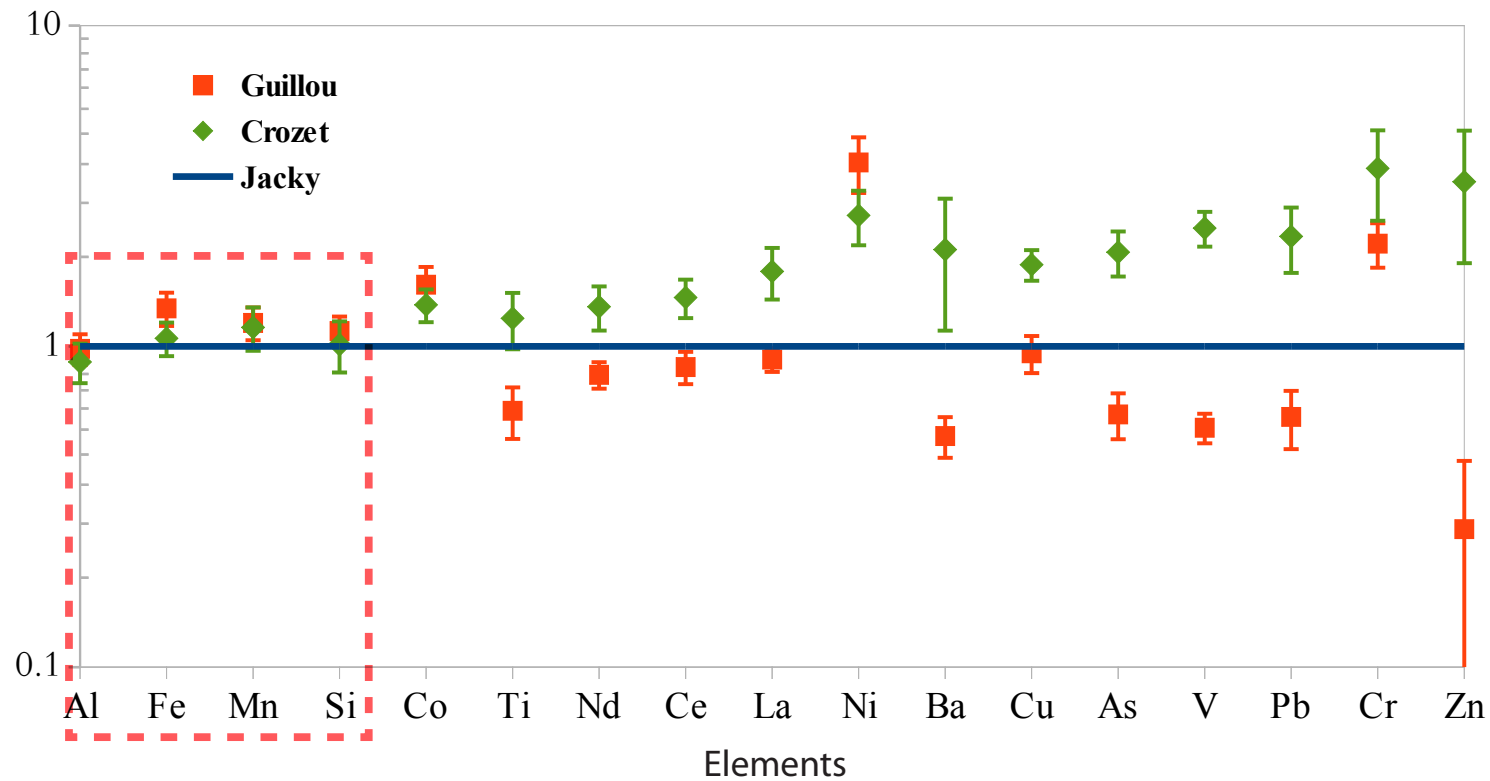
Hypothesis:

aerosols scavenged by rain are in altitude and not at surface level



Total deposition fluxes

Ratio:
flux at «X» / flux at «jacky»

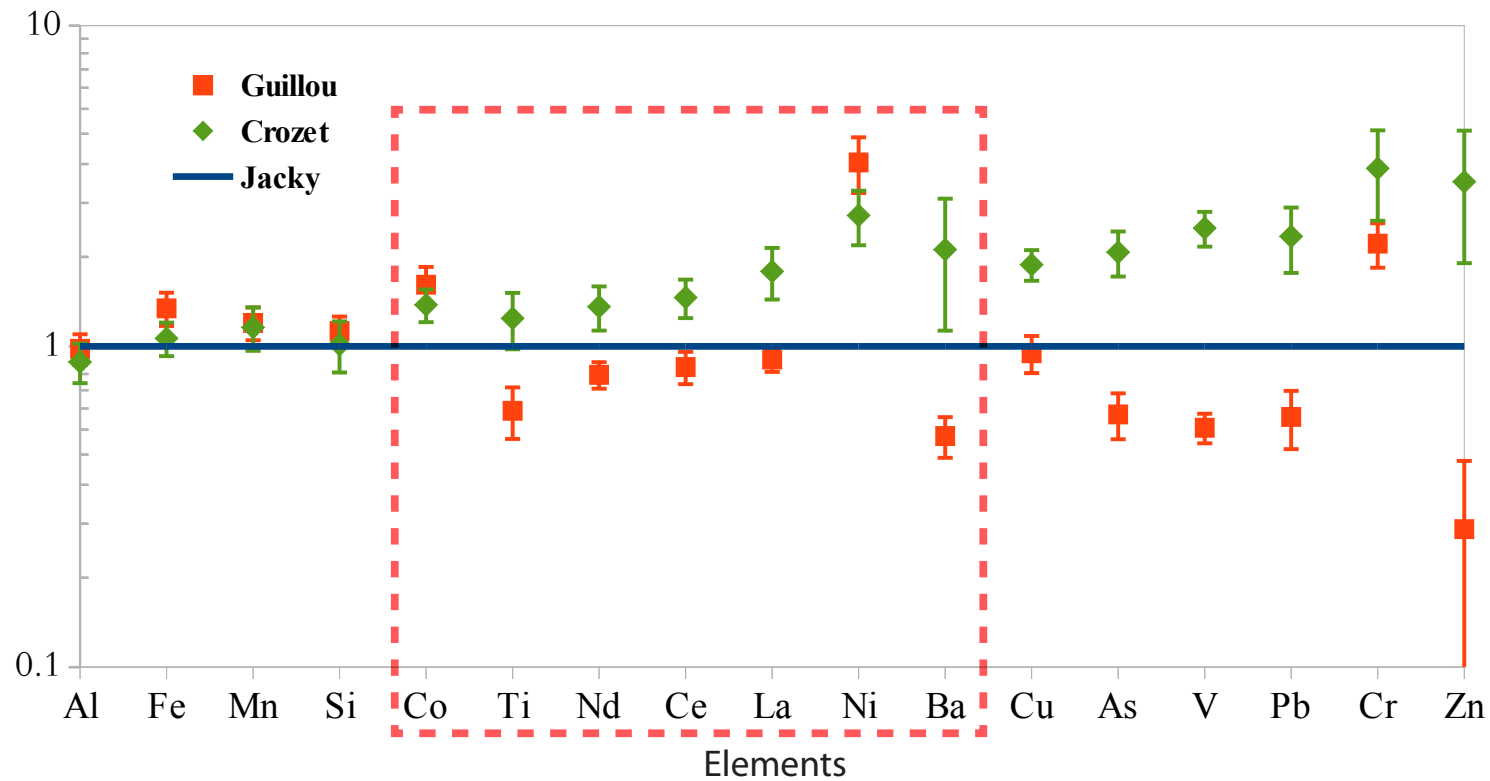


Al, Fe, Mn, Si:
Similar fluxes for the 3 sites (Ker and Cro: 1400km apart)



Total deposition fluxes

Ratio:
flux at «X» / flux at «jacky»

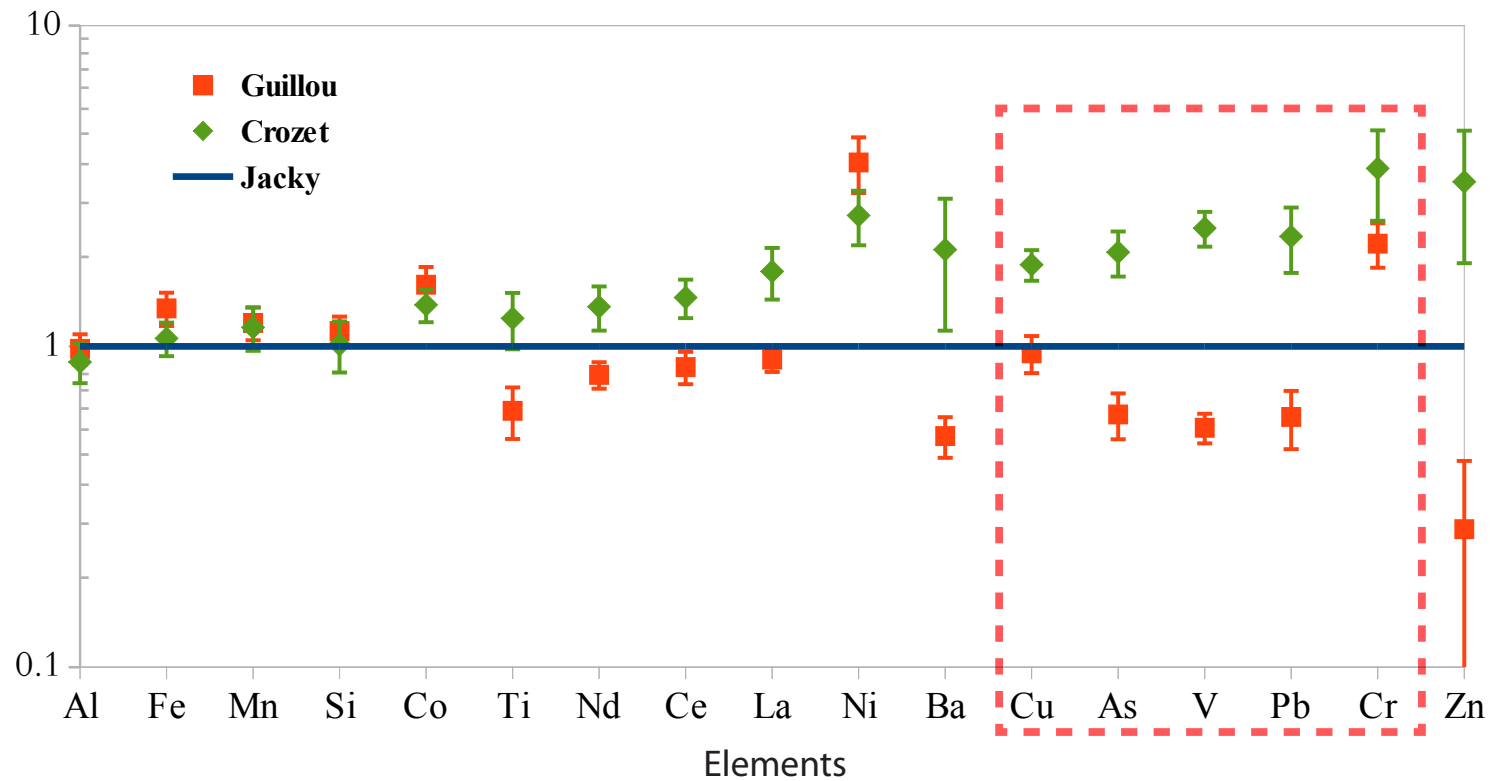


Differences between fluxes
for Co, Ti, Nd, Ce, La, Ni and Ba

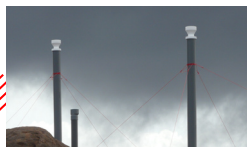


Total deposition fluxes

Ratio:
flux at «X» / flux at «jacky»

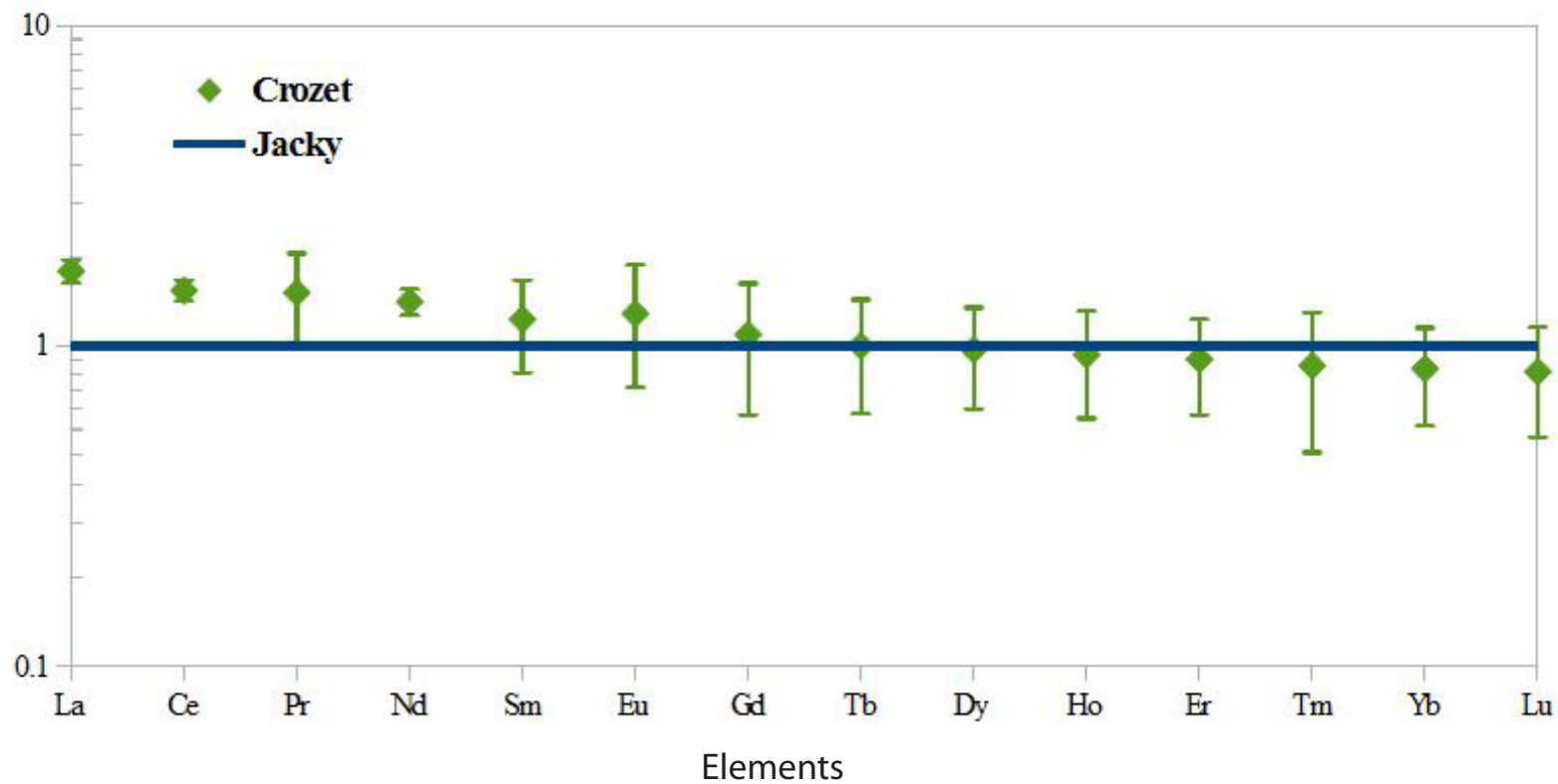


Differences between fluxes
for Cu, As, V, Pb and Cr



Total deposition fluxes: REE

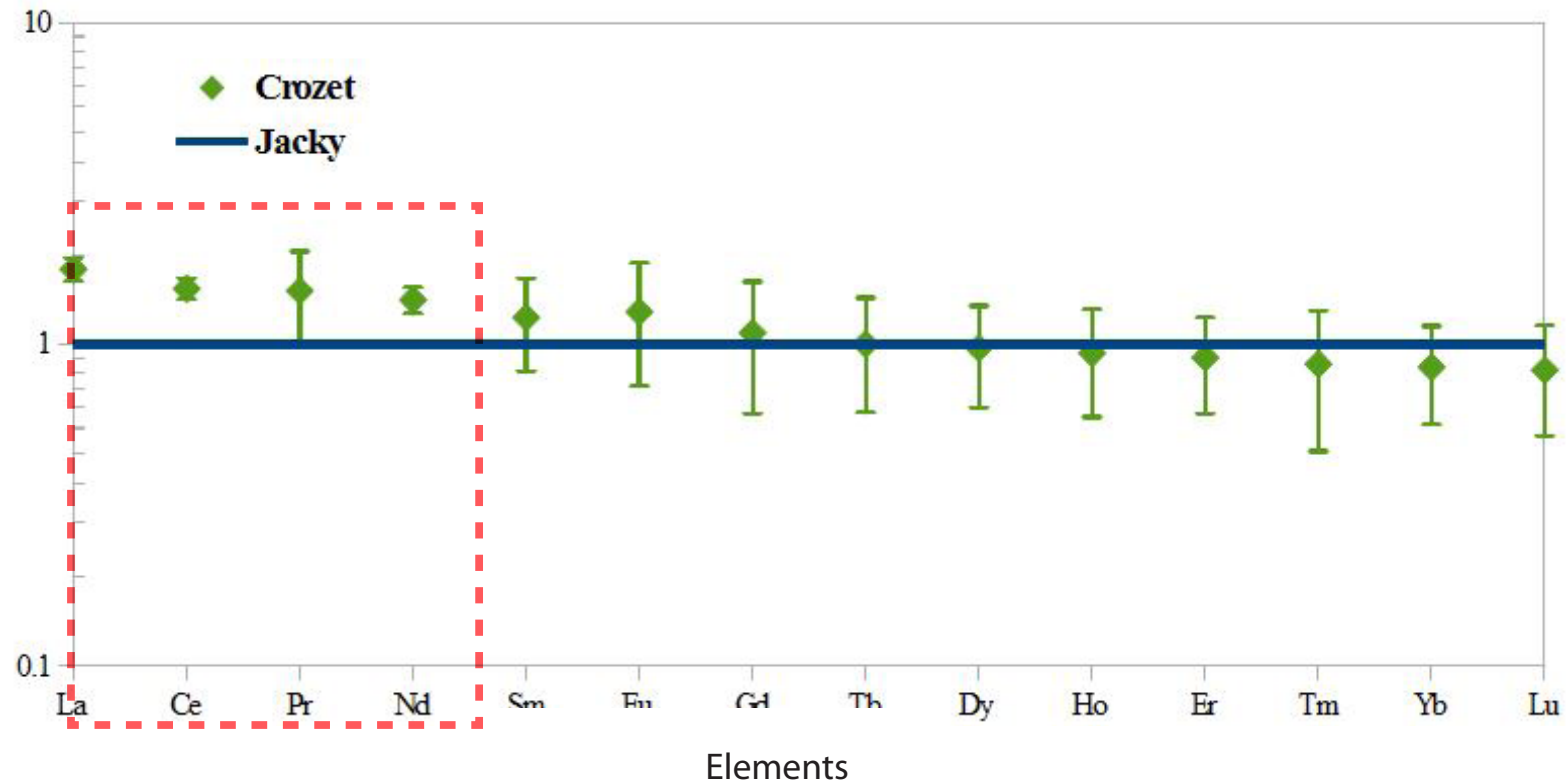
Ratio:
flux at «X» / flux at «jacky»





Total deposition fluxes: REE

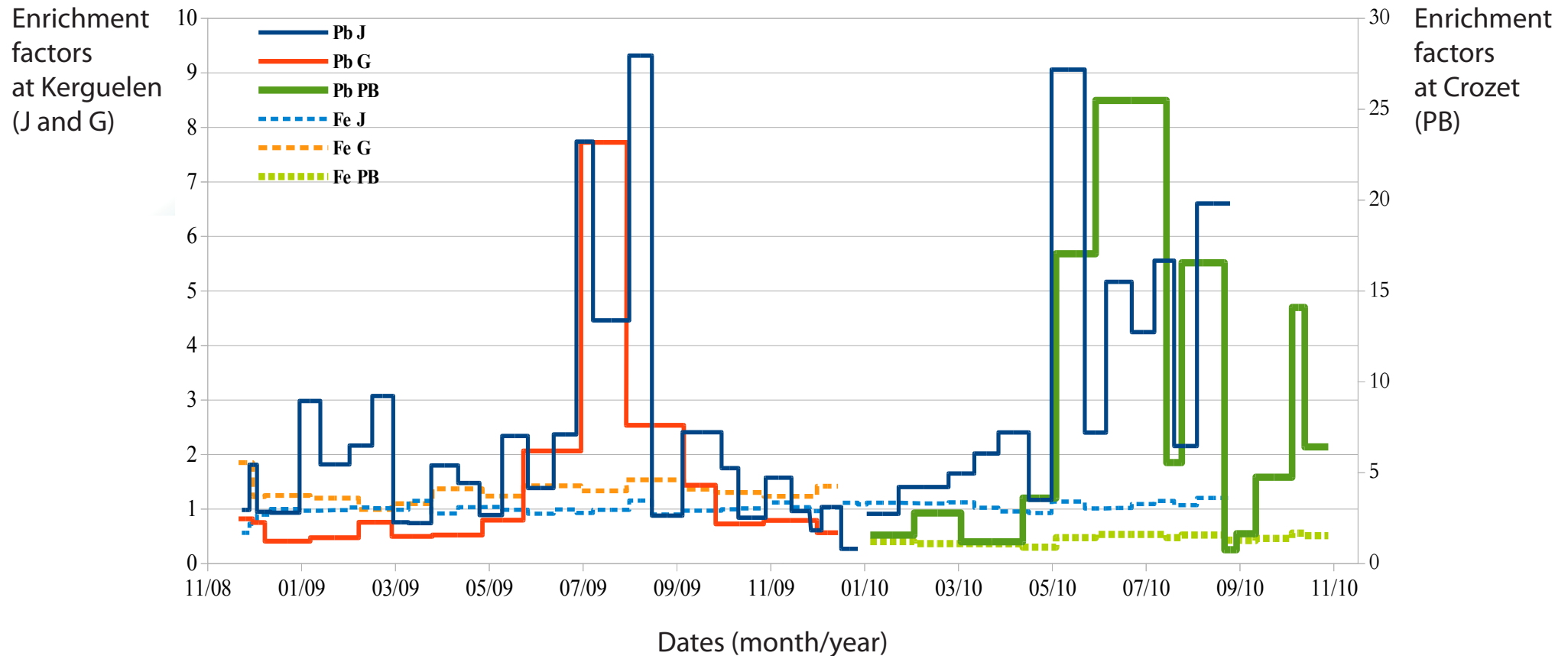
Ratio:
flux at «X» / flux at «jacky»



Enrichement of LREE
on Crozet Island



Enrichment factors



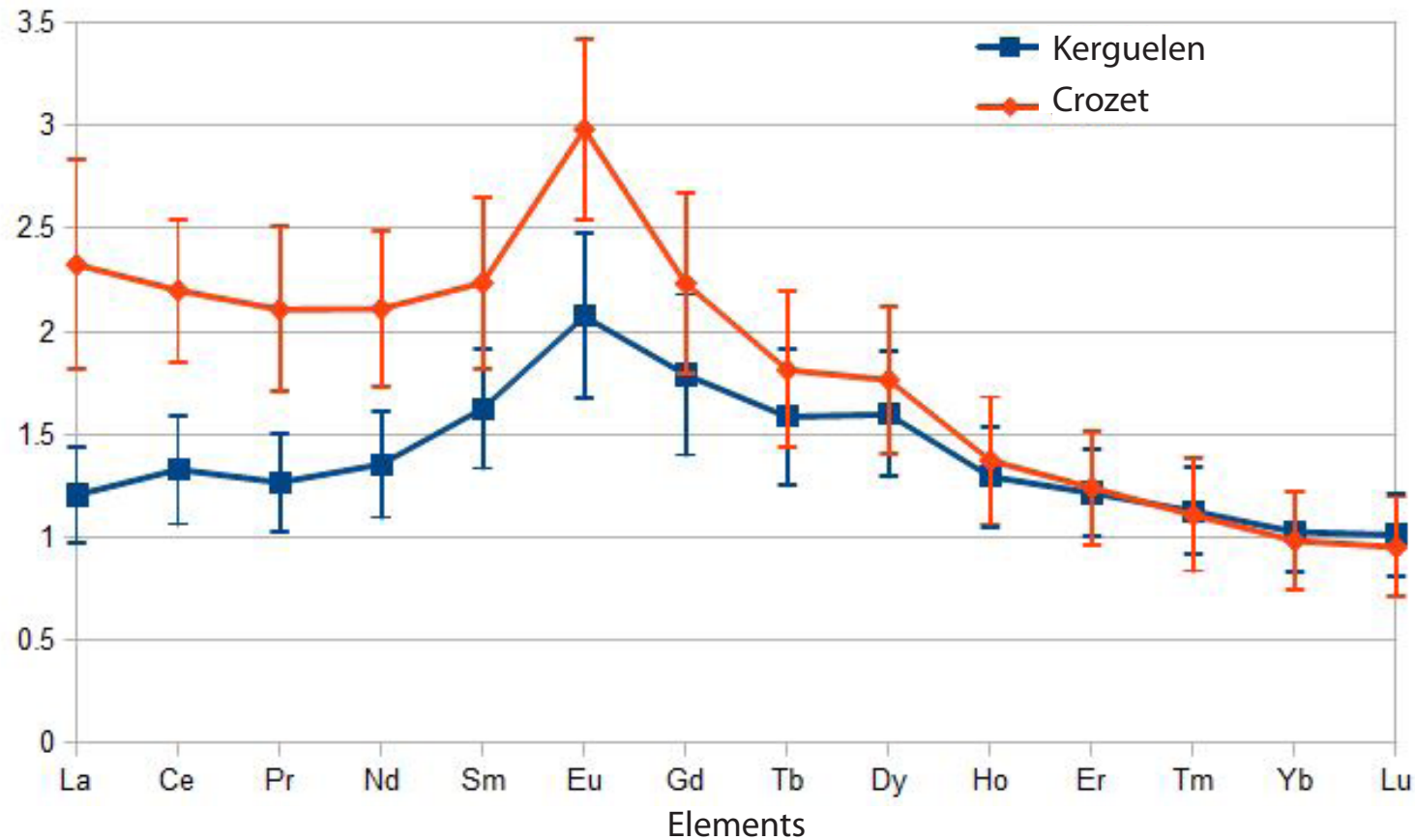
Anthropogenic enrichment for Pb (and As, Cu, Cr, V)
during the austral winter



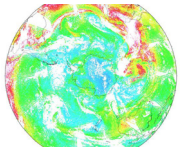
Rare earth elements

REE/UCC

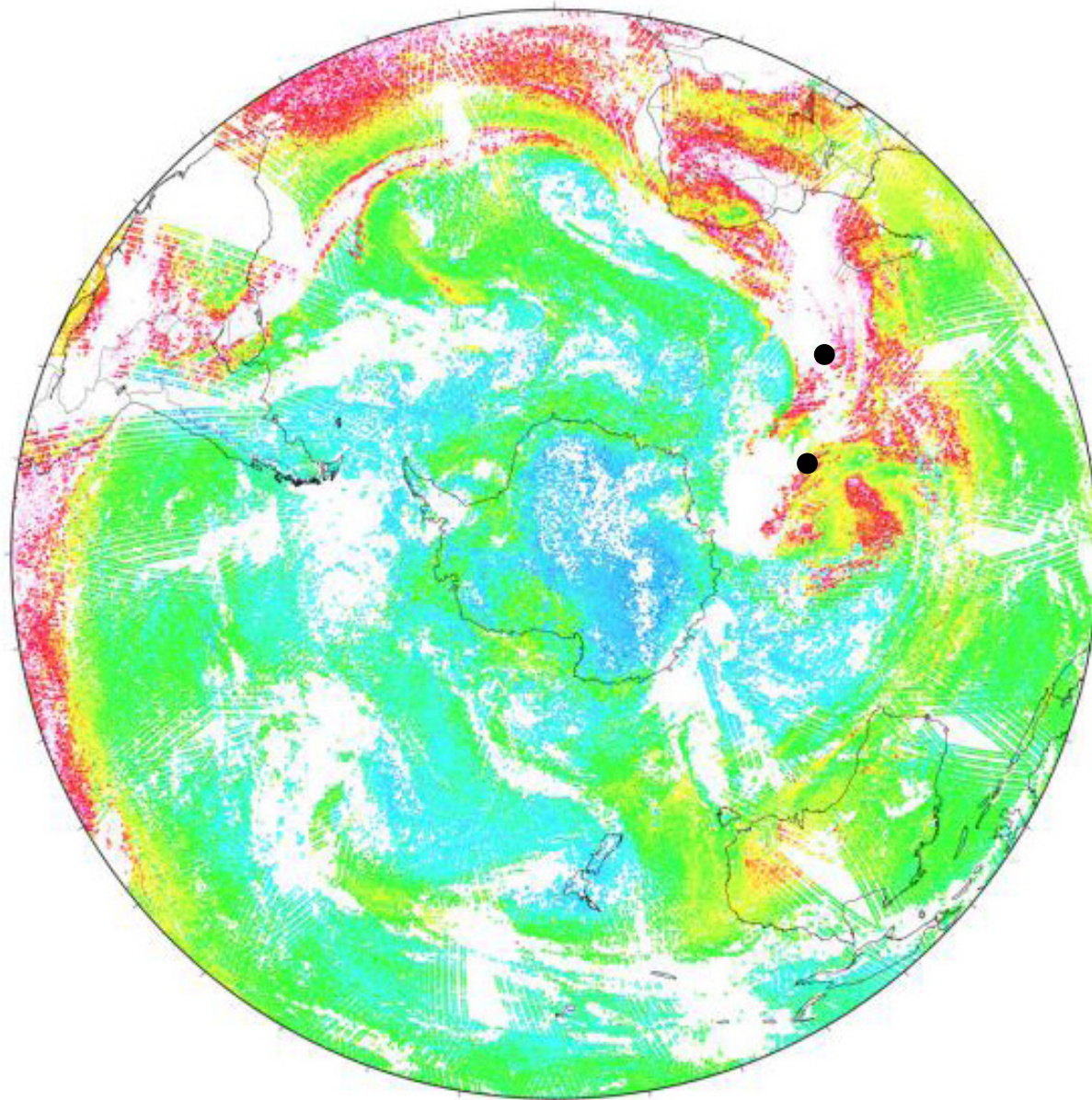
(Taylor and McLennan, 1985)



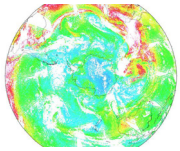
REE profiles are different between Kerguelen and Crozet suggesting that crustal source is not the same for both locations



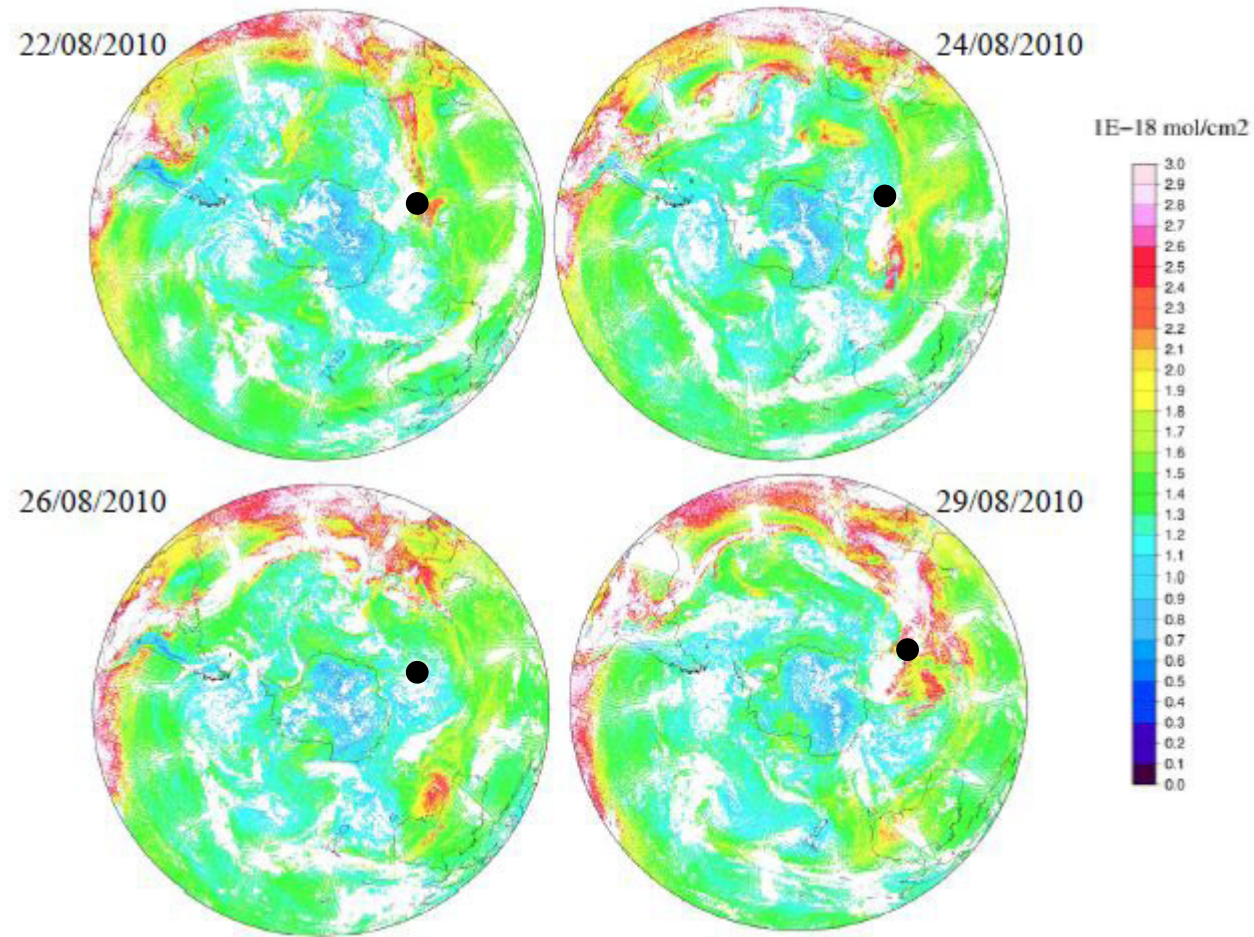
CO satellite images

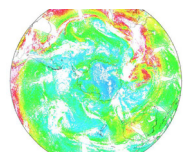


Images satellite CO, IASI

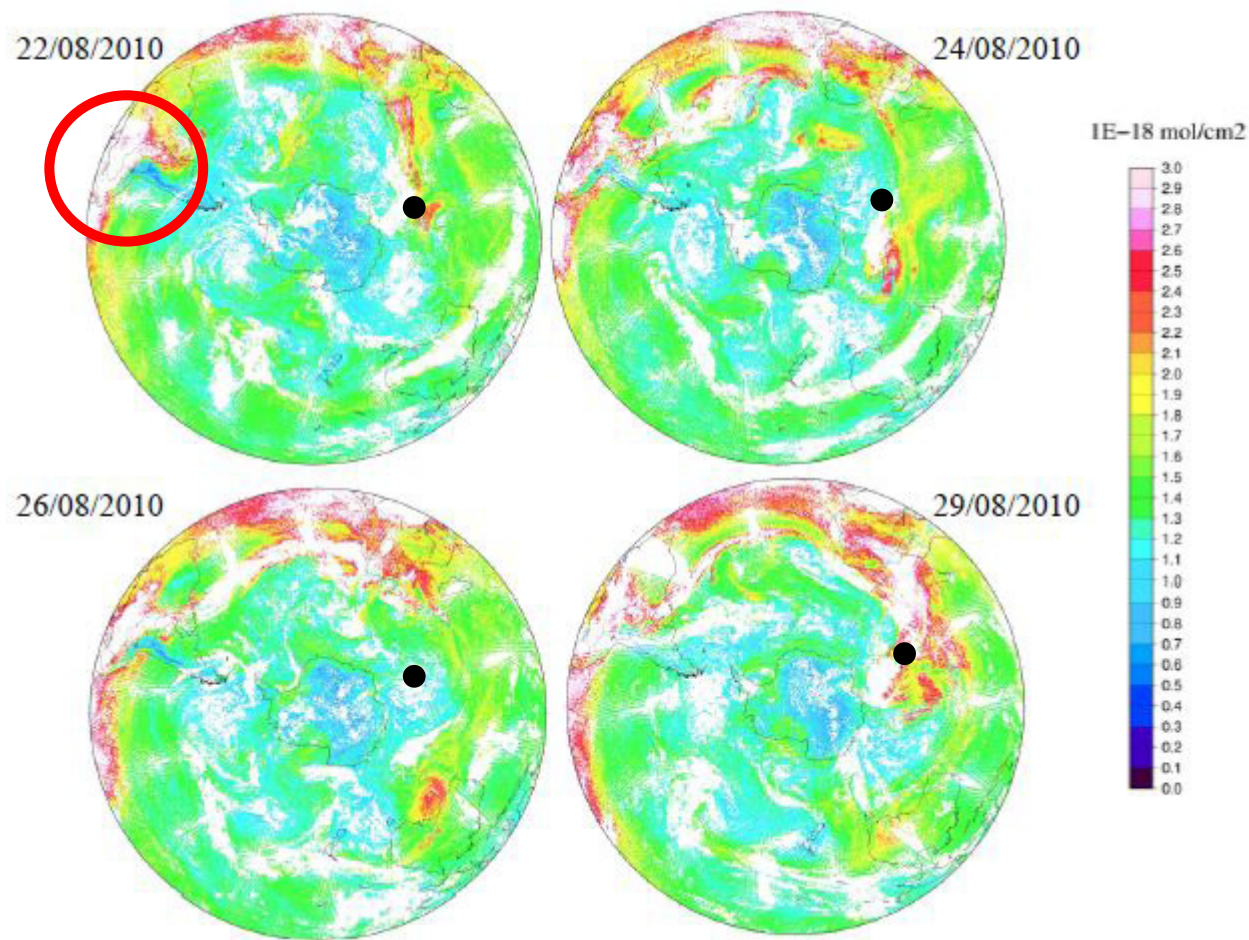


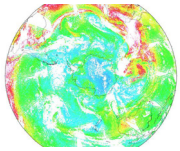
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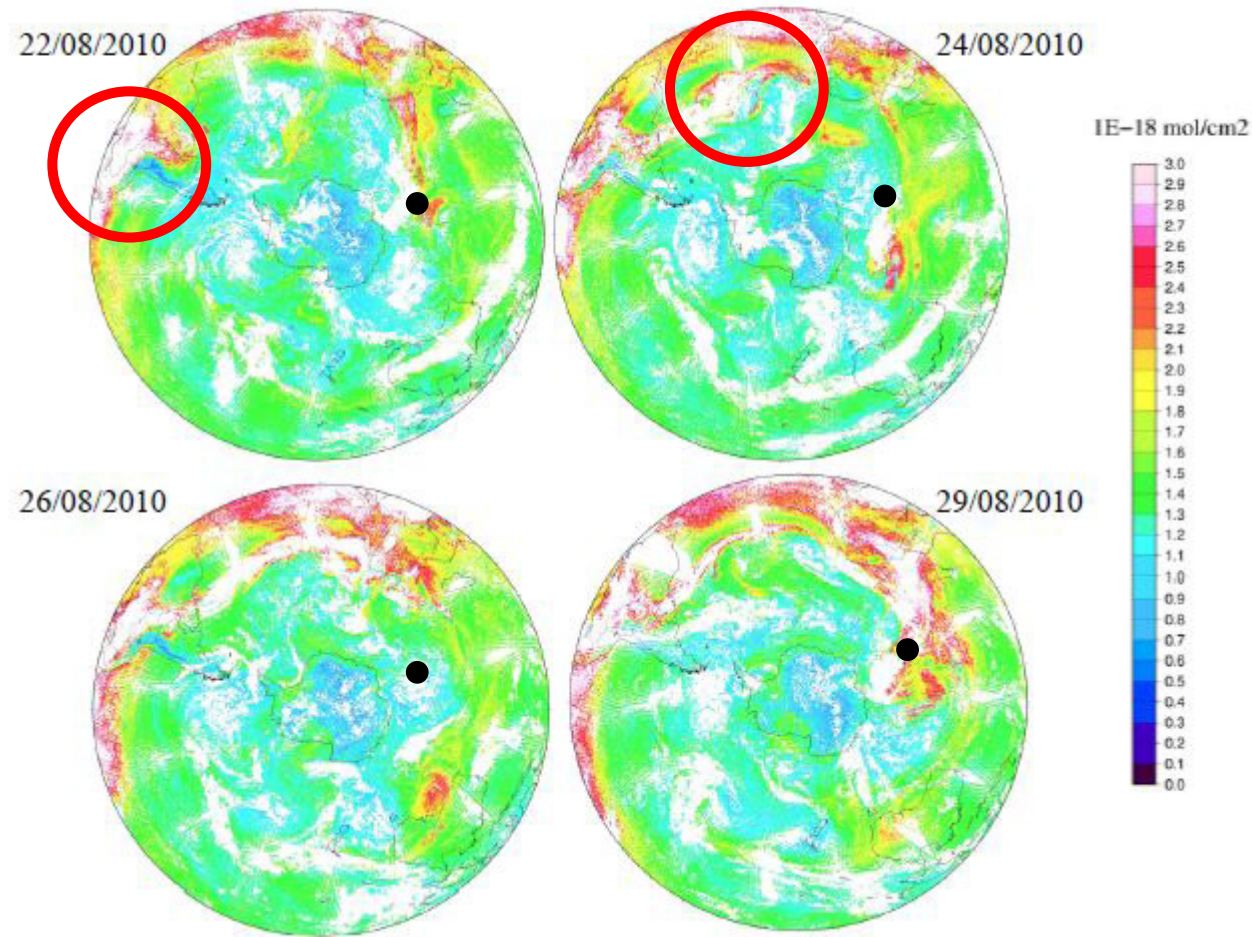


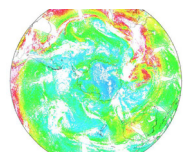
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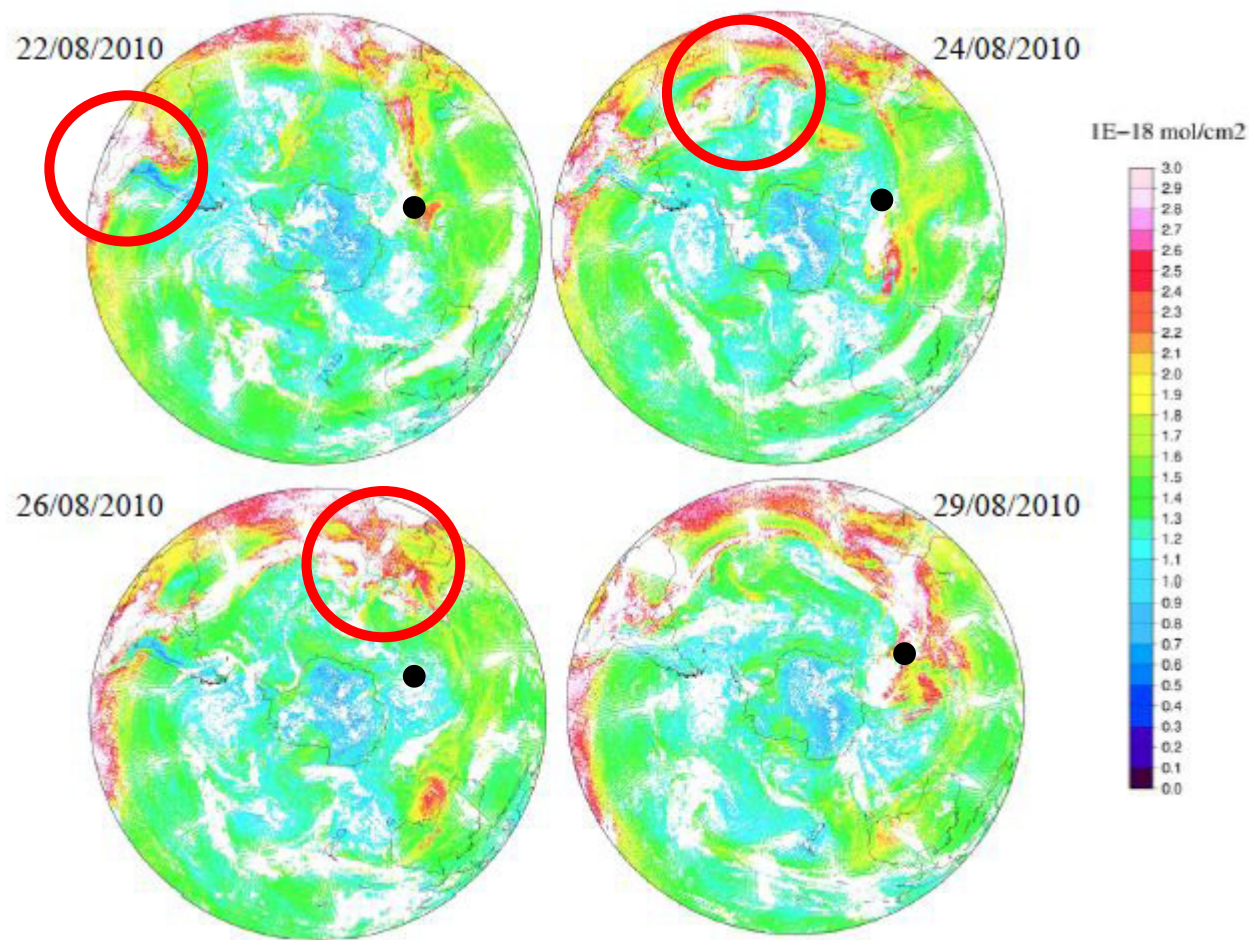


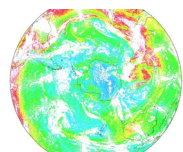
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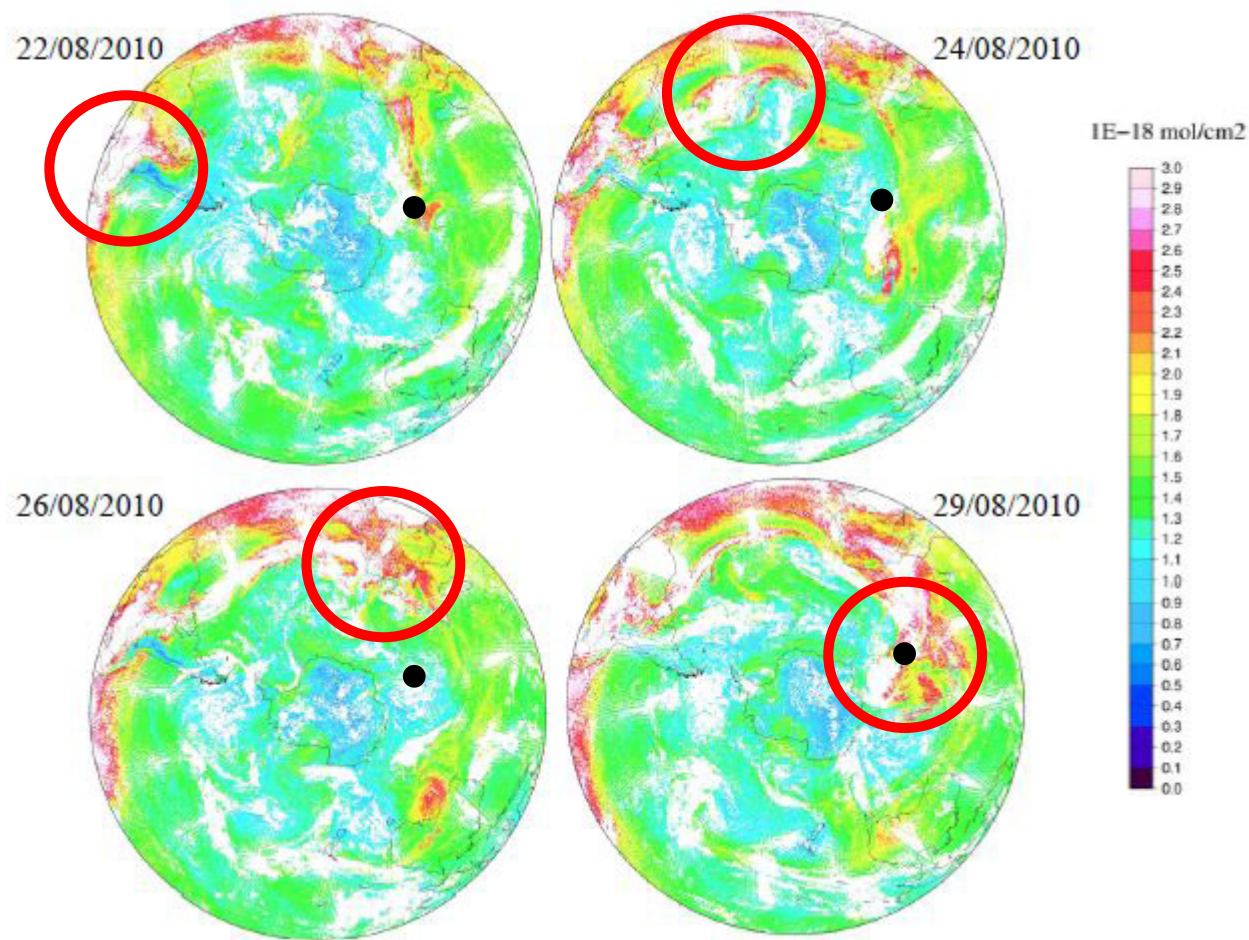


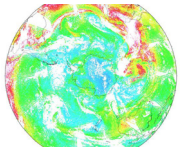
CO satellite images



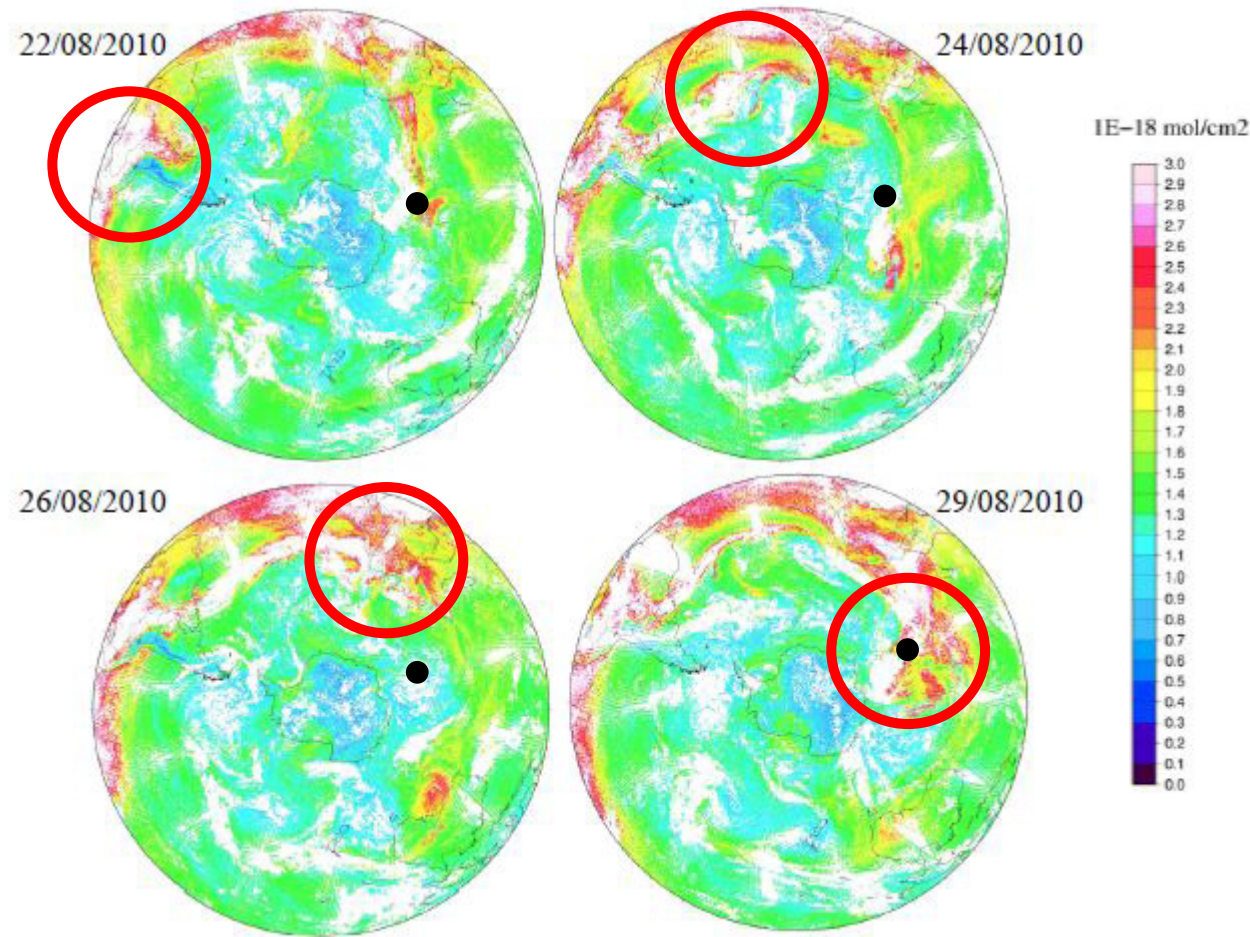


CO satellite images





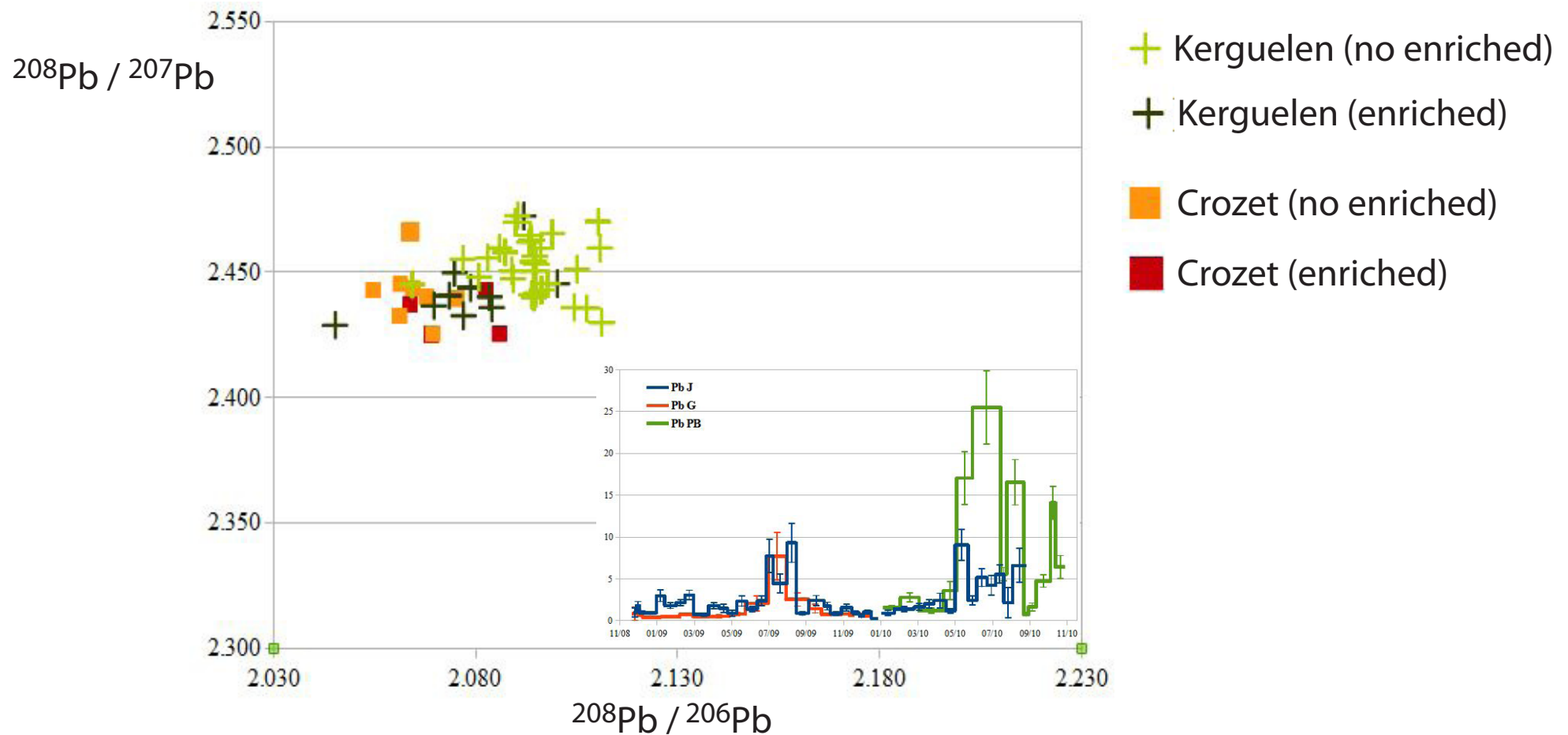
CO satellite images



On Kerguelen and Crozet Islands
air masses come from South America and South Africa

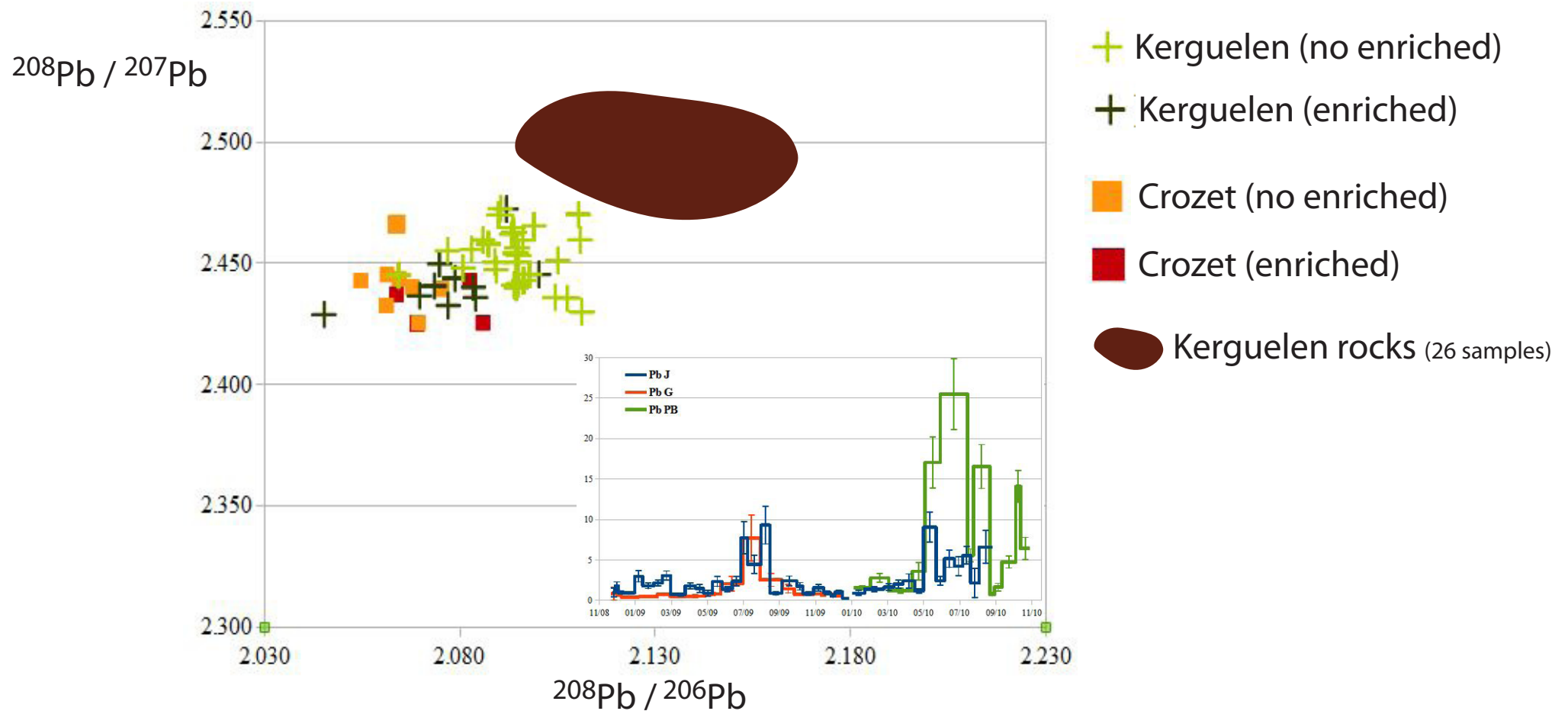


Pb isotopes





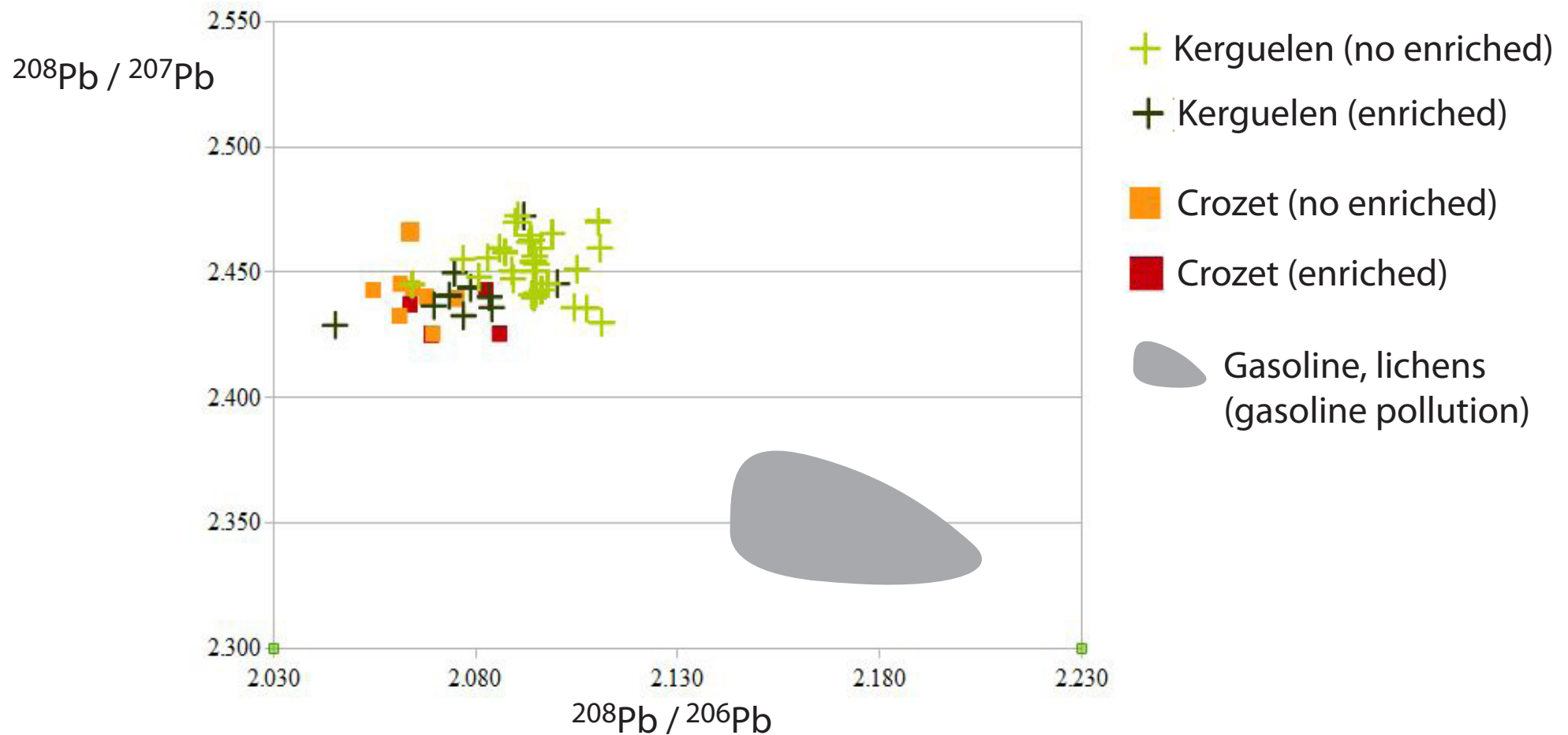
Pb isotopes: local soil contamination?



No correlation with Pb ratios in Kerguelen rocks



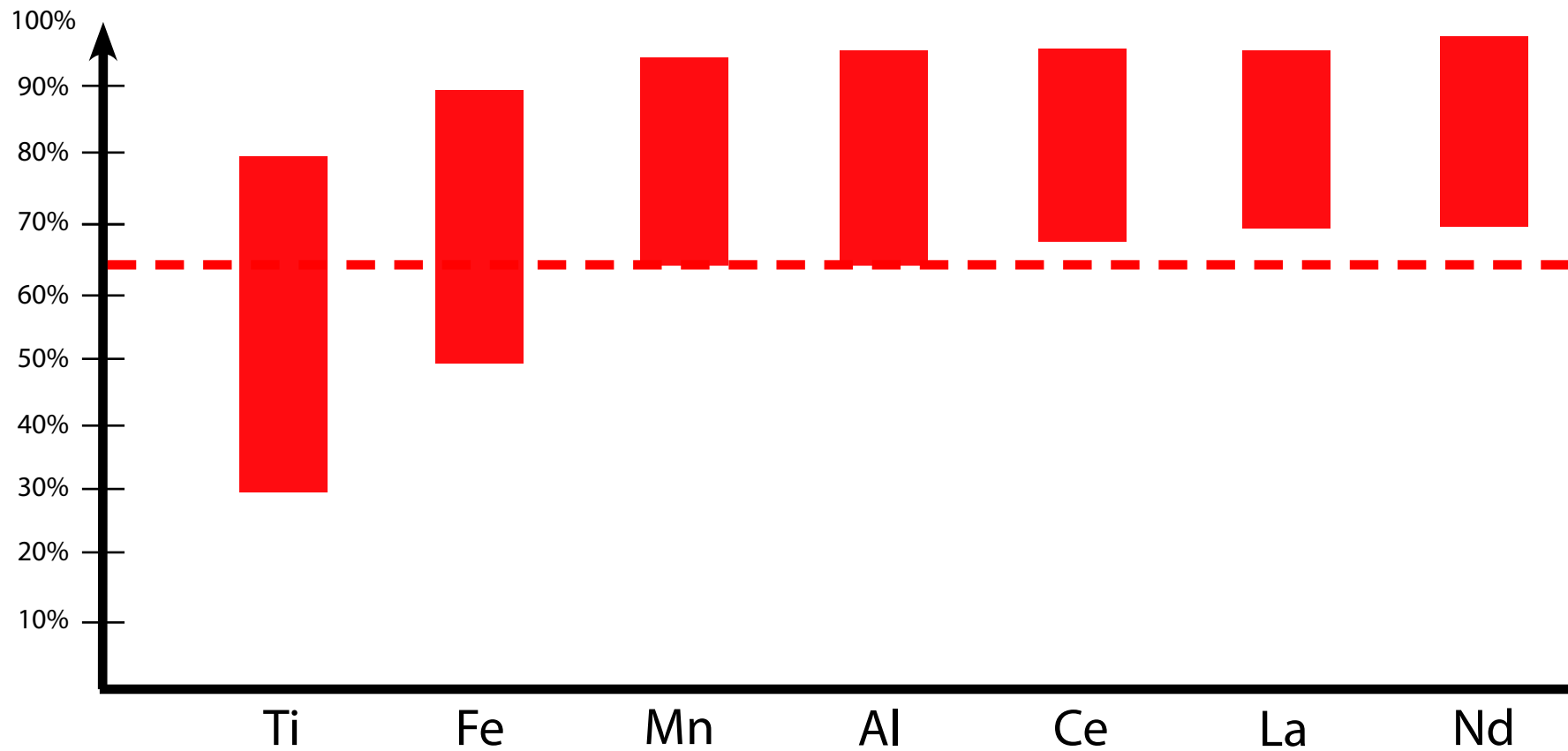
Pb isotopes: anthropogenic contamination?



No correlation with Pb ratios in gasoline and polluted lichens



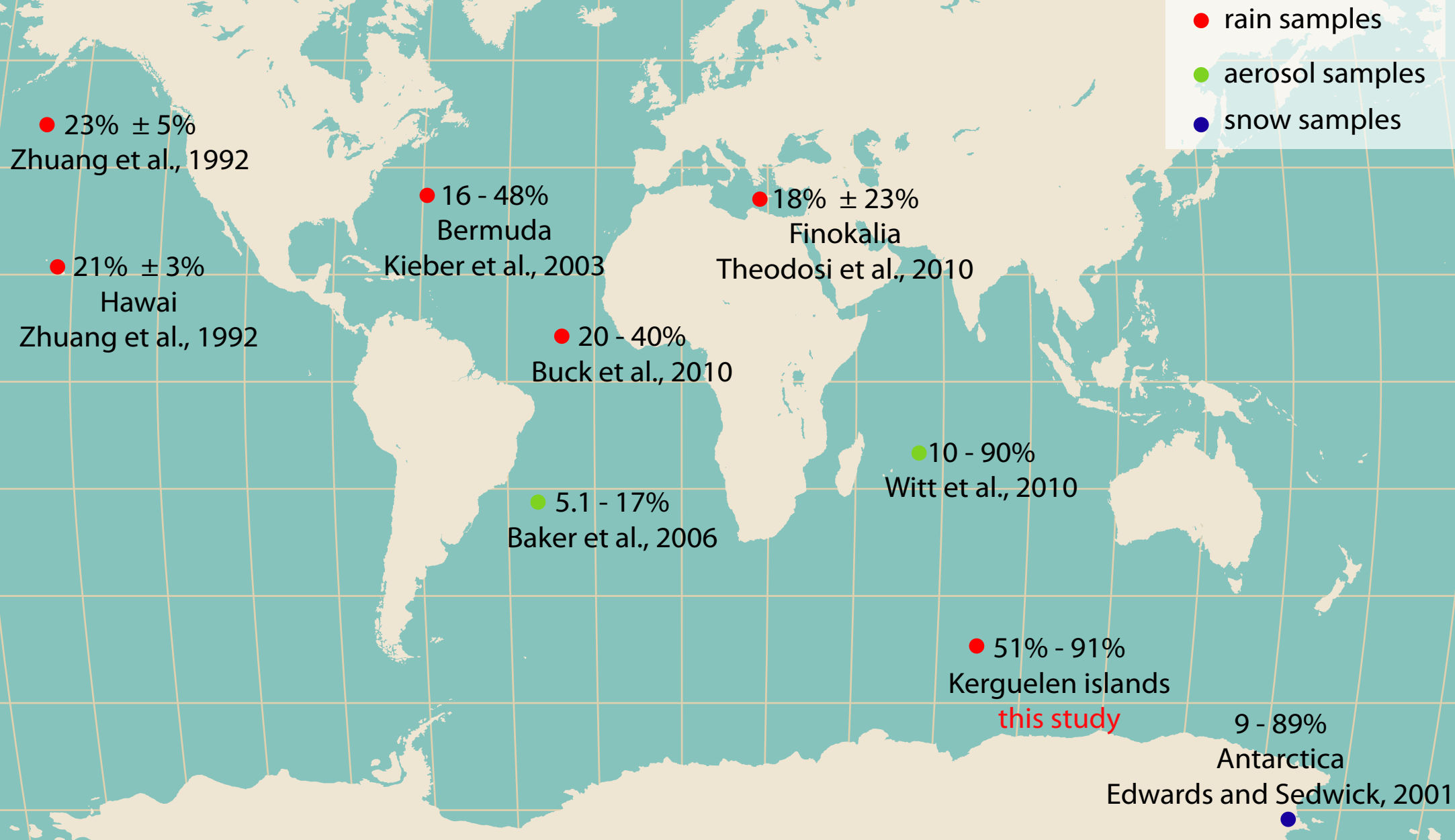
Solubility



High solubility
> 65%



Fe solubility in literature





Al solubility in literature

- rain samples
- aerosol samples
- snow samples

5 - 62%
● Bermuda
Lim et al., 1994

19% ± 26%
● Tour du Valat
Guieu et al., 2010

● 18 - 37%
Buck et al., 2010

● 7.8 - 51%
Backer et al., 2006

● 91% ± 66% (max, pH 1)
Witt et al., 2010

67% - 96%
● Kerguelen islands
this study



Mn solubility in literature

- rain samples
- aerosol samples
- snow samples

63% ± 28%
Tour du Valat
Guieu et al., 2010

7 - 88% ●
Buck et al., 2010

69% ± 26% ●
Finokalia
Theodosi et al., 2010

● 49% ± 12%
Hsu al., 2010

● 18 - 75%
Backer et al., 2006

66% - 94% ●
Kerguelen islands
this study

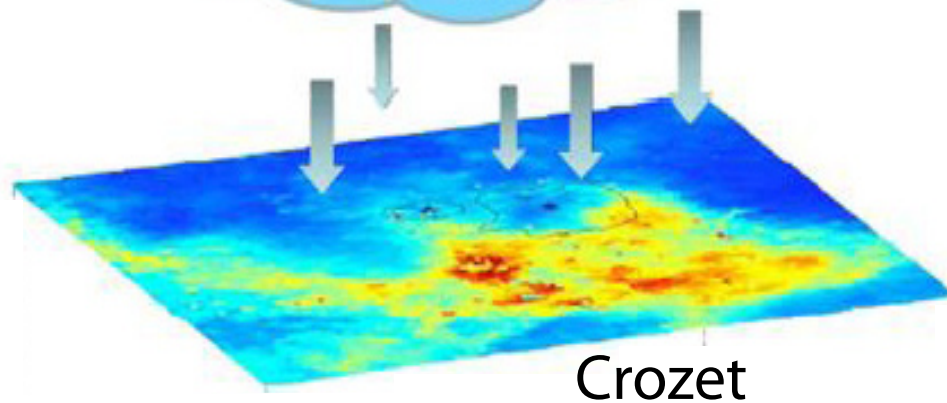


Dissolved Fe flux in literature

Observations

$23 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ($412 \text{ nmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)
with a solubility of 82%
(Heimburger et al., in review)

Total Fe flux = $50 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
 $28 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Heimburger et al., 2012)



Observed dissolved Fe deposition flux
using a solubility of 14% (wet deposition):
 $6 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
($100 \text{ nmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)
(Planquette et al., 2007)



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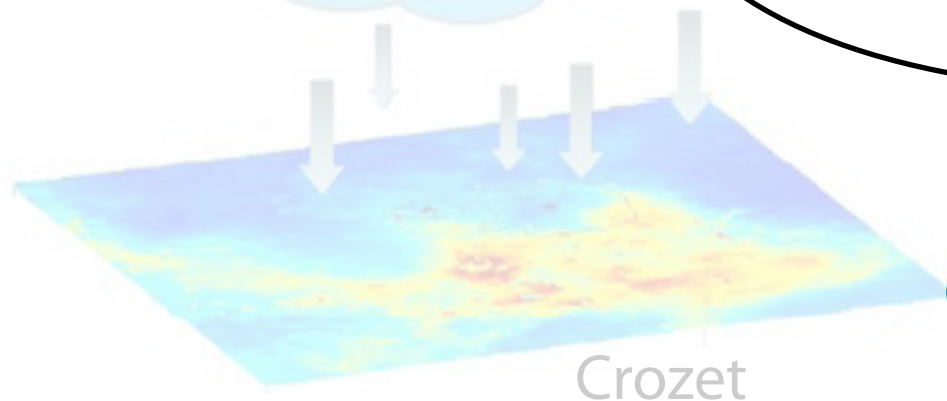
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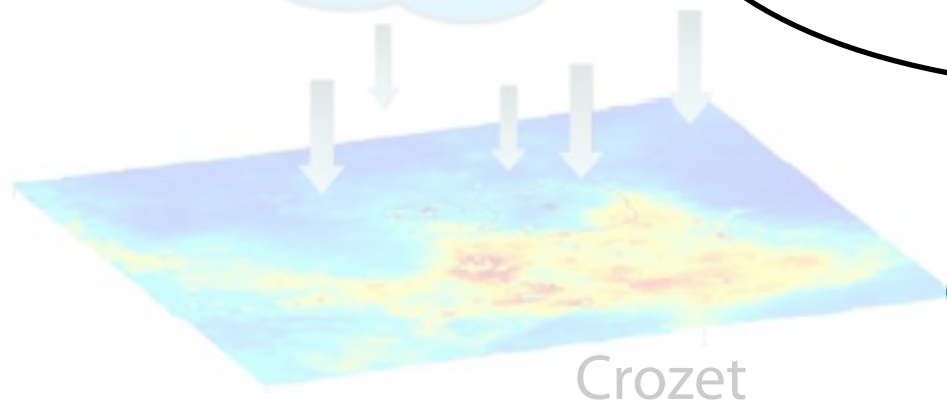
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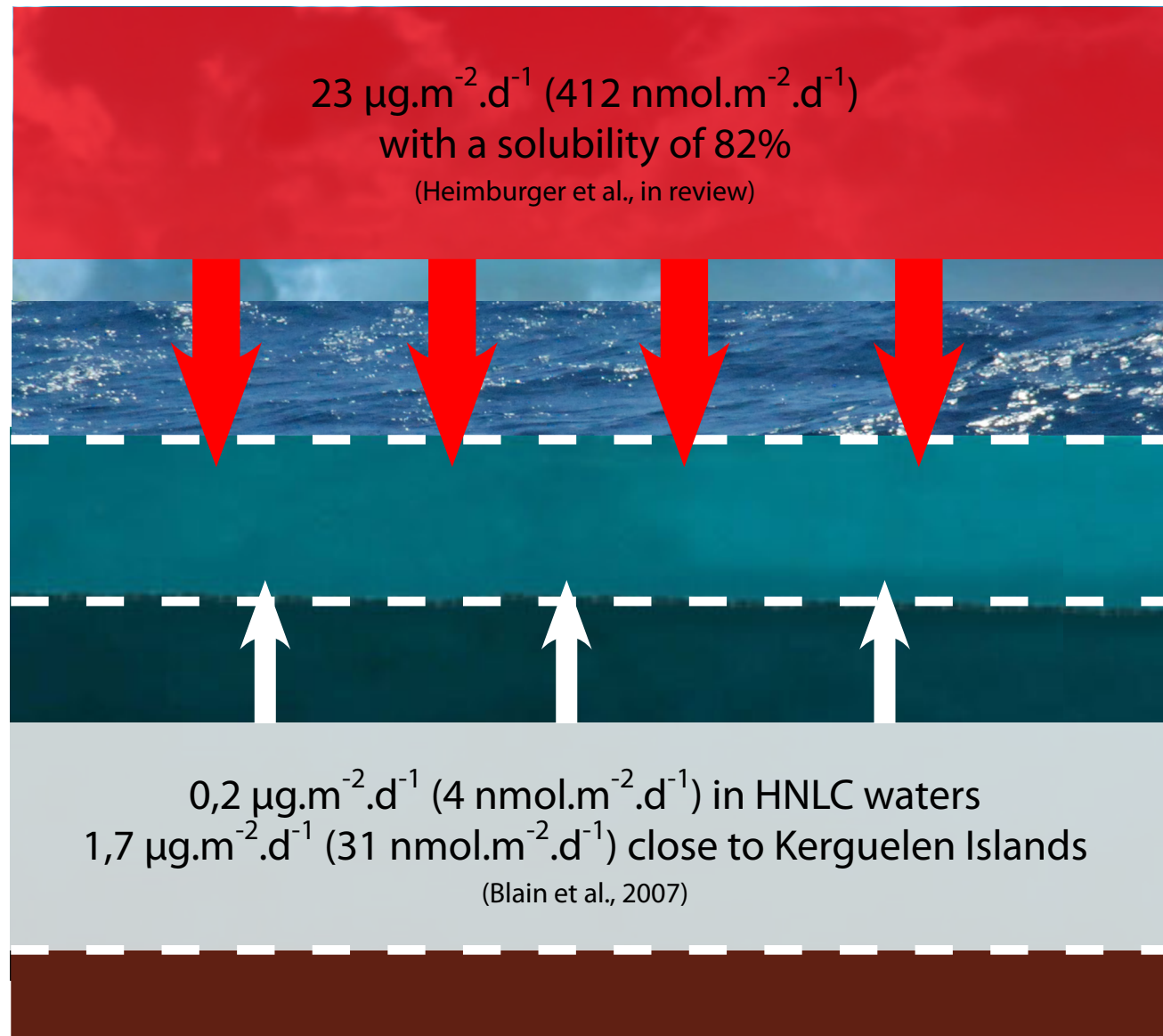
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Atmospheric vs. oceanic inputs



Publications

3 published articles as first author,

1 published article as second author,

1 article in review (first author),

1 article in prep (first author),

1 blog on my field campaign on Kerguelen Islands (on Paris 7 website),

1 short film (popularization of my thesis research),

9 communications (talk and poster) during international conferences.

Recommendations

1. Testing our hypothesis that aerosols collected at surface level do not allow total atmospheric deposition flux to be calculated:
 - a) sampling of aerosols in altitudes,
 - b) direct measurements of total atmospheric deposition coupled with aerosol measurements have to be performed over other remote oceanic areas in a continuous basis during one year at least.
2. Automatic samplers are needed to minimize human factors.
3. Studies on chemical composition (solubility, REE profiles, Pb isotopes) of aerosols emitted in area sources of the South Hemisphere (South America, South Africa).
Evolution of aerosols during their transport in the atmosphere.
4. Review the micronutrient budget in the South Indian Ocean with our results.

Islands in remote ocean

