High-resolution measurements and modeling of $\Delta^{14}CO_2$ over Western Europe

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The carbon isotopes – ^{12}C, ^{13}C, ^{14}C

- Isotope mixing ratios of CO_2 in the atmosphere
  - Stable ^{12}C isotope ~ 98.9%
  - Stable ^{13}C isotope ~ 1.1%
  - Radioactive ^{14}C isotope ~ 0.0000000001% (10^{-12})
- ^{14}C is reported usually as normalized difference with a reference standard
  - $\Delta^{14}C = (A_s/A_{ref} - 1)*Corrections*1000$ [‰]
  - Corrections for two major processes
    - Fractionation – using ^{13}C information
    - Radioactive decay in the reference since 1950

The unstable ^{14}C

- Destroyed by radioactive decay
  - ^{14}C half-life time = 5730±40 years
  - Well-researched for archaeological dating purposes
    - Effects are negligible for modern samples and measurable only in reservoirs with long turn-over time
- Production requires high energy particles
  - Natural by cosmic radiation in the upper atmosphere

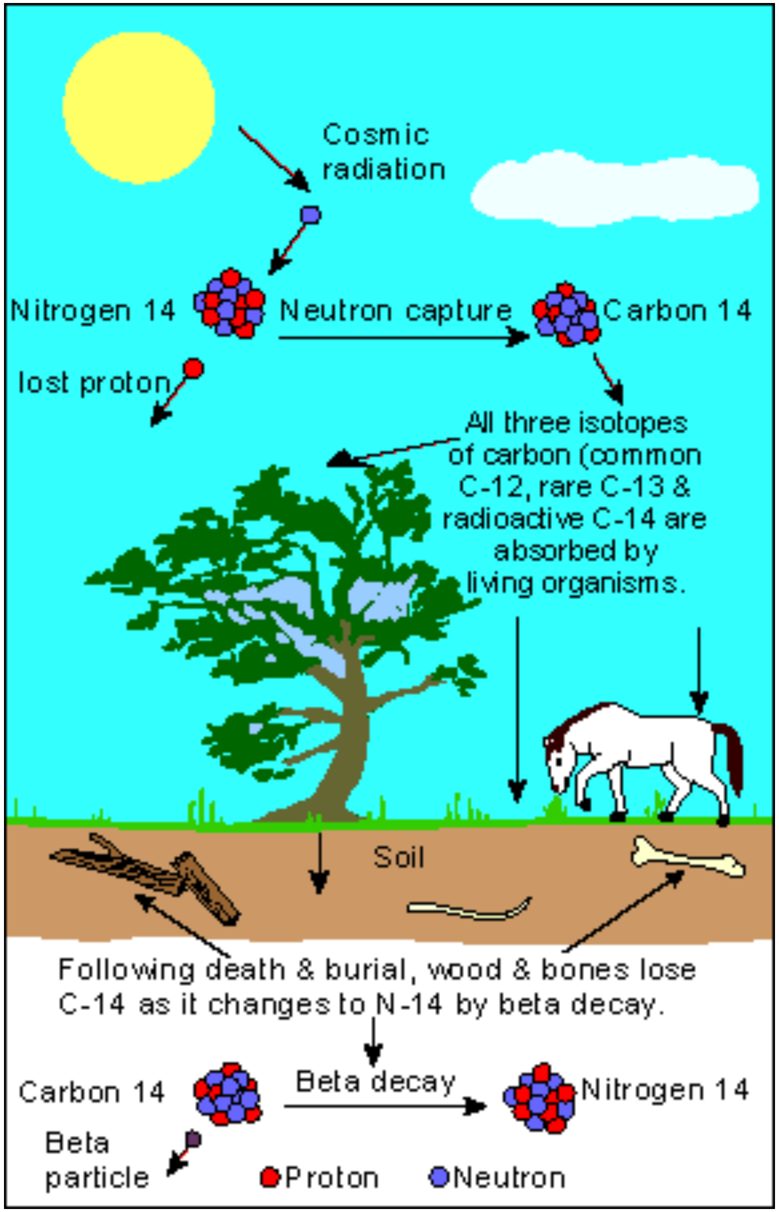

Figure source: Virtual Courseware Project - ScienceCourseware.org

The unstable ^{14}C

- Destroyed by radioactive decay
  - ^{14}C half-life time = 5730±40 years
  - Well-researched for archaeological dating purposes
    - Effects are negligible for modern samples and measurable only in reservoirs with long turn-over time
    - Fossil fuels are old enough to have no ^{14}C left
- Production requires high energy particles
  - Natural by cosmic radiation in the upper atmosphere
  - Manmade by atmospheric nuclear bomb tests (mostly before 1963)
  - Manmade by nuclear power industry

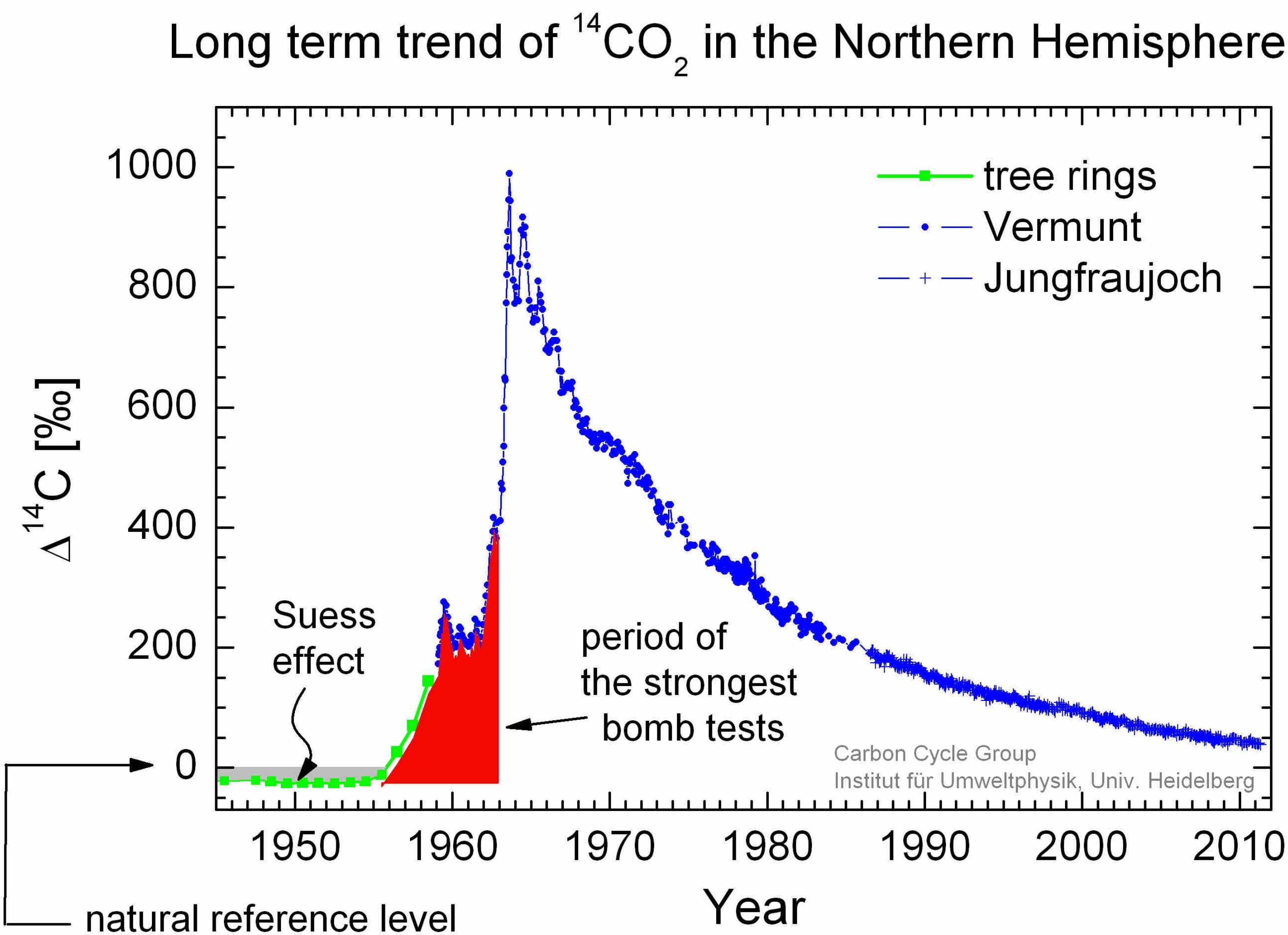

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Figure source: Institute for Environmental Physics, University of Heidelberg, 2011

(http://www.iup.uni-heidelberg.de/institut/forschung/groups/kk/en/14CO2_html)

The budget of $^{14}CO_2$

- $d\Delta_{atm}C_{atm}/dt = \Delta_{fossil}C_{fossil} +$

    - $\Delta_{fossil} = -1000$ ‰ – no $^{14}CO_2$ in the flux

The budget of $^{14}CO_2$

- $d\Delta_{atm}C_{atm}/dt = \Delta_{fossil}C_{fossil} + \Delta_{14}{}^{14}C_{cosmic} + \Delta_{14}{}^{14}C_{nuclear}$

  - Δ_{fossil} = -1000 ‰ – no $^{14}CO_2$ in the flux
  - Δ_{14} ~ +7.5x10^{15} ‰ – pure $^{14}CO_2$ fluxes

The budget of $^{14}CO_2$

- $d\Delta_{atm}C_{atm}/dt = \Delta_{fossil}C_{fossil} + \Delta_{14}{}^{14}C_{cosmic} + \Delta_{14}{}^{14}C_{nuclear}$

  $+ \Delta^{bio}{}_{diseq}(C_{landuse}+C_{bio}{}^{net}) + \Delta^{ocean}{}_{diseq}C_{ocean}{}^{net}$

  - Δ_{fossil} = -1000 ‰ – no $^{14}CO_2$ in the flux
  - $\Delta_{14} \sim +7.5x10^{15}$ ‰ – pure $^{14}CO_2$ fluxes
  - Disequilibrium terms induced by the bomb-peak curve represent the difference in the Δ signature of the incoming and outgoing flux for the reservoirs

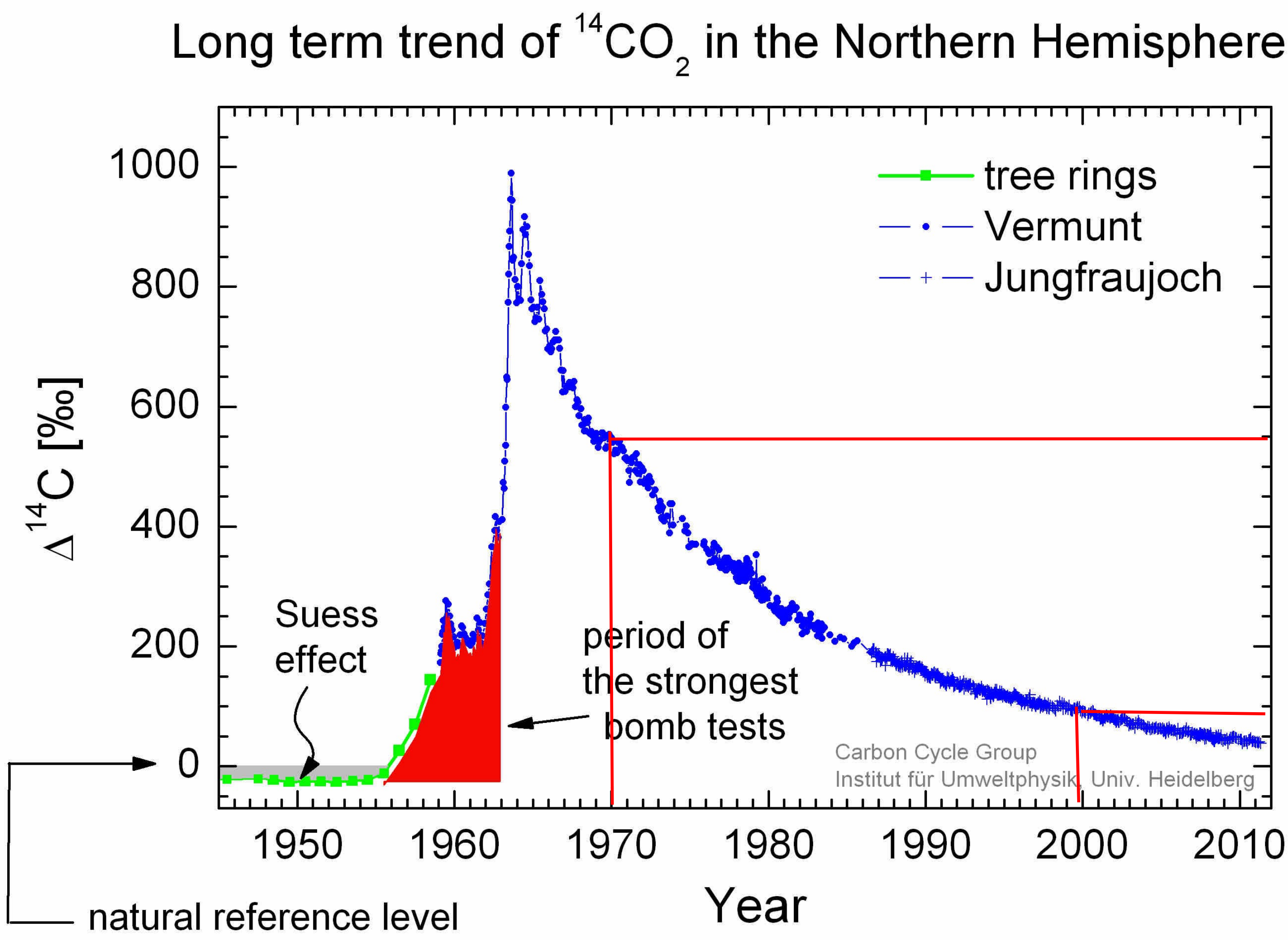

Figure source: Institute for Environmental Physics, University of Heidelberg, 2011

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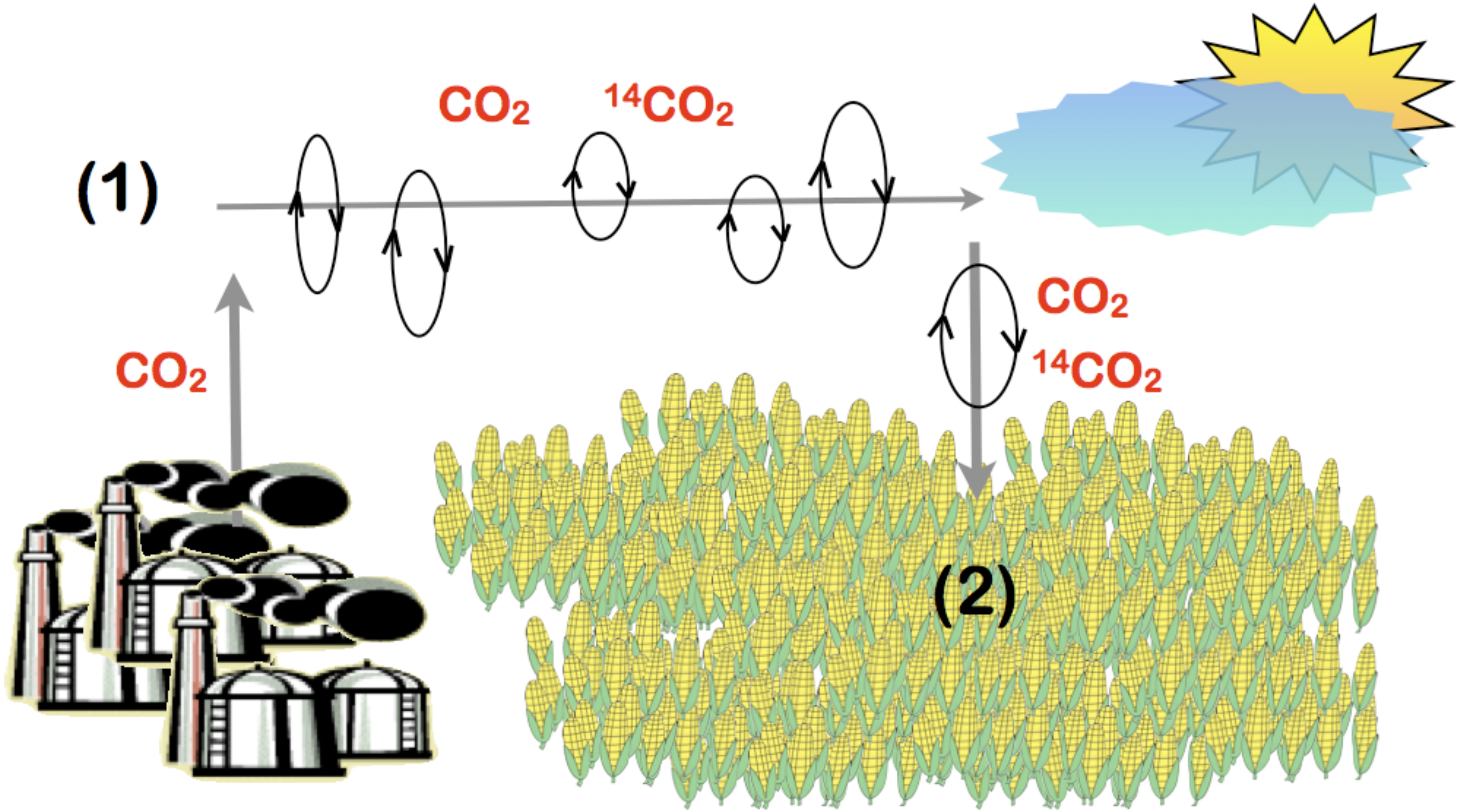
Project: ^{14}C in crops in Western Europe



PhD project Denica Bozhinova

Fixation of 'old' (fossil) CO_2 in maize (corn)

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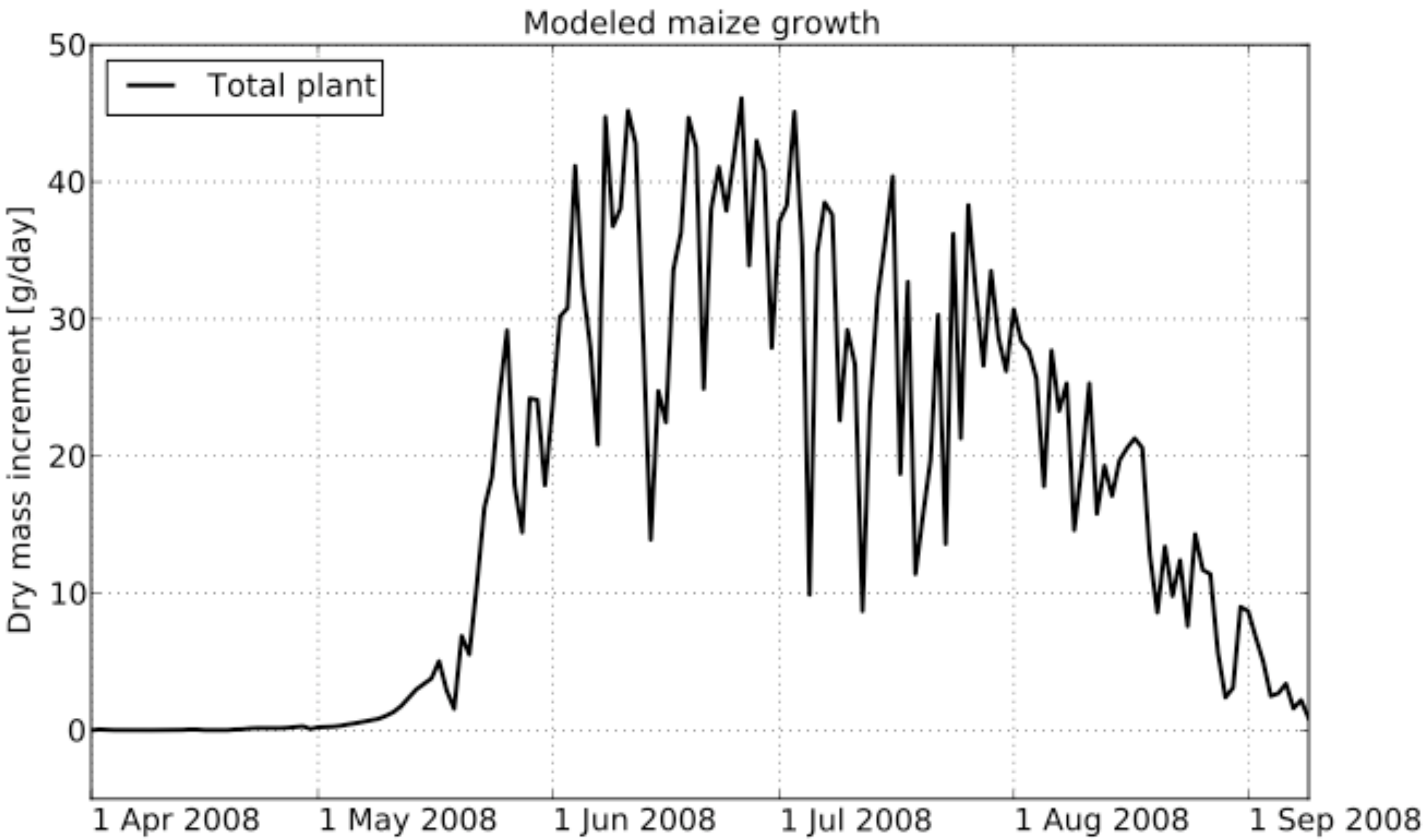
Our project and methods

- Can we use plant samples in a quantitative manner?
  - Model the plant CO_2 assimilation with crop growth model – SUCROS2
  - Model the atmospheric CO_2 concentrations (C_{atm}) and their $\Delta^{14}CO_2$ signature (Δ_{atm}) – WRF-Chem
  - Gather plant samples to verify the method

The crop growth model

- A simple and universal crop growth simulator - SUCROS
  - Mechanistic process-based LUE model
  - Includes water limited plant growth (SUCROS2)
  - Requires basic meteorological information
  - Time step of 1 day
  - Models the allocation of mass in the different plant parts
  - Simulates own development and leaf area index



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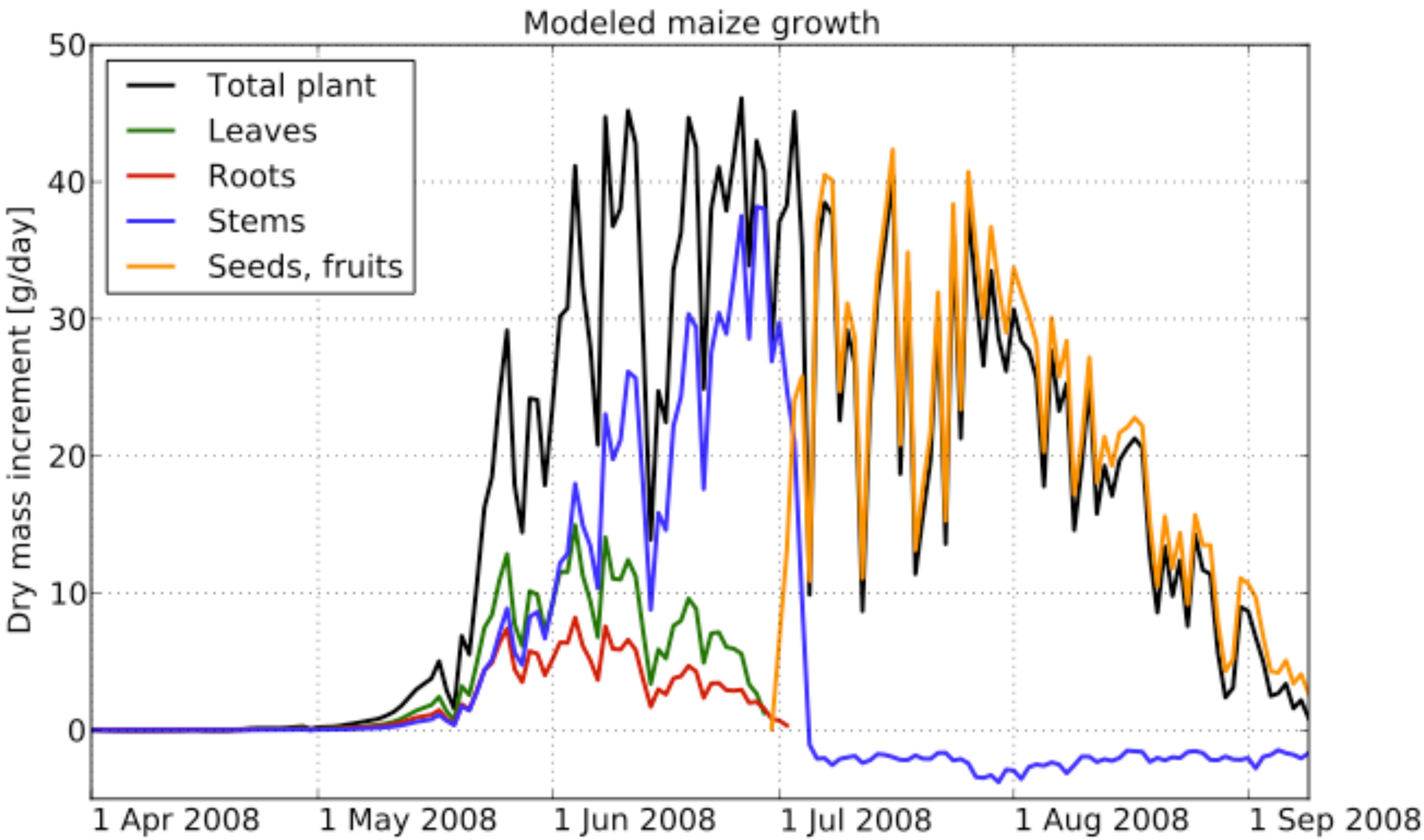

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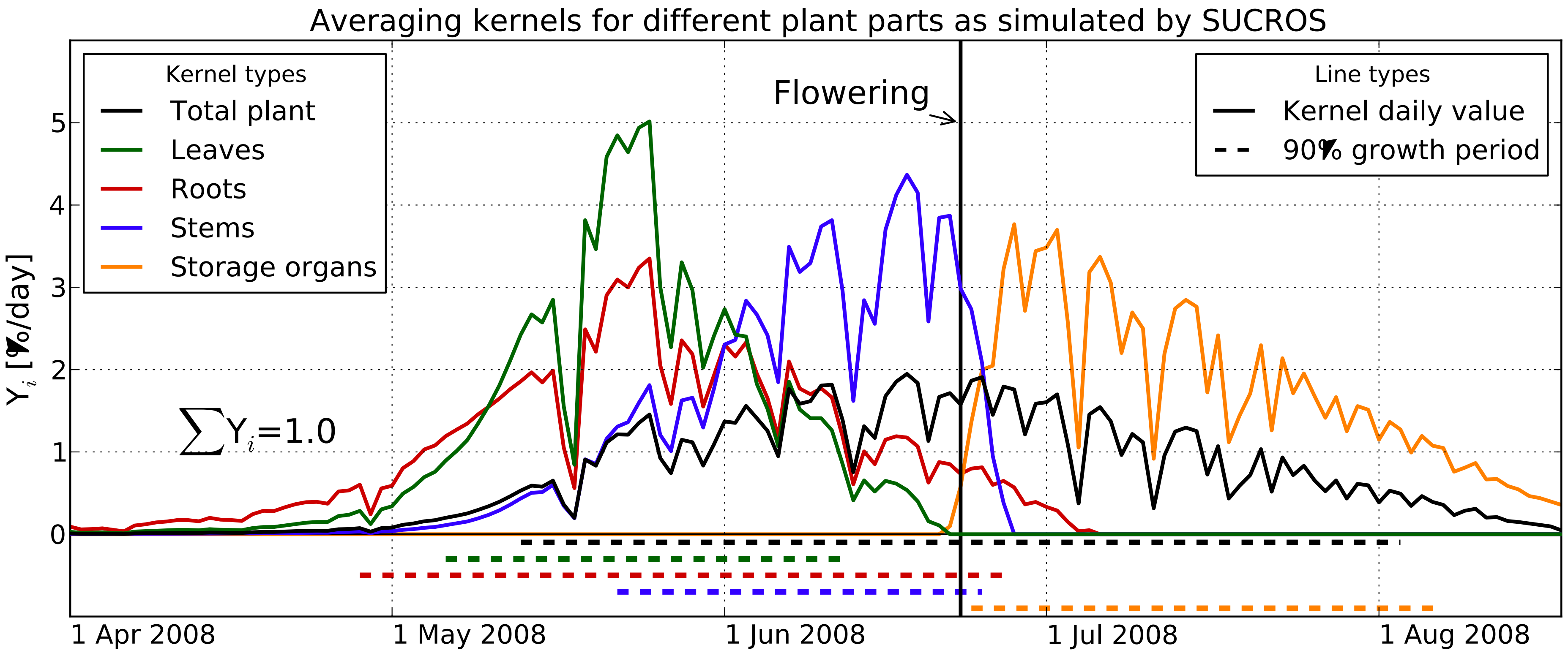
Averaging kernel

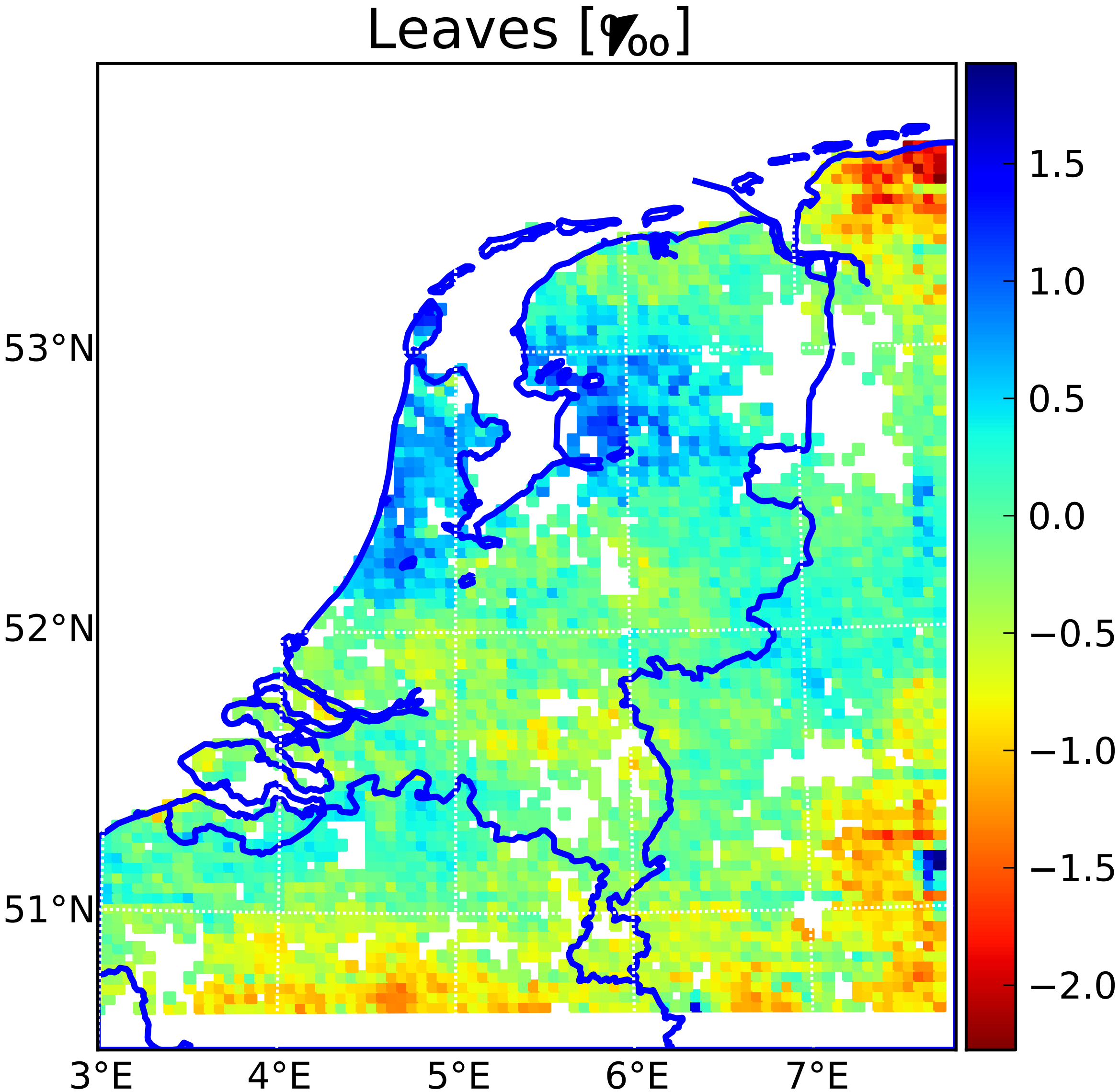


Averaging kernel: matters !

$\Delta^{14}C_{res}$[‰]	Spring wheat		Maize	
	flowering	maturity	flowering	maturity
Leaves	39.0	39.0	39.6	39.6
Roots	38.6	38.8	39.6	39.5
Stems	41.0	41.3	42.6	41.7
St. organs	-	40.6	-	38.1
Total plant	40.5	40.8	41.4	39.5
Flat-kernel	37.8	38.3	38.4	38.9
Radiation	37.6	38.0	38.4	38.4
Temperature	37.6	37.8	38.3	38.5

Same weather
Same atm Δ14C
Different growth period

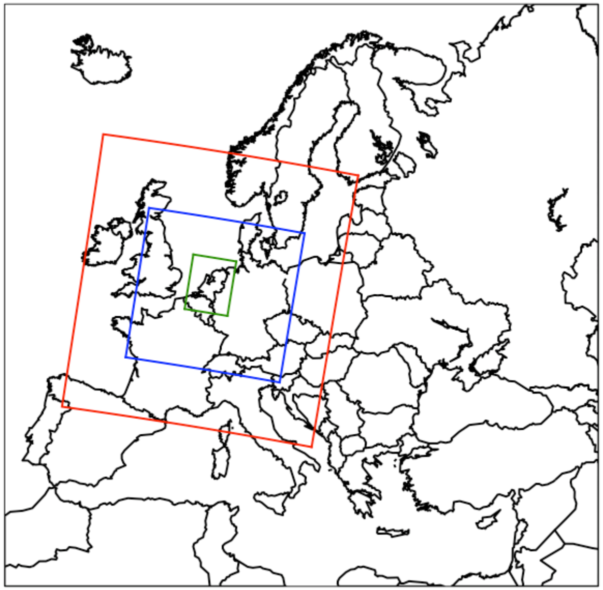
Averaging kernel: matters !



Same growth period
Same atm Δ14C
Different weather

WRF-Chem version 3.2.1

- 3 nested domains with horizontal resolution of 36, 12 and 4 km
- 27 vertical eta-levels, with 18 in the lower 2 km of the troposphere
- Time step of 180s in the mother domain, hourly output
- 6 hourly meteorological boundary conditions – NCEP FNL model
- MYNN 2.5 boundary layer and surface layer scheme
- Noah Unified Land-Surface Model
- 6 hourly update of surface conditions
- 2 days of model spin-up
- Period simulated 30-03-2008 – 01-10-2008



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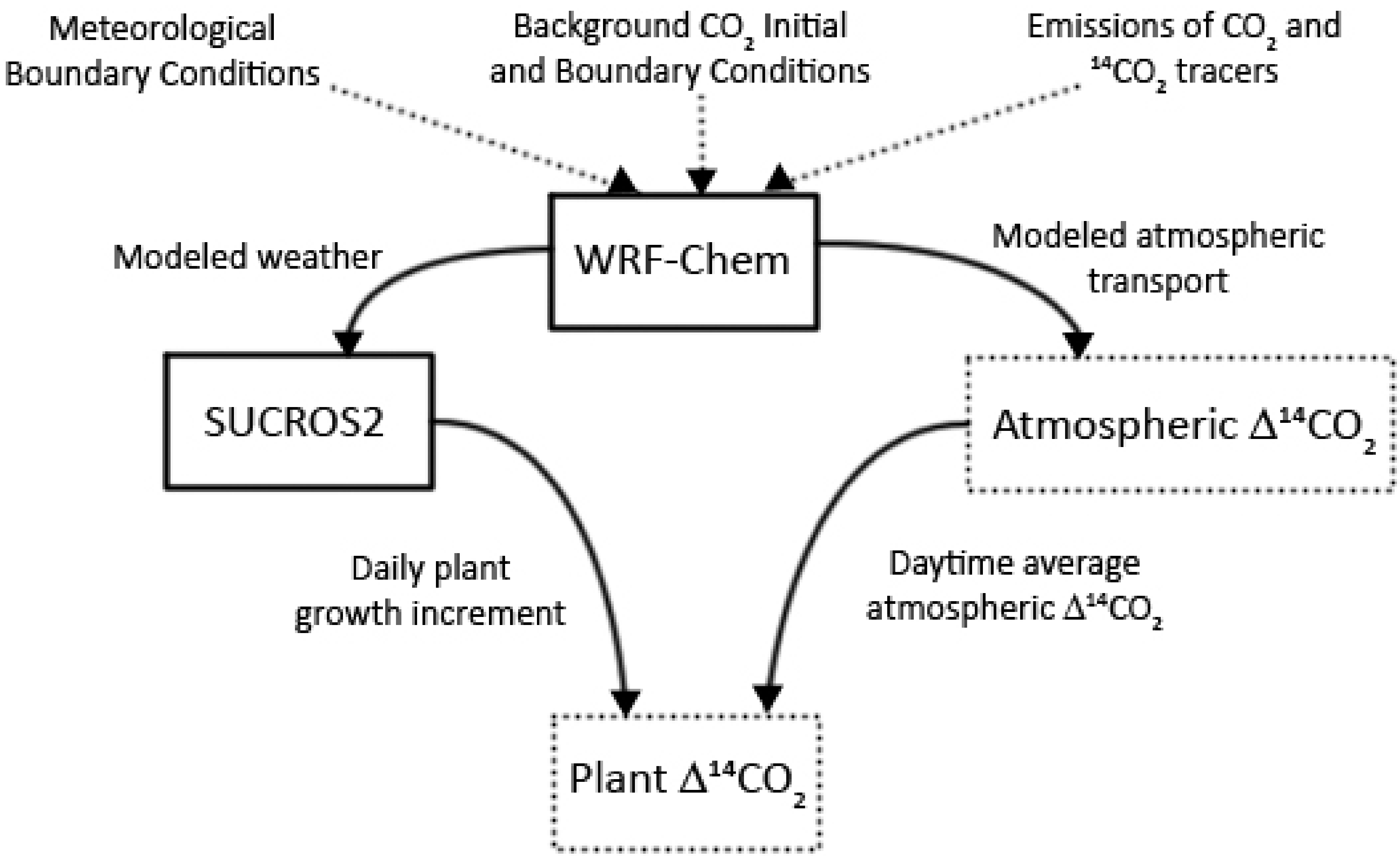
WRF-Chem - SUCROS



WRF-Chem with CO_2 tracers

$$CO_{2obs} = CO_{2bg} + CO_{2ff} + CO_{2p} + CO_{2r}$$

$$\begin{aligned} \Delta_{obs} CO_{2obs} &= \Delta_{bg}(CO_{2bg} + CO_{2p} + CO_{2r}) + \Delta_{ff} CO_{2ff} + \\ &+ {}^{14}\Delta({}^{14}CO_{2bio}^{dis} + {}^{14}CO_{2o}^{dis} + {}^{14}CO_{2n} + {}^{14}CO_{2c}) \end{aligned}$$

- Background CO_2 concentrations from CarbonTracker
- Biospheric CO_2 fluxes from SiBCASA
- Δ_{bg} from Jungfraujoch observatory (courtesy I. Levin)
- Fossil fuel CO_2 emissions from CarboEurope (IER, Stuttgart)
- Nuclear $^{14}CO_2$ emissions (IAEA, Graven and Gruber [2011])
- disequilibrium fluxes and stratospheric production from Miller et al., 2012

- The $\Delta^{14}CO_2$ signatures are calculated offline

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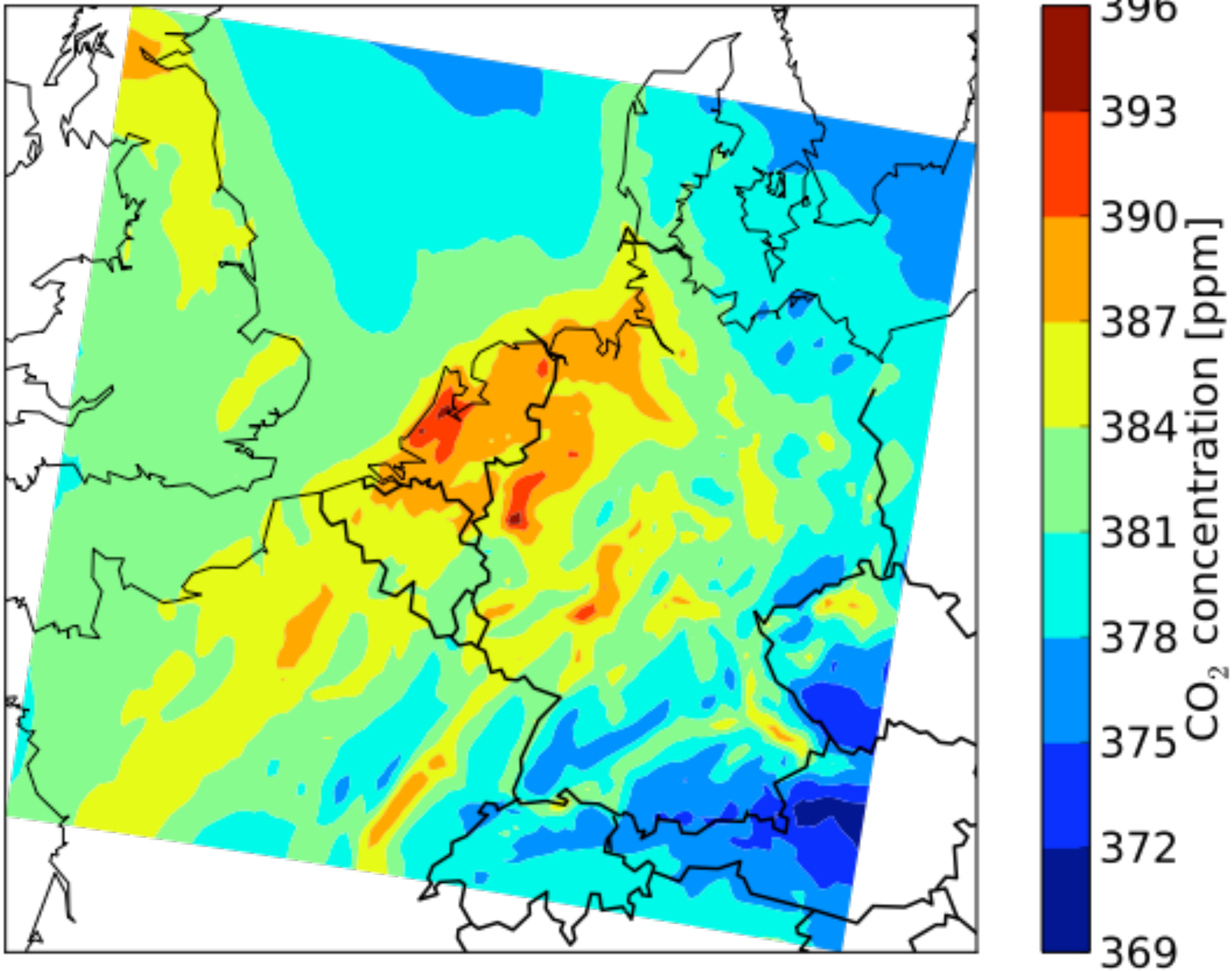

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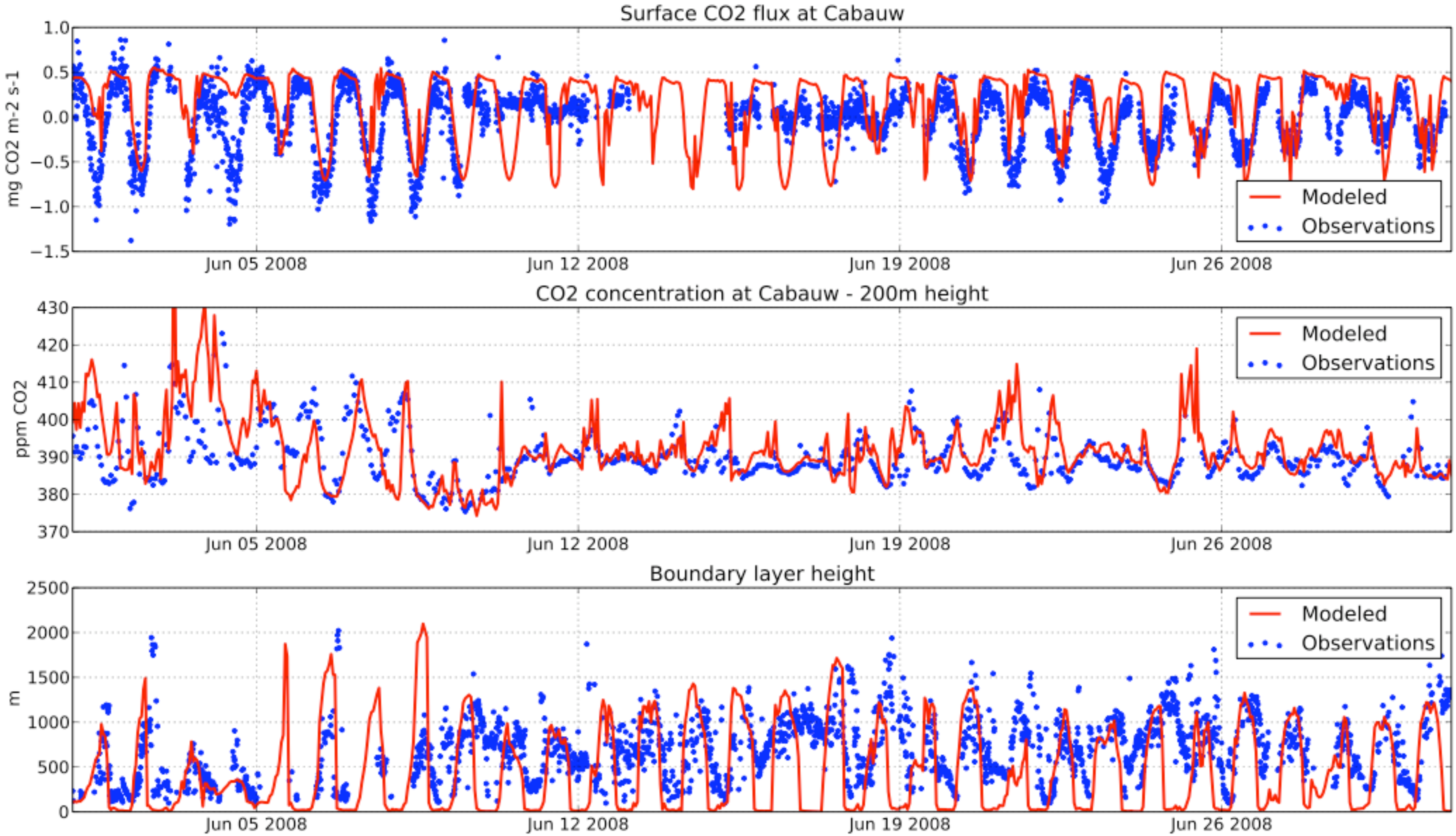

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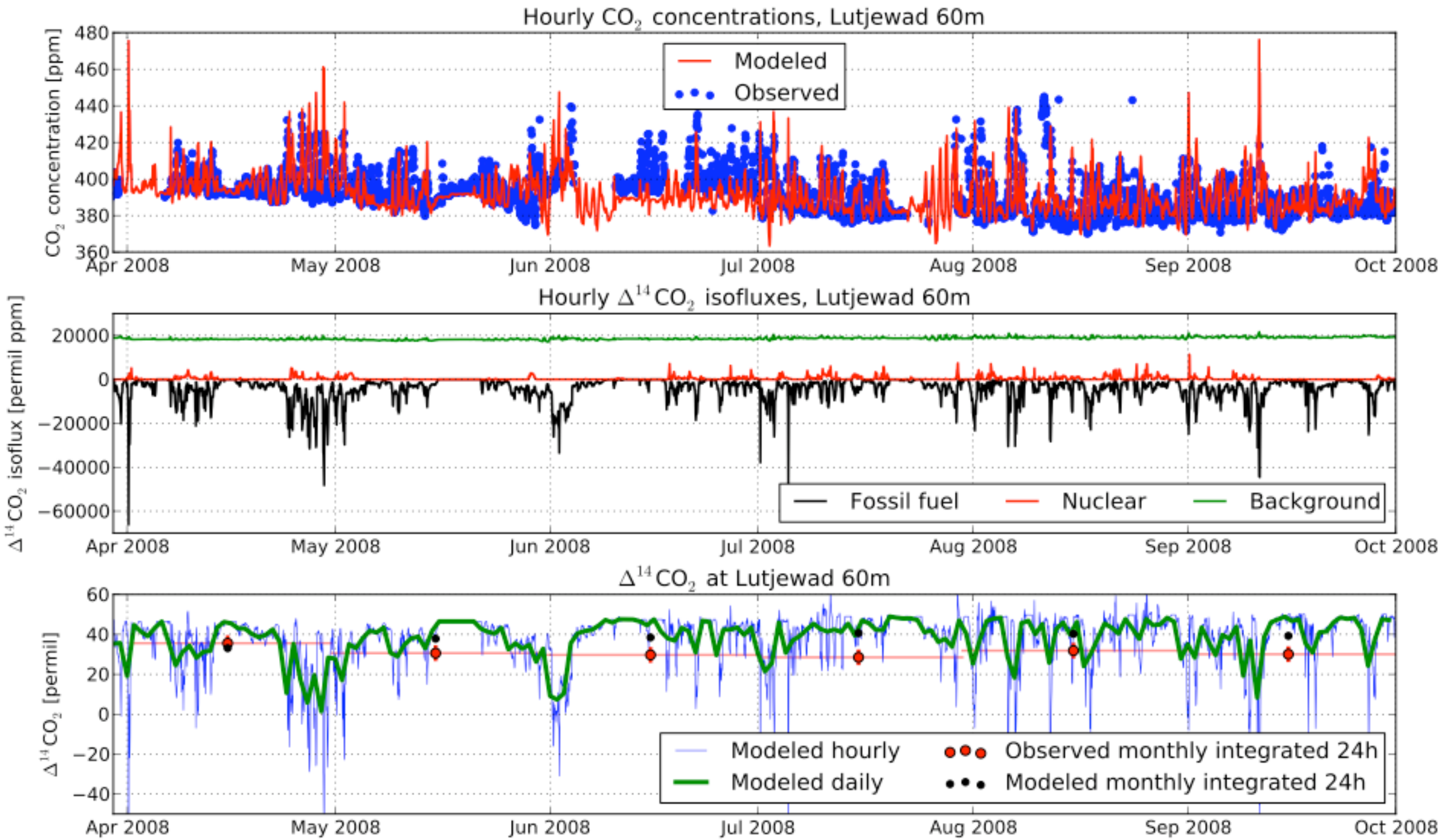

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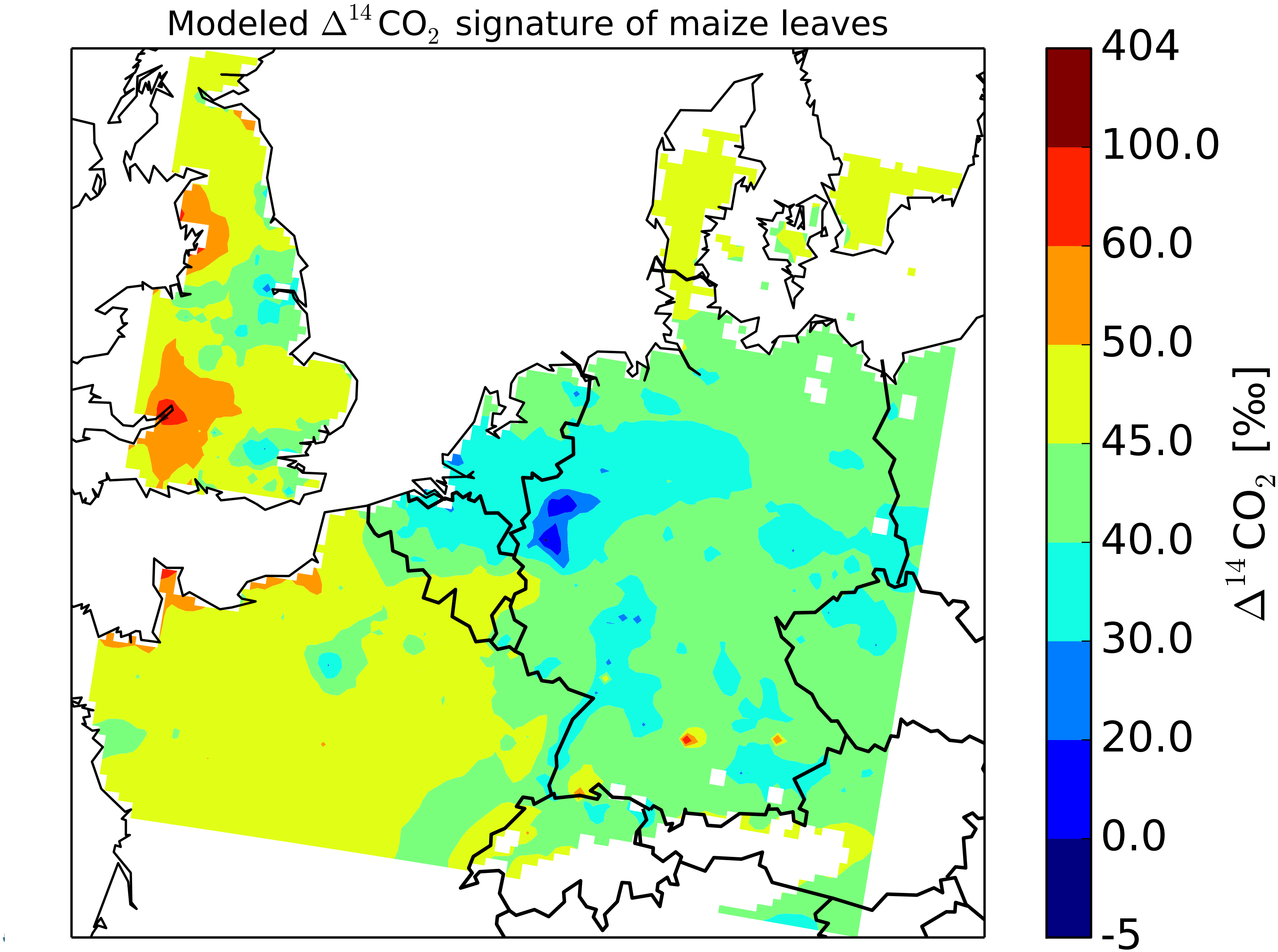
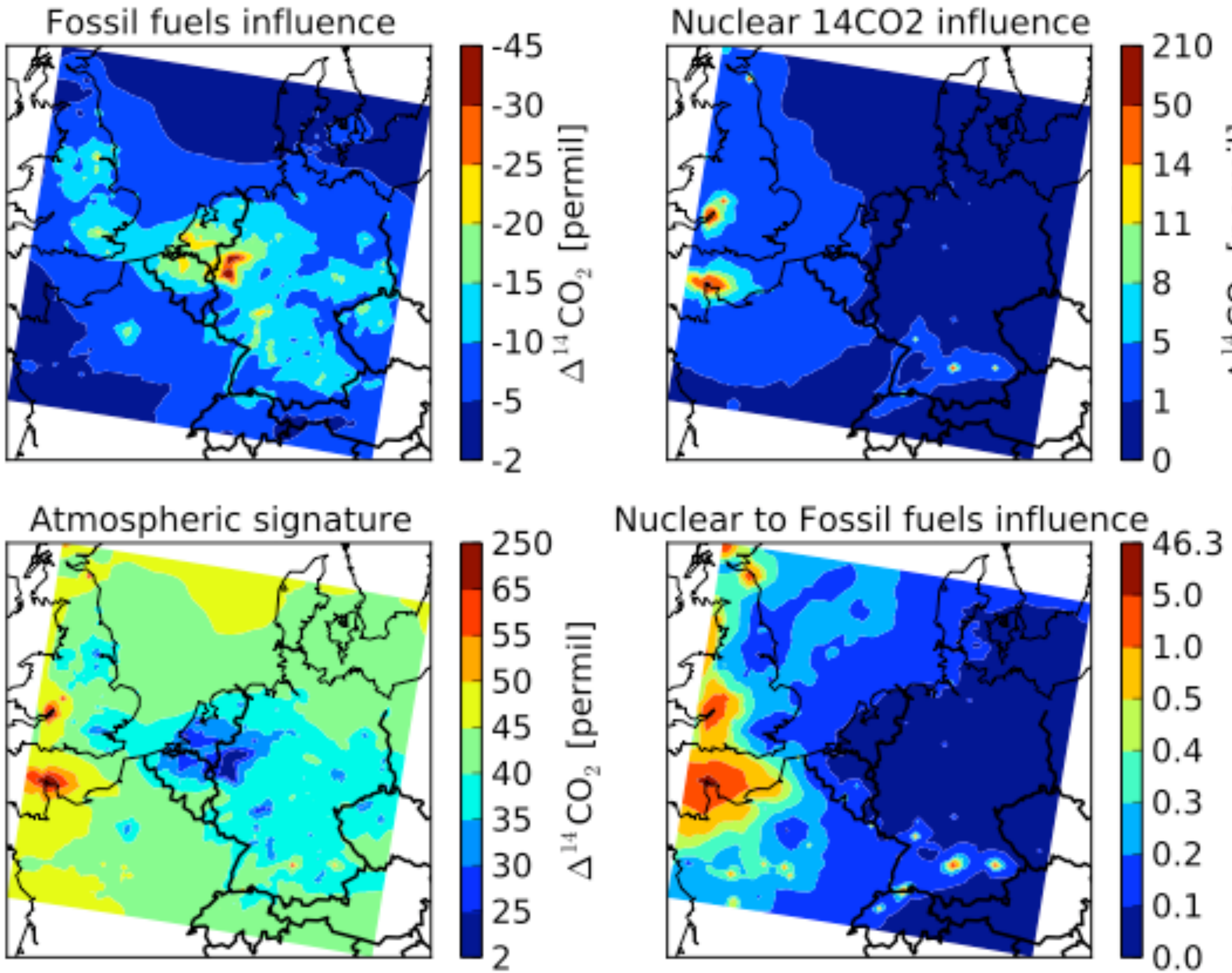
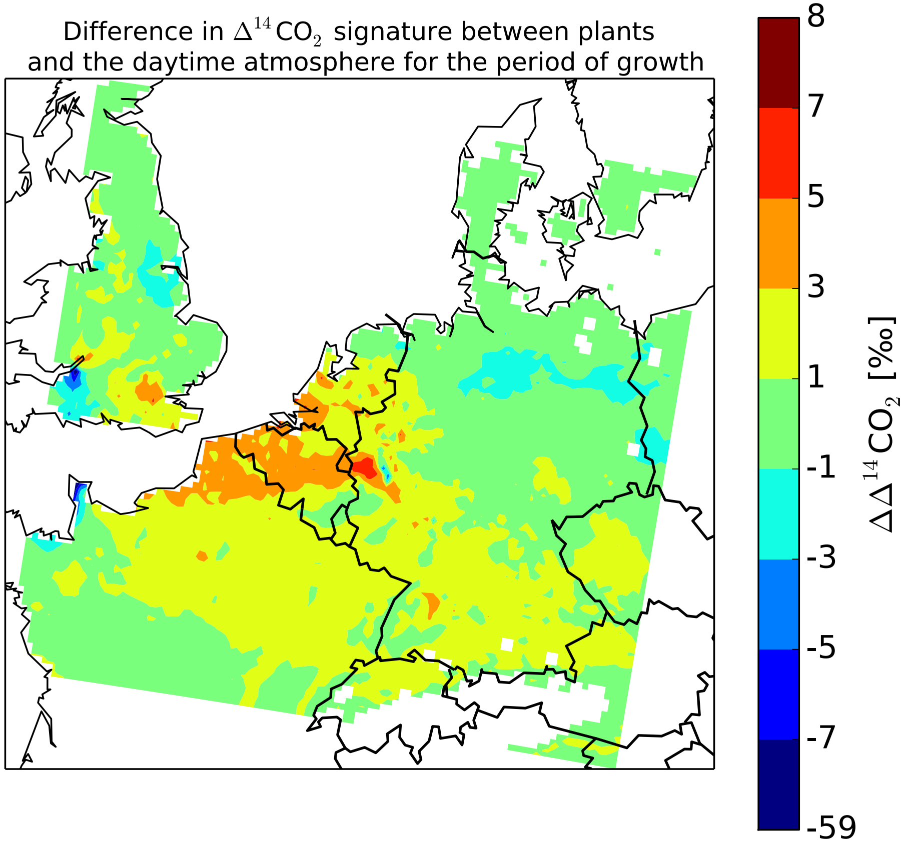


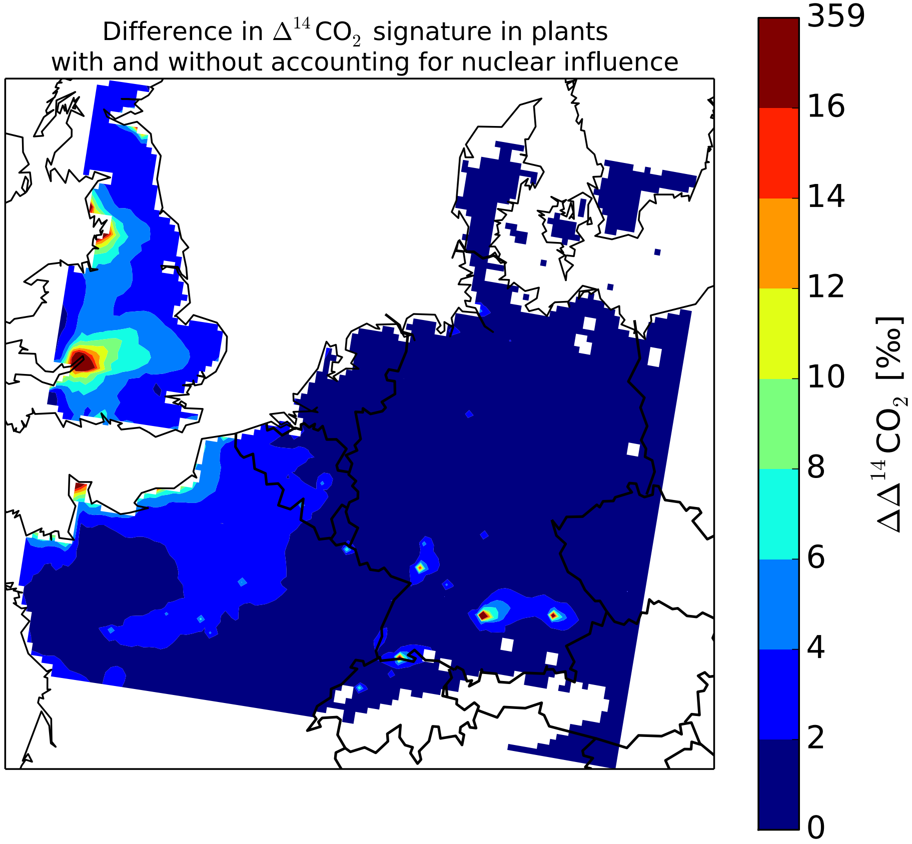
Modeled $\Delta^{14}CO_2$ signature of maize leaves
404
100.0
60.0
50.0
45.0
40.0
30.0
20.0
0.0
-5
$\Delta^{14}CO_2$ [‰]



Fossil fuels influence
-45
-30
-25
-20
-15
-10
-5
-2
Δ14CO2 [permil]
Nuclear 14CO2 influence
210
50
14
11
8
5
1
0
Δ14CO2 [permil]
Atmospheric signature
250
65
55
50
45
40
35
30
25
2
Δ14CO2 [permil]
Nuclear to Fossil fuels influence
46.3
5.0
1.0
0.5
0.4
0.3
0.2
0.1
0.0

Using plants to sample the atmosphere?





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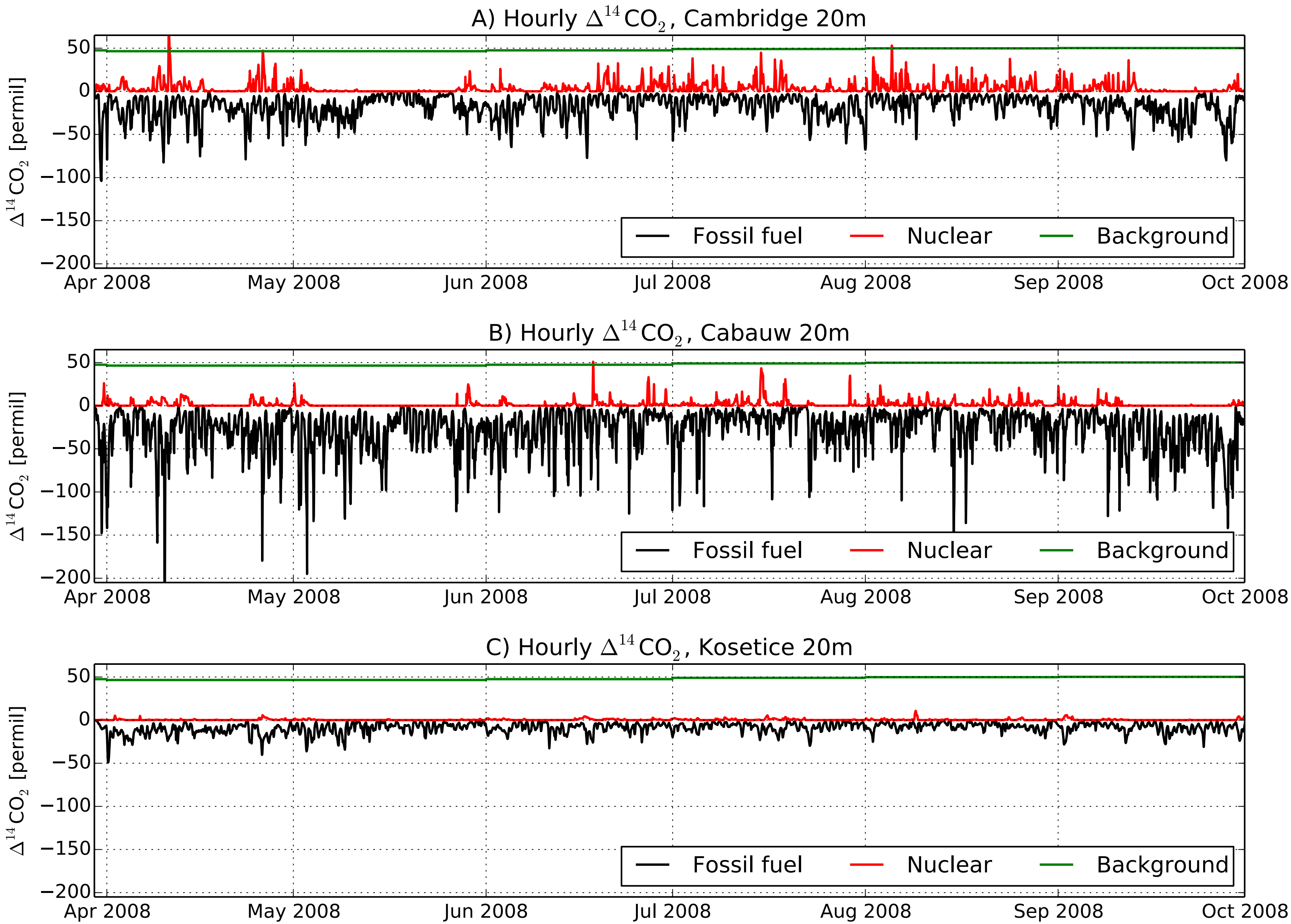
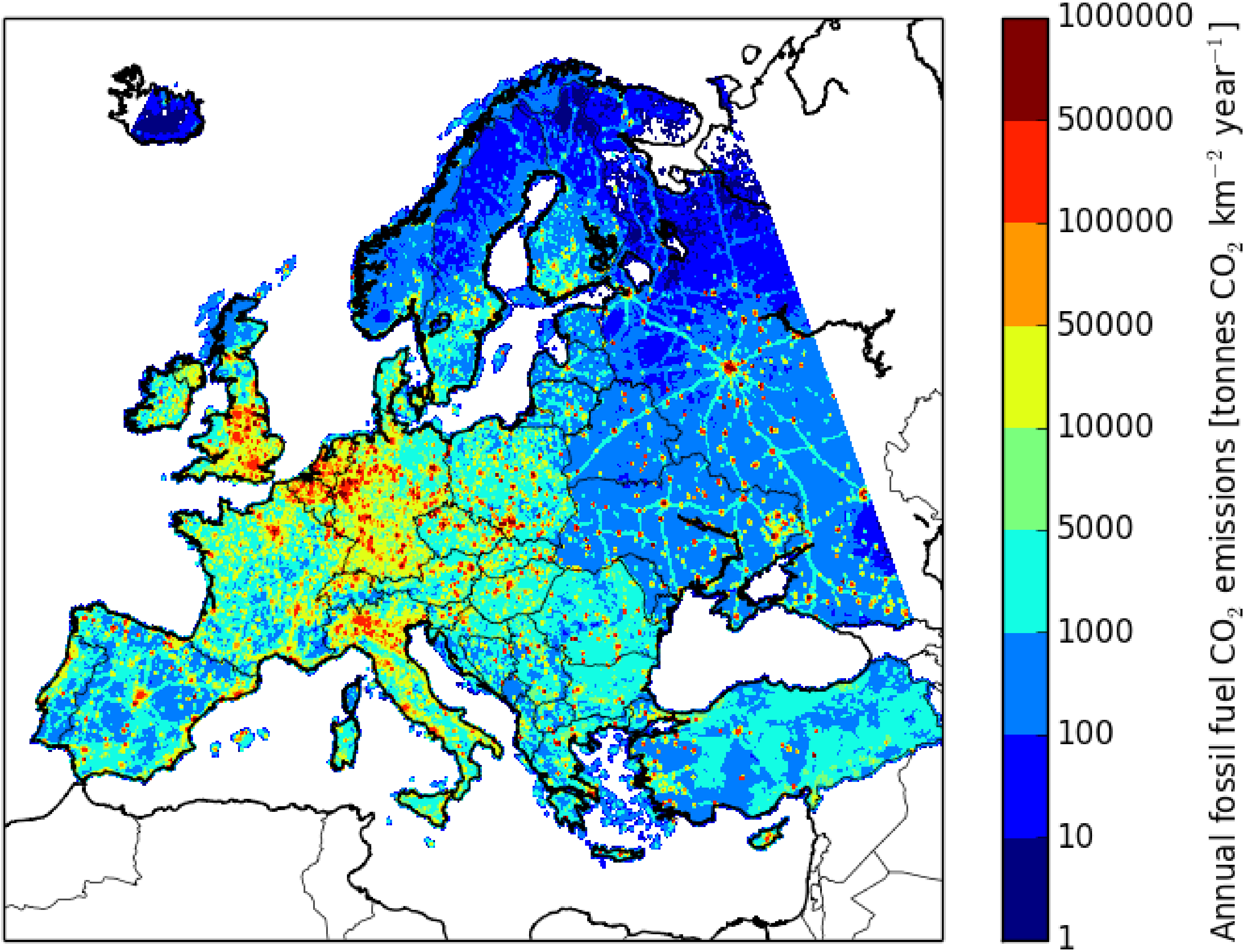
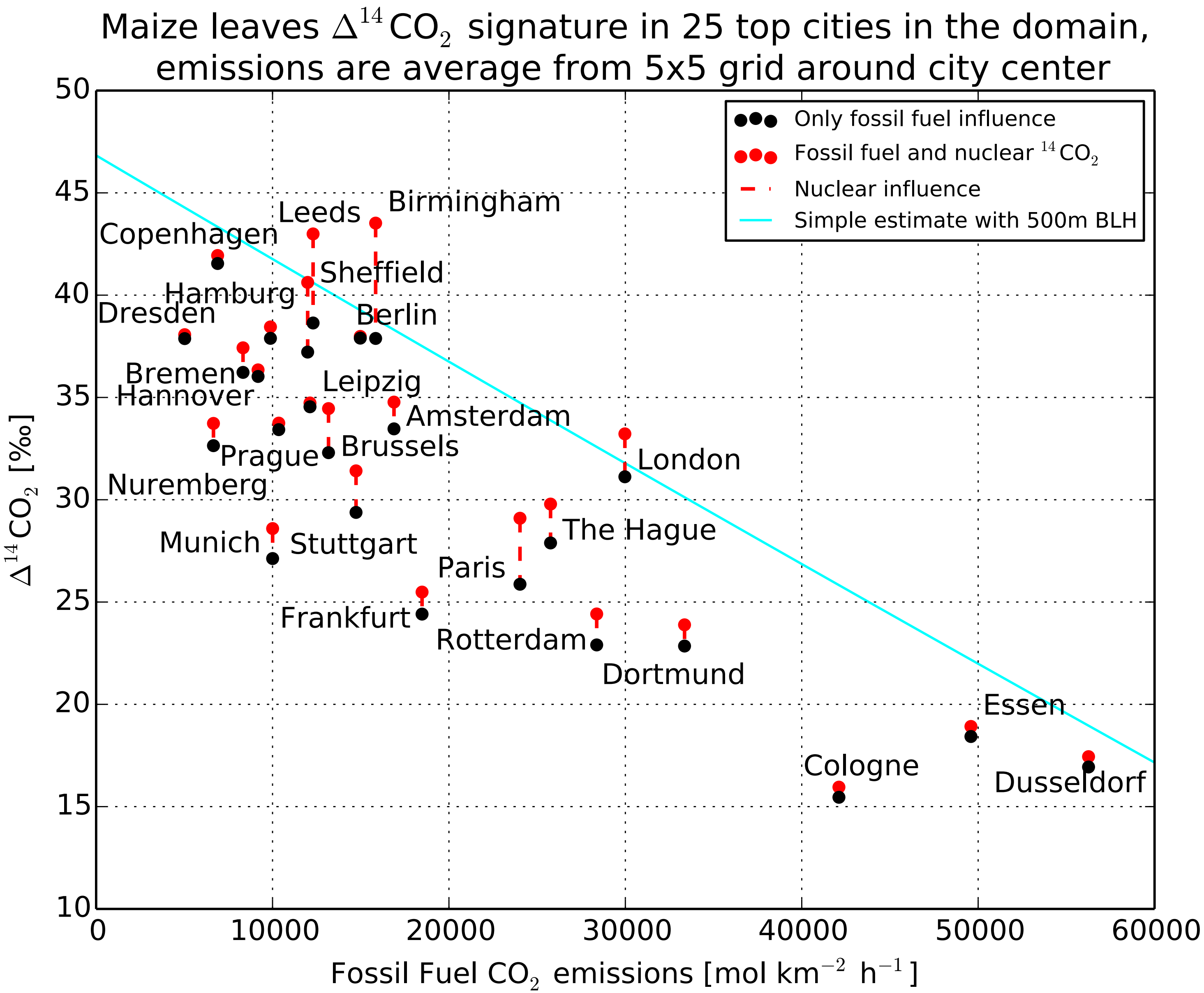
Using plants to sample the atmosphere?



IER fossil fuel emissions





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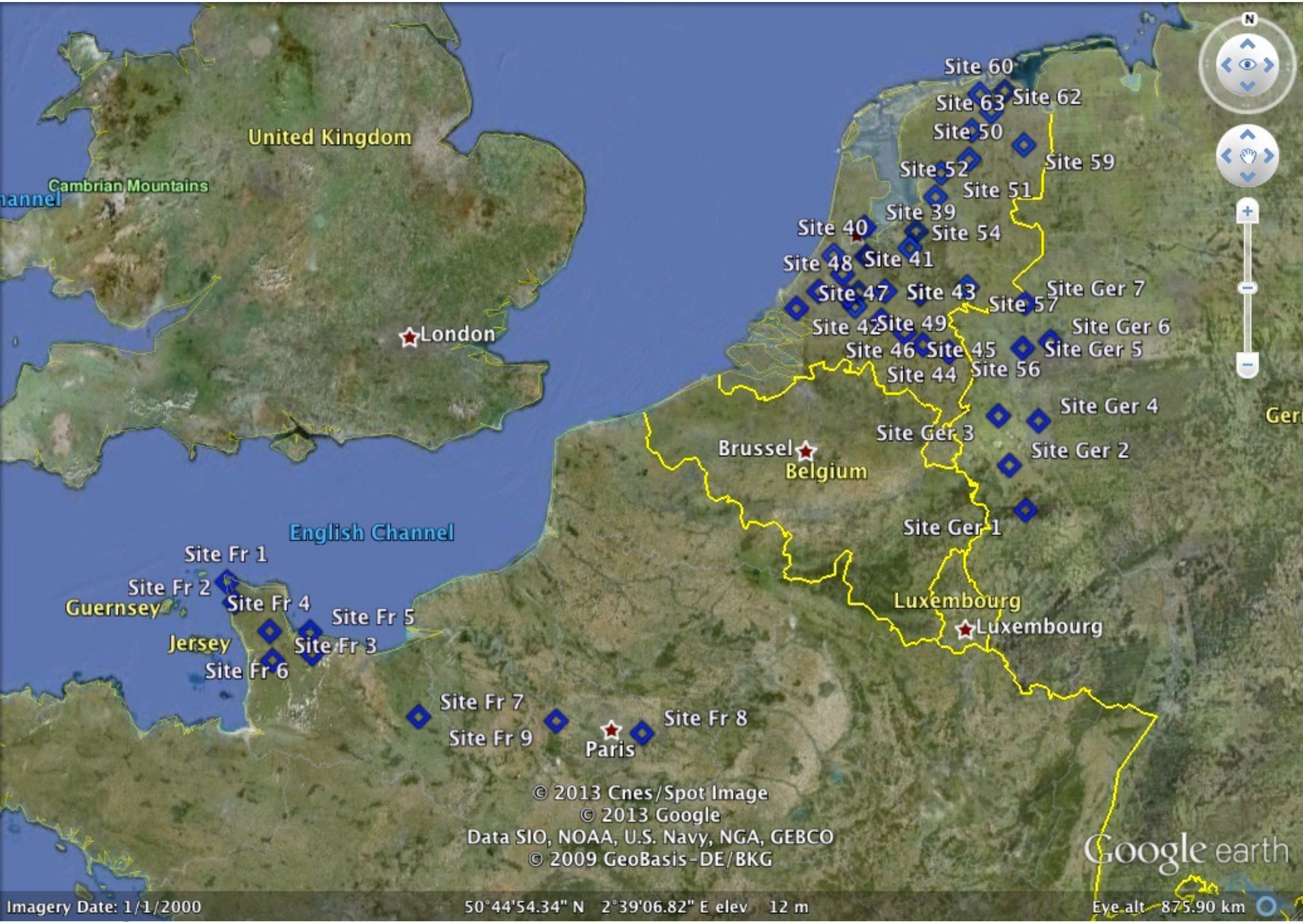
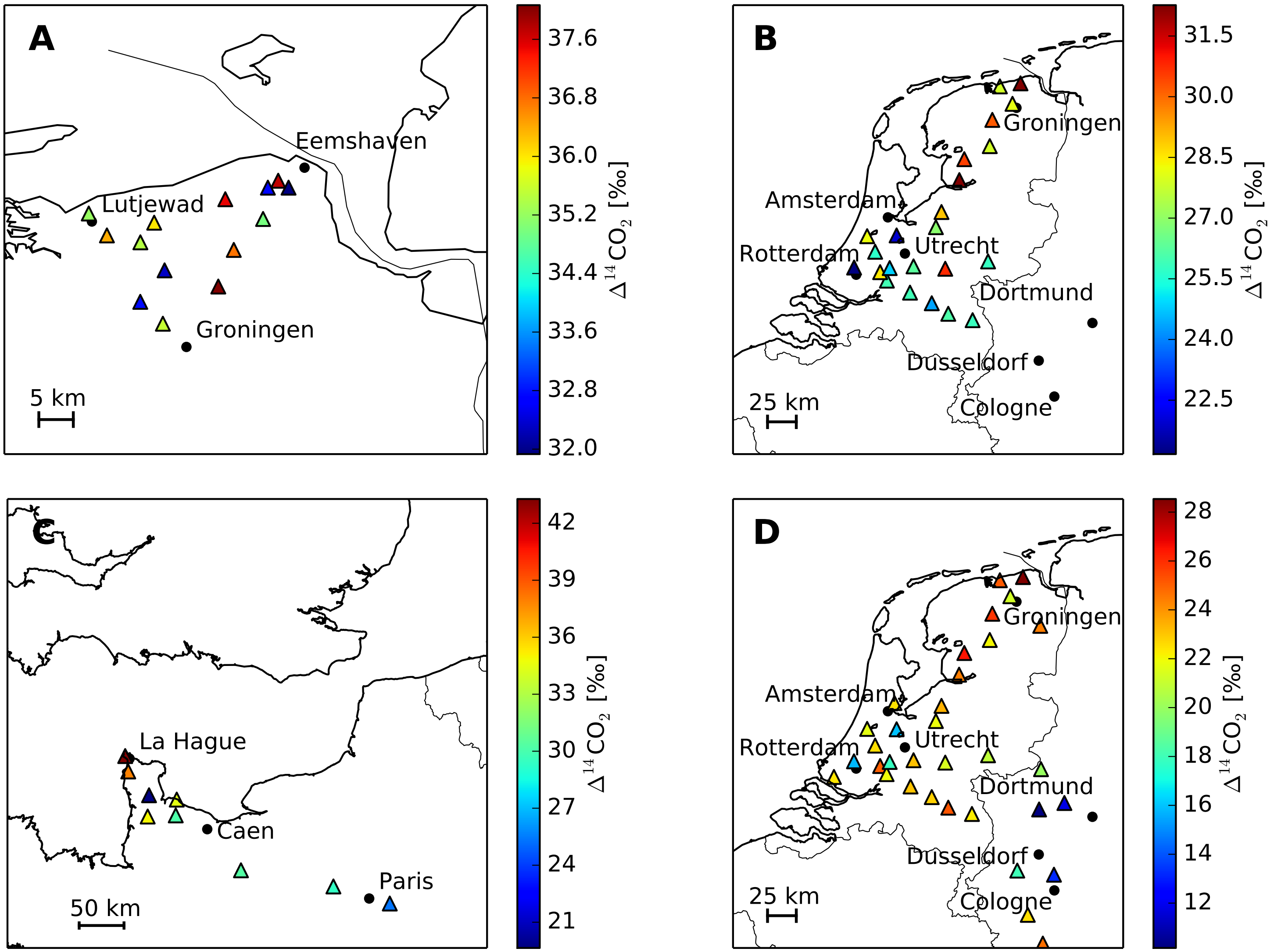
Our regional sampling campaigns

- 2010 – 12 sites in Groningen province, the Netherlands
  - Differences between samples were within the measurement precision of ~ 2‰
- 2011 – 24 sites in three directions in the Netherlands
  - The cleaner north and more polluted south were clearly differentiated, samples from polluted areas more than 10‰ lower than cleaner ~ 3.5 ppm fossil fuel CO_2
- 2012 – 42 samples from the Netherlands, Germany (the Ruhrgebiet) and France (Normandy)
  - clear signatures per country/area
  - nuclear and fossil influence sampled



Site 60
Site 63
Site 62
Site 50
Site 59
Site 52
Site 51
Site 39
Site 40
Site 54
Site 48
Site 41
Site 47
Site 43
Site Ger 7
Site 57
Site 42
Site 49
Site Ger 6
Site 46
Site 45
Site Ger 5
Site 44
Site 56
Site Ger 4
Site Ger 3
Site Ger 2
Site Ger 1
United Kingdom
Cambrian Mountains
London
Brussel
Belgium
Luxembourg
Luxembourg
English Channel
Site Fr 1
Site Fr 2
Guernsey
Site Fr 4
Site Fr 5
Jersey
Site Fr 3
Site Fr 6
Site Fr 7
Site Fr 9
Site Fr 8
Paris
© 2013 Cnes/Spot Image
© 2013 Google
Data SIO, NOAA, U.S. Navy, NGA, GEBCO
© 2009 GeoBasis-DE/BKG
Google earth
Imagery Date: 1/1/2000
50°44'54.34" N 2°39'06.82" E elev 12 m
Eye alt 875.90 km

Maize leaf sampled Δ14C



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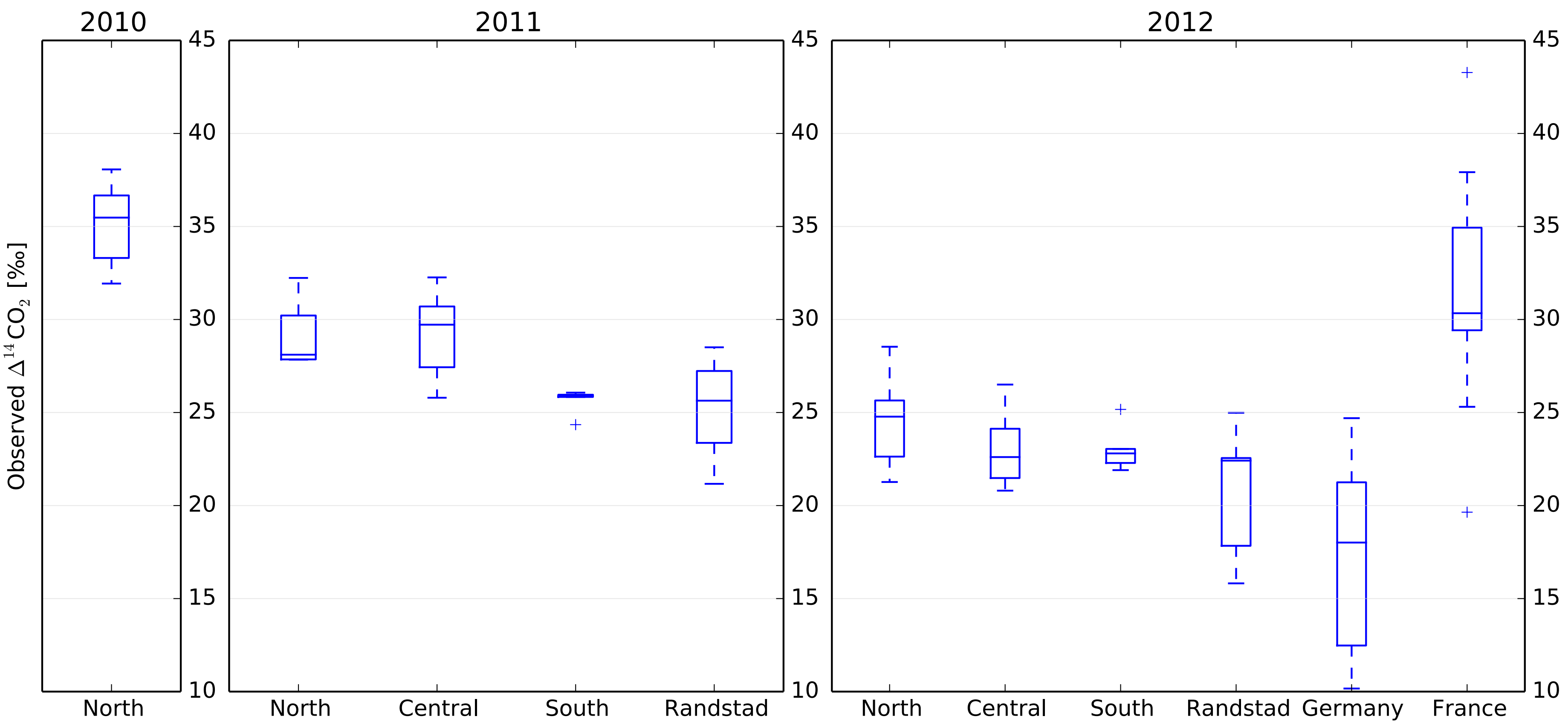
Measured plant samples 2010-2012



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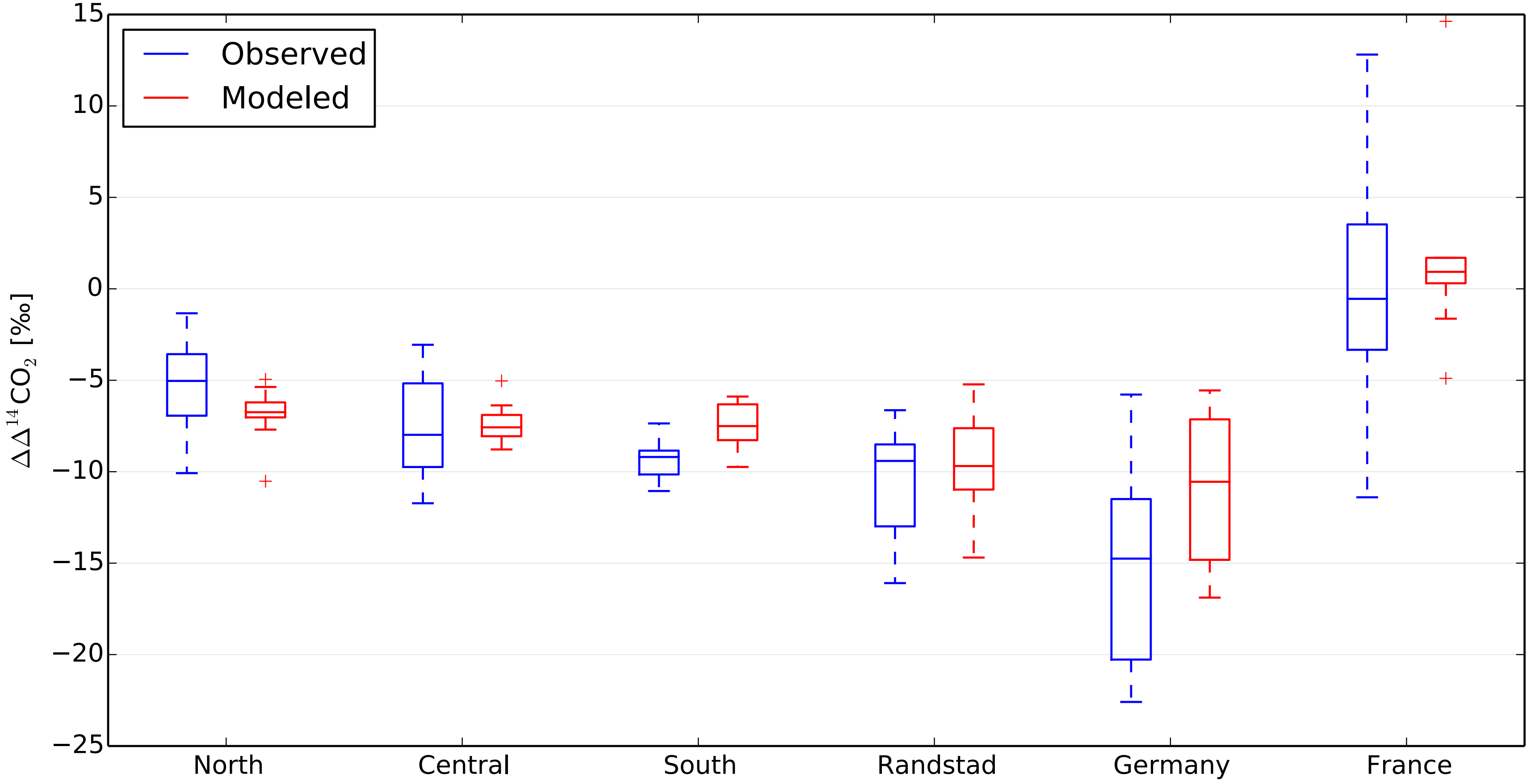
Measured vs modeled Maize samples



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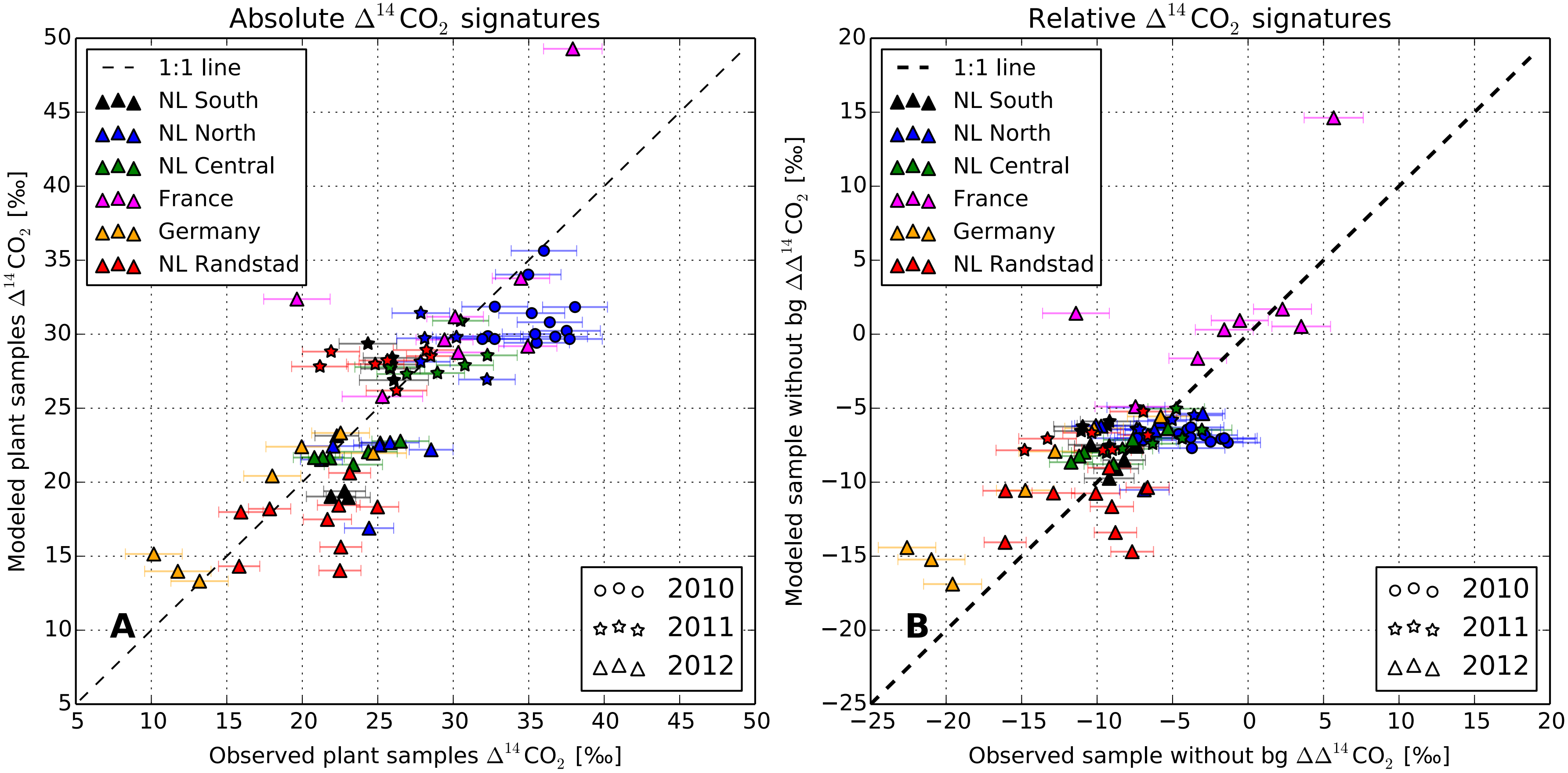
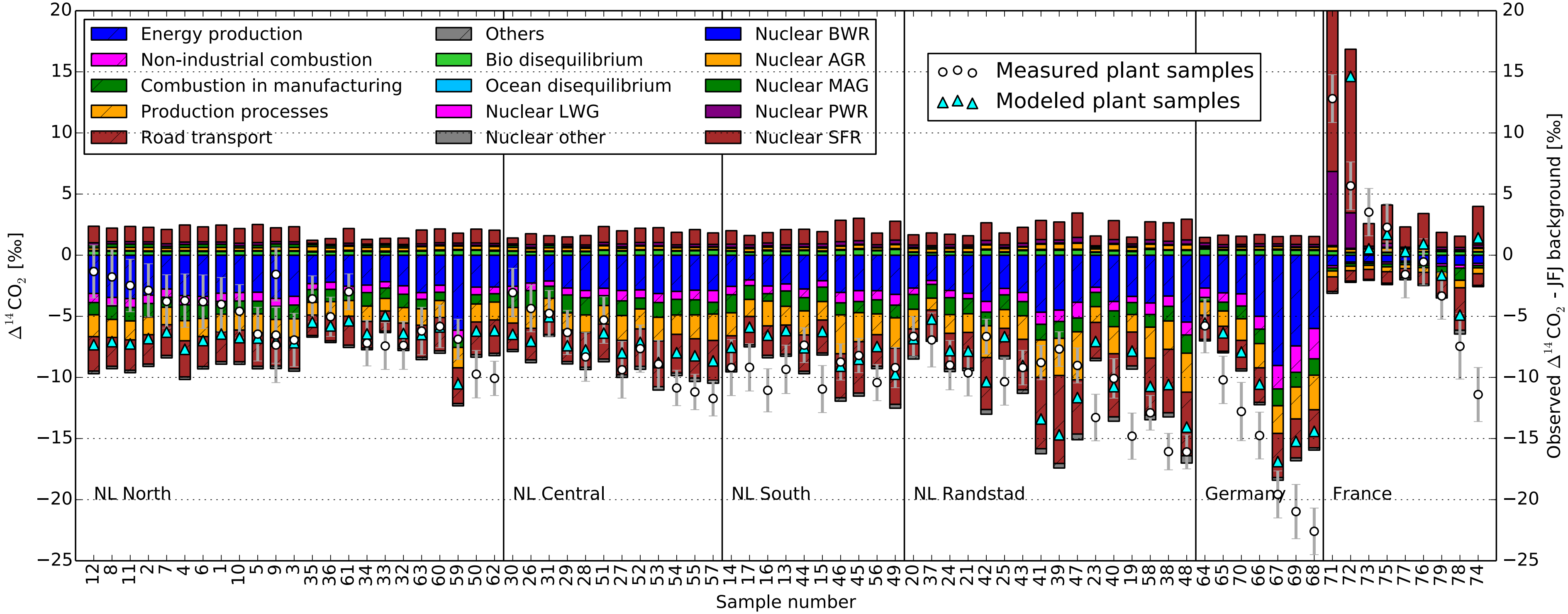
Measured vs modeled Maize samples



Measured vs modeled plant samples: contributions



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Reactor types considered

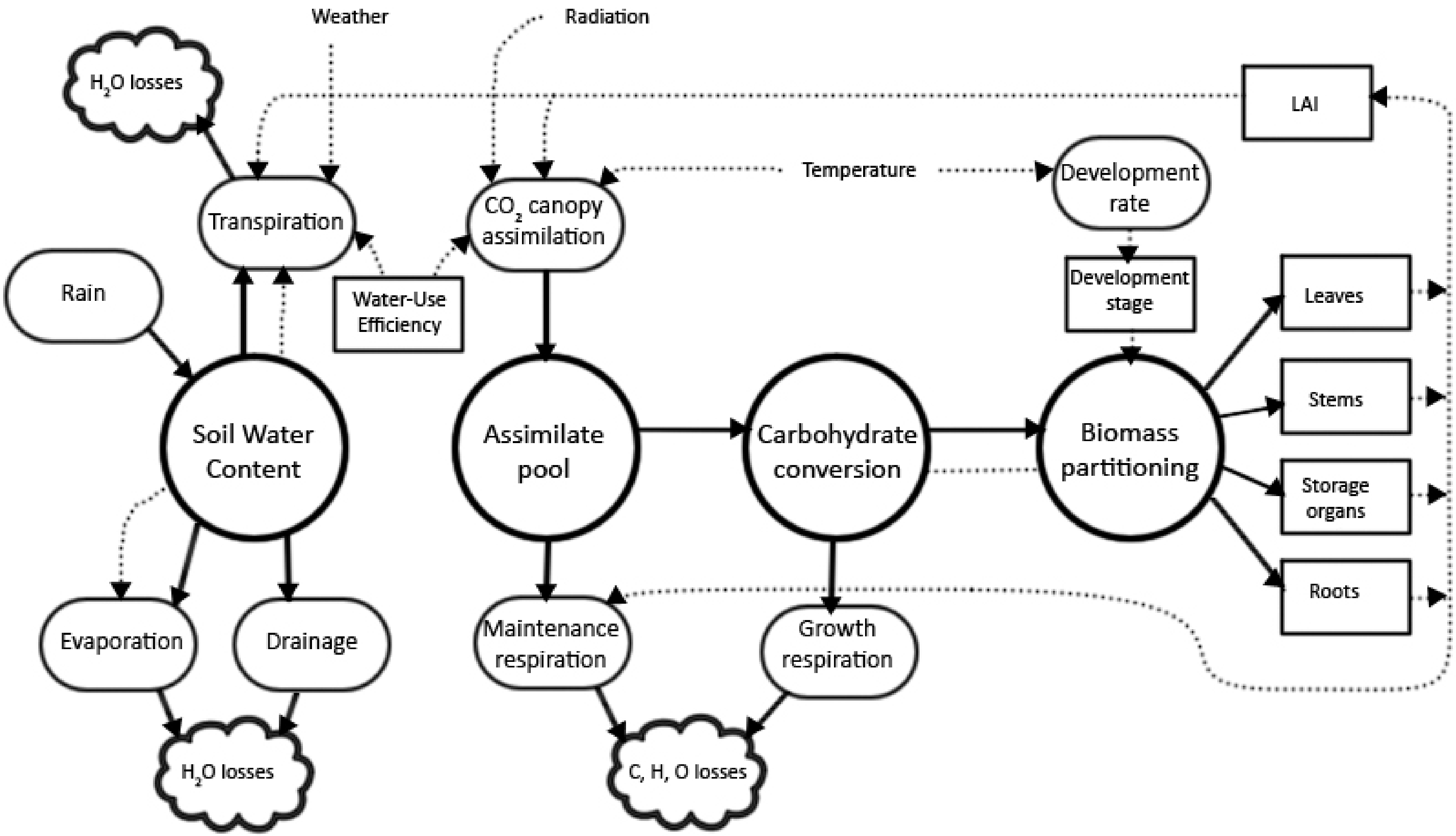
10	$^{14}CO^{0}_{2nuc}$	Total $^{14}CO_2$ emissions from the nuclear industry. EAS.
11	$^{14}CO^{h}_{2nuc}$	Total $^{14}CO_2$ emissions from the nuclear industry. ECAH.
12	$^{14}CO^{SFR}_{2nuc}$	Spent-fuel reprocessing plants. ECAH.
13	$^{14}CO^{PWR}_{2nuc}$	Pressurized water reactors. ECAH.
14	$^{14}CO^{BWR}_{2nuc}$	Boiling water reactors. ECAH.
15	$^{14}CO^{AGR}_{2nuc}$	Advanced gas-cooled reactors. ECAH.
16	$^{14}CO^{MAG}_{2nuc}$	Magnox advanced gas-cooled reactors. ECAH.
17	$^{14}CO^{LWG}_{2nuc}$	Light-water-cooled graphite-moderated reactors. ECAH.
18	$^{14}CO^{rest}_{2nuc}$	Heavy water reactors and Fast-breeder reactors. ECAH.
19	$^{14}CO^{AGR_{temp}}_{2nuc}$	2012 AGR tracer, with temporary shutdowns. ECAH.
20	$^{14}CO^{SFR_{day}}_{2nuc}$	2012 SFRP tracer, only 4h-at-noon emissions. ECAH.
21	$^{14}CO^{SFR_{night}}_{2nuc}$	2012 SFRP tracer, only 4h-at-midnight emissions. ECAH.

What is the temporal profile of 14C release from each site?

Conclusions

- ^{14}C in plants is a promising verification approach over much of Europe
- Absolute signatures in integrated plants still too variable: local background samples must accompany each verification site
- Details matter:
  - Nuclear emissions can dominate fossil fuel signals
  - Plants grow as a function of the weather
  - A plant sample is not the same as an air sample
- We now have a complete framework to describe these details

SUCROS



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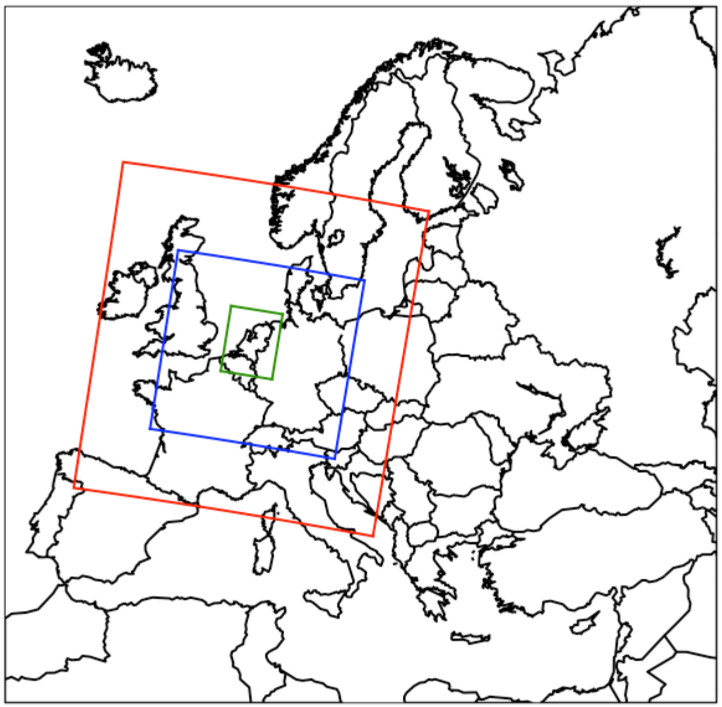
The regional budget of CO_2 and $^{14}CO_2$

- $dC_{atm}/dt = C_{bg} + C_{fossil} + C_{landuse} + C_{bio}^{net} + C_{ocean}^{net}$

- $d\Delta_{atm}C_{atm}/dt = \Delta_{bg}C_{bg} + \Delta_{fossil}C_{fossil} + \Delta_{14}{}^{14}C_{cosmic} +$



$+ \Delta^{bio}{}_{diseq}(C_{landuse}+C_{bio}^{net})$

$+ \Delta^{ocean}{}_{diseq}C_{ocean}^{net} + \Delta_{14}{}^{14}C_{nuclear}$

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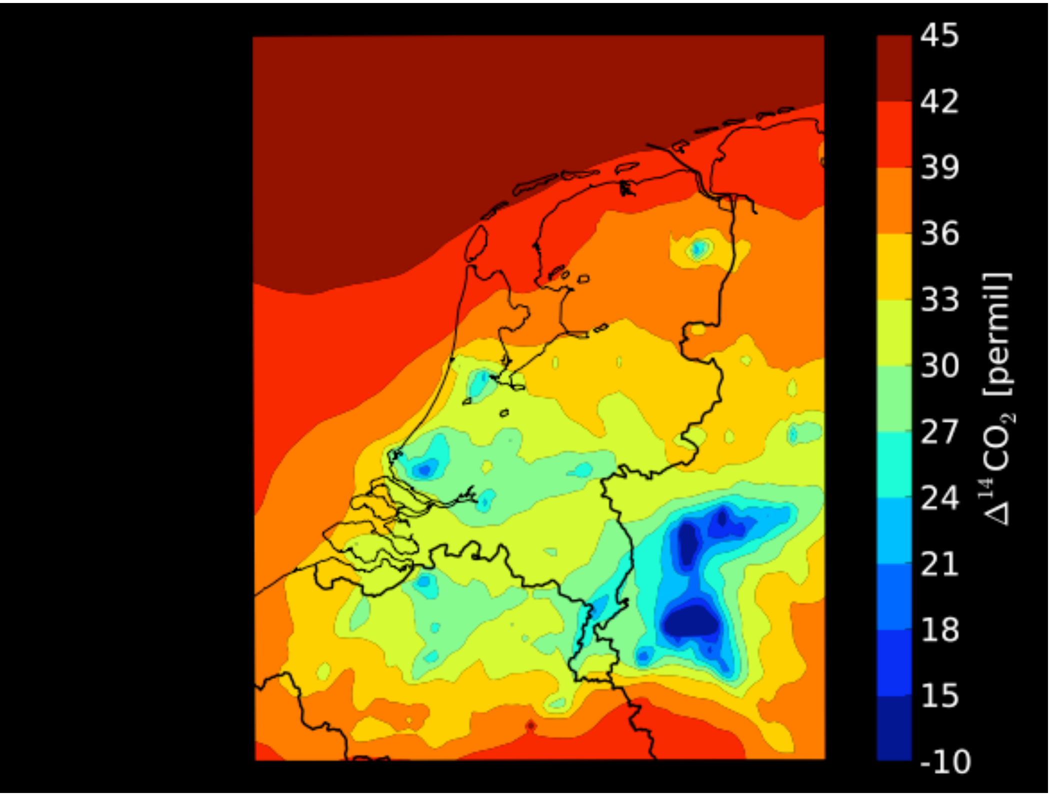
Discussion



- Nuclear emissions uncertainties
- Pollution dispersion in the model
- Biospheric disequilibrium
- Is there a limit in the horizontal resolution?
- Other challenges?

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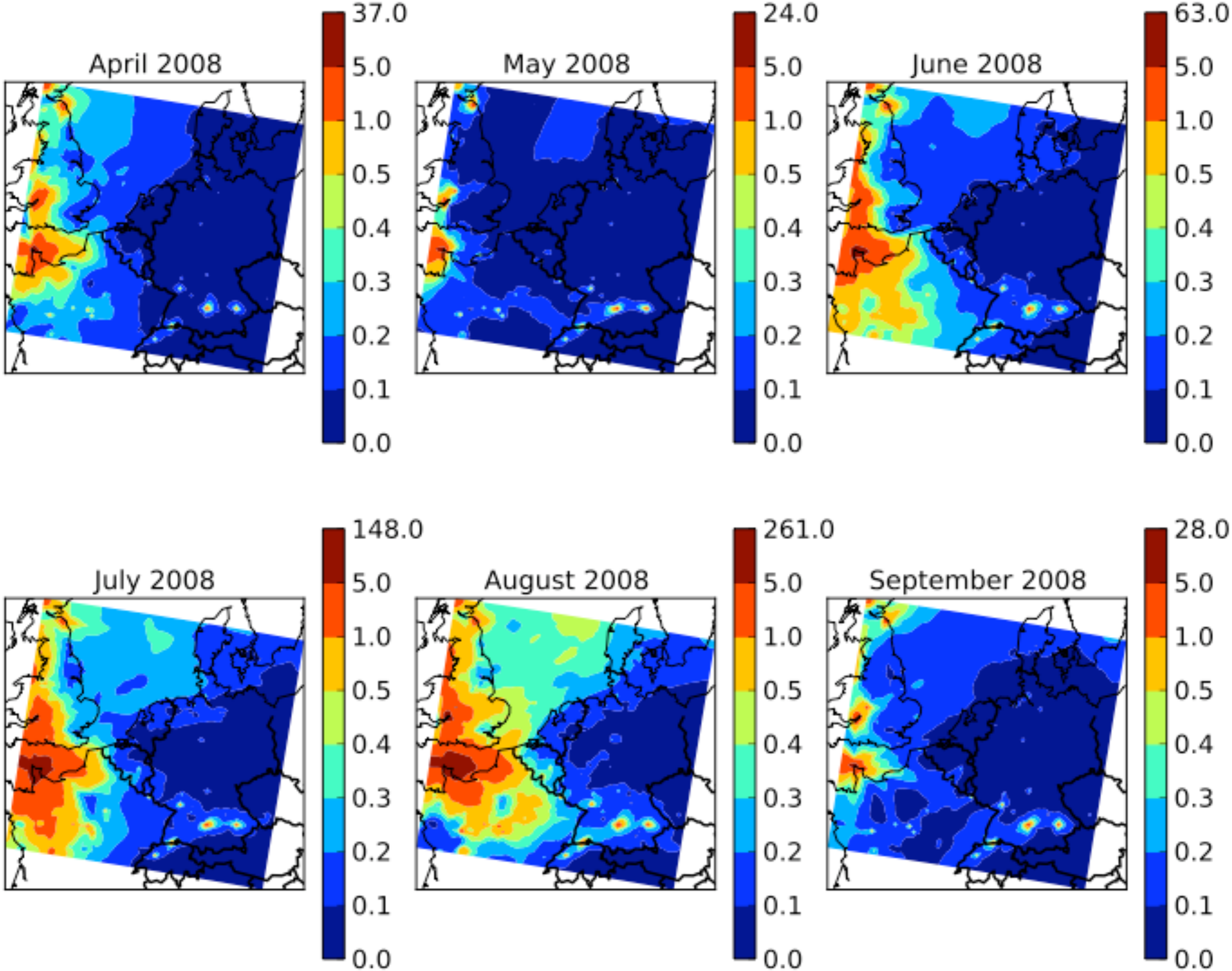
April 2008
May 2008
June 2008
July 2008
August 2008
September 2008
37.0
24.0
63.0
148.0
261.0
28.0
5.0
1.0
0.5
0.4
0.3
0.2
0.1
0.0