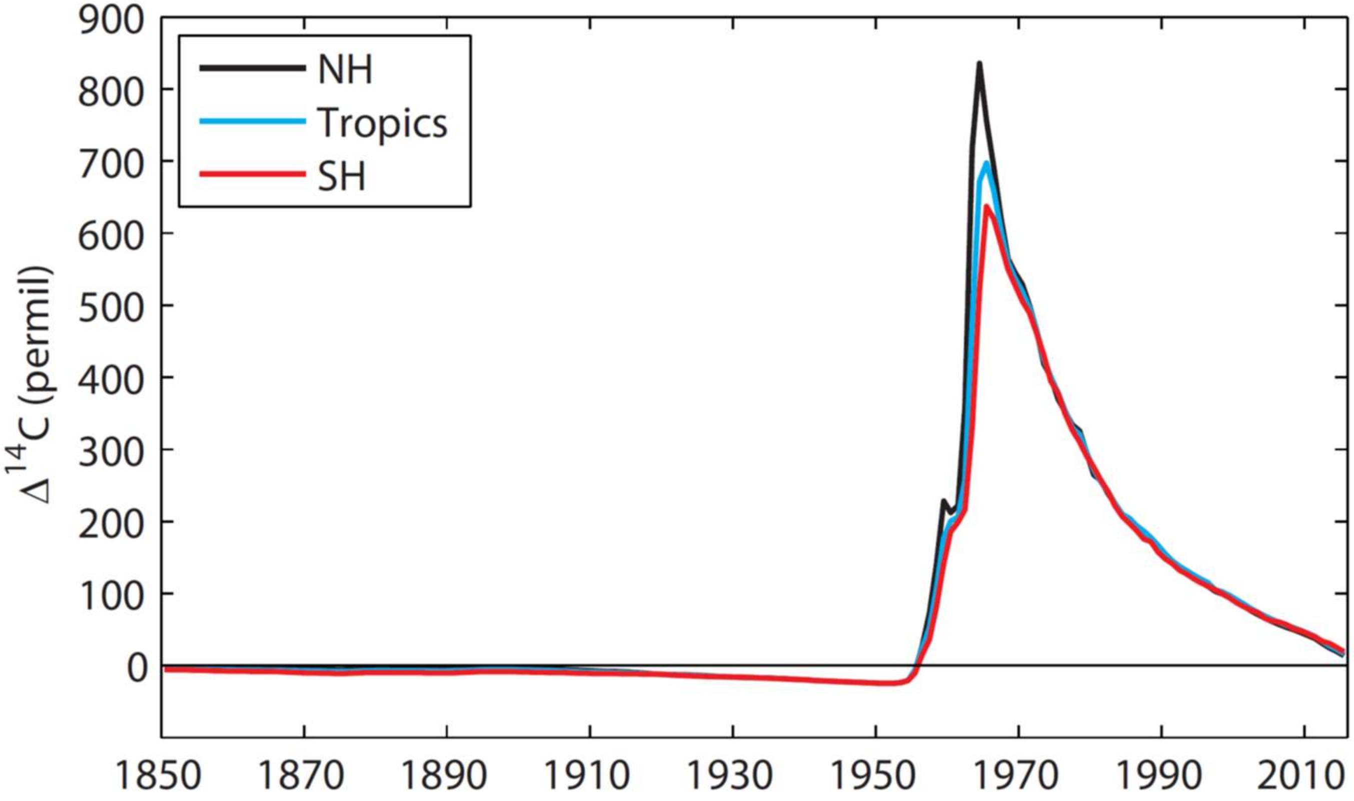
Radiocarbon in Atmospheric CO_2

Heather Graven



Graven et al.
GMDD 2017

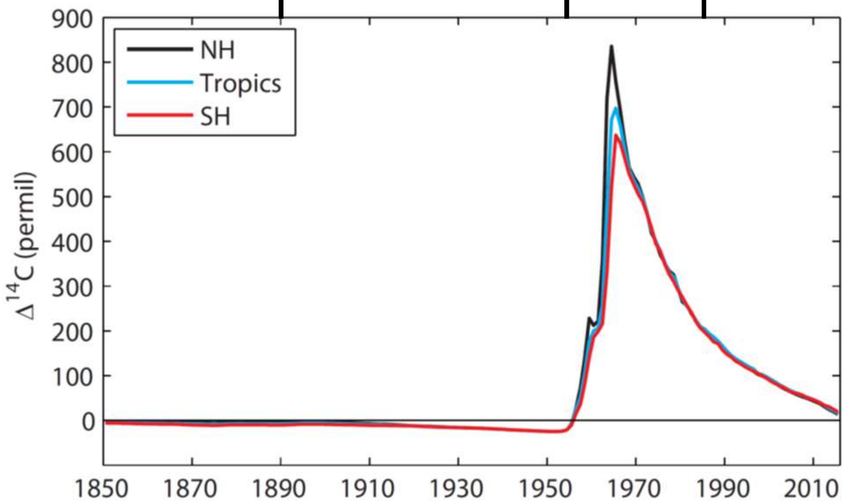
Radiocarbon in Atmospheric CO_2

Influences:

Natural ^{14}C production
Fossil fuel emissions
Anthropogenic ^{14}C production

Stratosphere-troposphere exchange
Ocean-atmosphere exchange
Biosphere-atmosphere exchange

Key Periods: Preindustrial | Early Industrial | Bomb | Post-Bomb | Future



Notation and mixing model concept

I will use $\Delta^{14}C$ in per mil (‰), including fractionation and age corrections (defined as Δ in Stuiver and Polach 1977)

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Approximate mass balances for carbon and ^{14}C, considering the addition of CO_2 from a source (s):

$Cm = Cbg + Cs$ $\quad\quad$ $Cm\ \Delta m = Cbg\ \Delta bg + Cs\ \Delta s$

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Approximate mass balances for carbon and ^{14}C, considering the addition of CO_2 from a source (s):

Cm = Cbg + Cs   Cm Δm = Cbg Δbg + Cs Δs

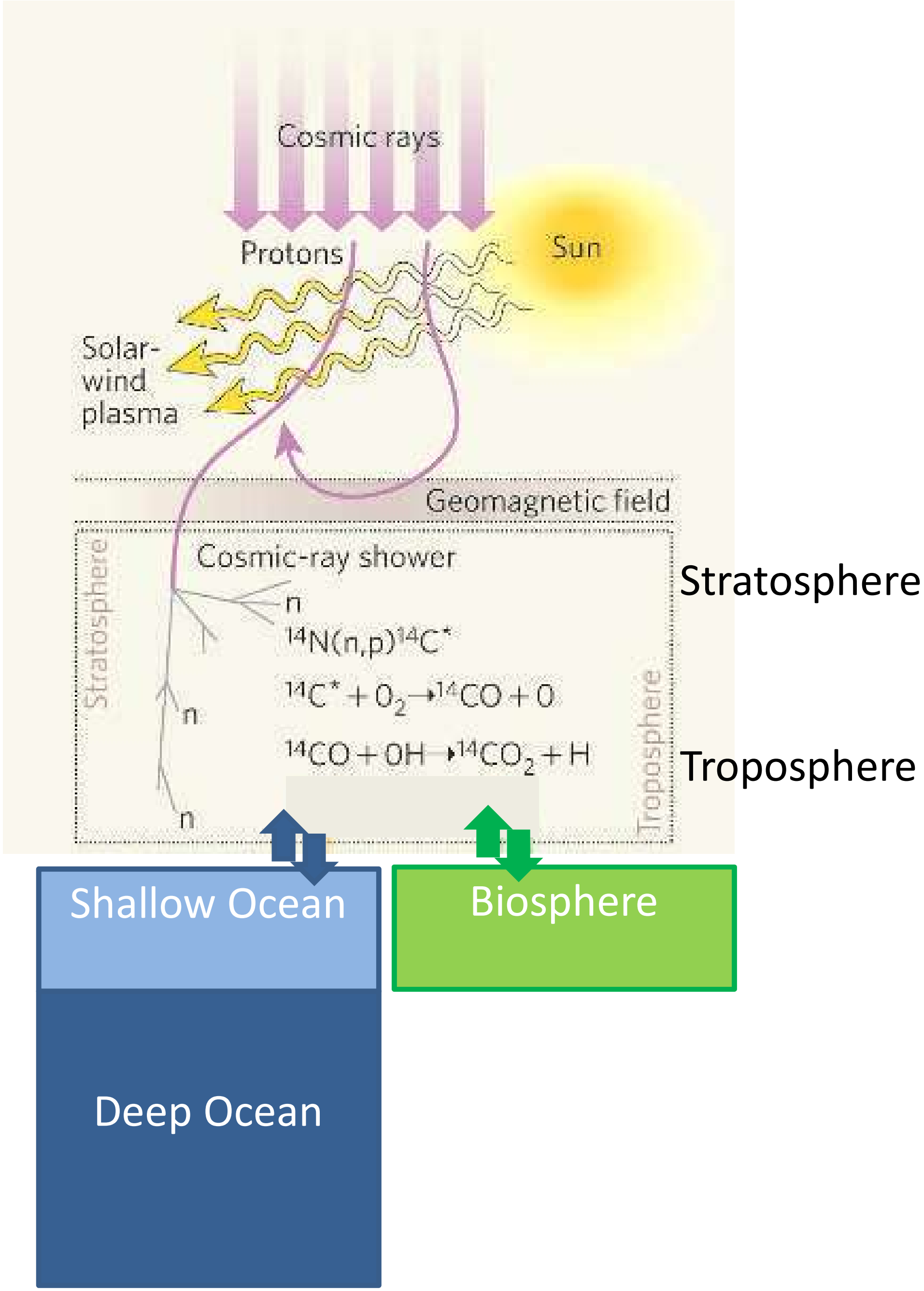
The change in $\Delta^{14}CO_2$ is the difference between the measured (m) and the background (bg) before the source was added:

Δm – Δbg = Cs/Cm (Δs – Δbg)

The sign and the magnitude of the change in $\Delta^{14}C$ depends on the amount Cs and the difference in $\Delta^{14}C$, the "disequilibrium" (Δs – Δbg)

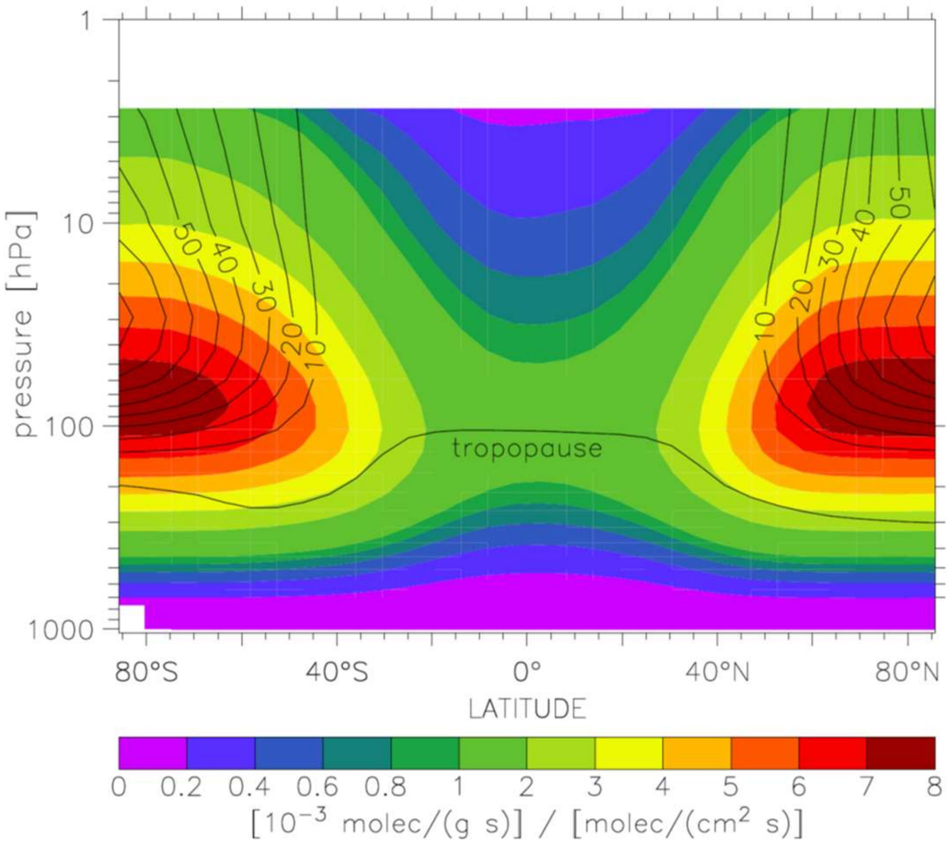
Preindustrial period

- ^{14}C is produced naturally in the atmosphere
- ^{14}C participates in the carbon cycle



Jockel and Brenninkmeijer, 2005

Cosmogenic ^{14}C production occurs primarily in the high latitude stratosphere

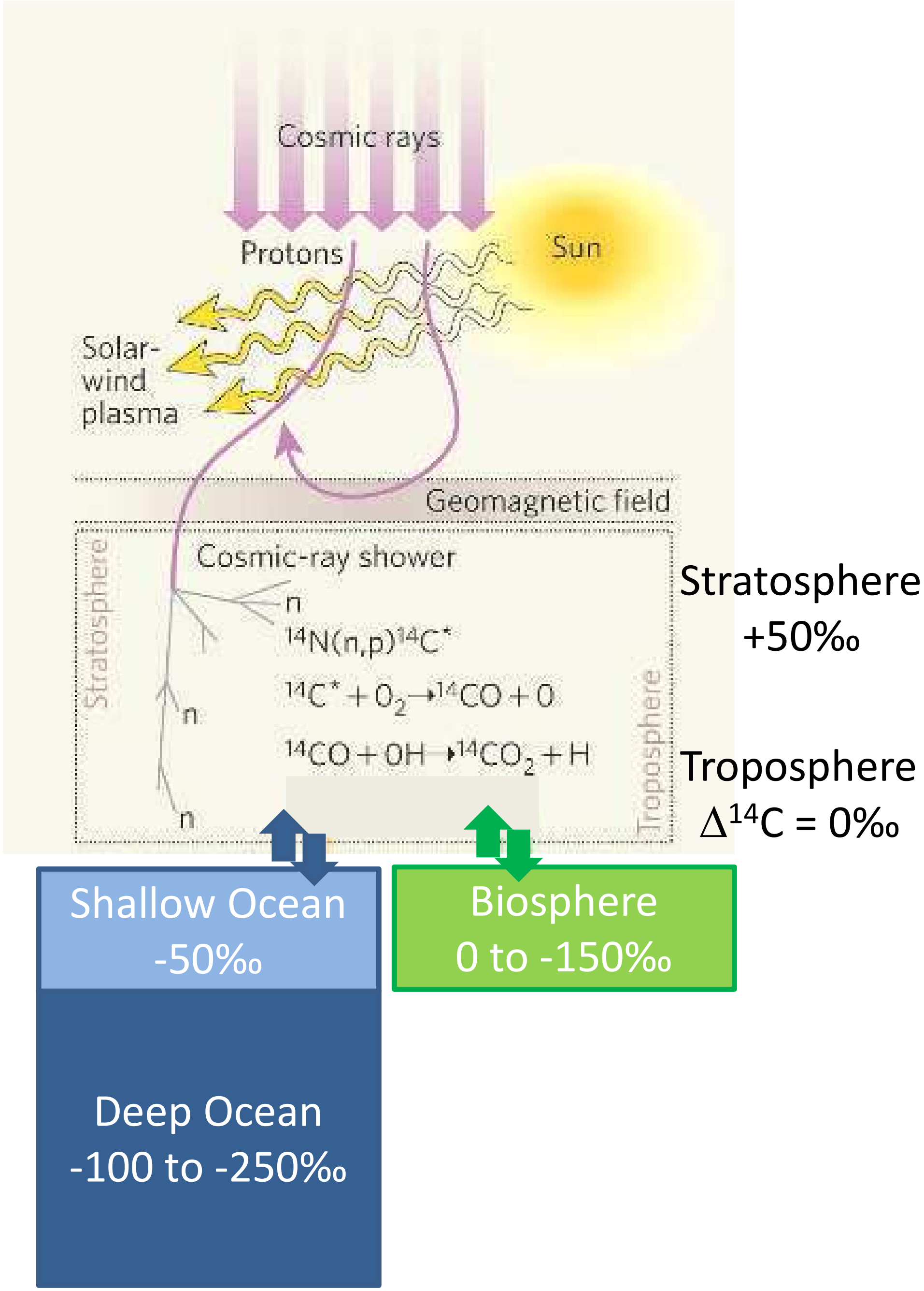


Annual zonal mean galactic cosmic ray induced ^{14}CO production rate (colors), normalized to a global average production rate of 1 molec cm^{-2} s^{-1}

From Jöckel et al. 2003, other work by Lingenfelter, Lal, Masarik, Beer, Jockel, Lowe, and new estimate by Lifton et al. 2014

Preindustrial period

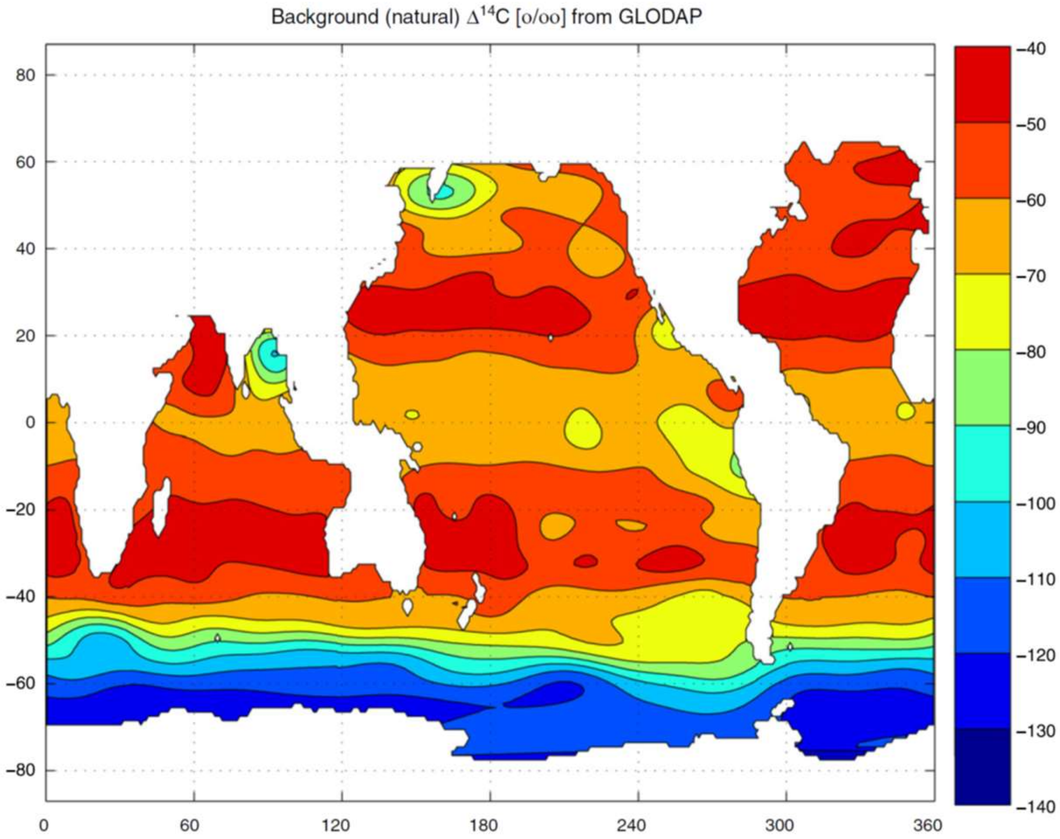
- ^{14}C is produced naturally in the atmosphere
- ^{14}C participates in the carbon cycle
- Biospheric and oceanic carbon are depleted in ^{14}C due to radioactive decay
- $\Delta^{14}C$ standard defined to be similar to preindustrial troposphere

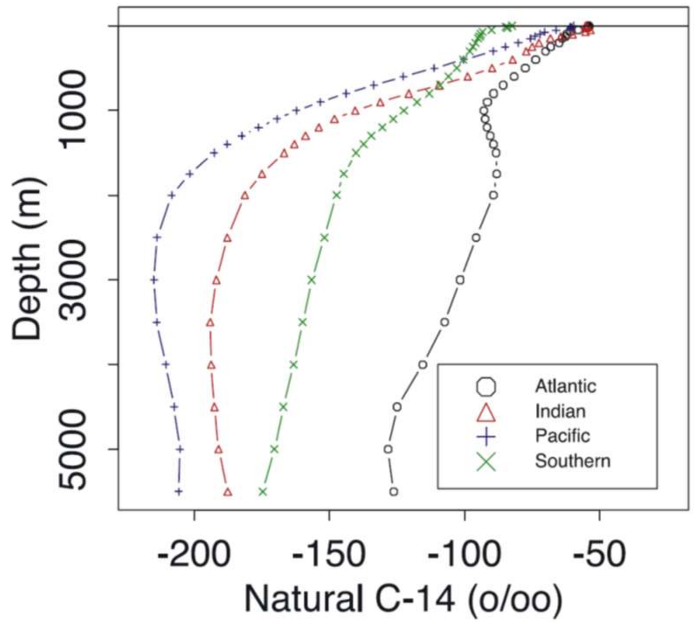


Jockel and Brenninkmeijer, 2005

Preindustrial ocean $\Delta^{14}C$ distribution

Based on oceanic survey data and estimates of anthropogenic ^{14}C (GLODAP)

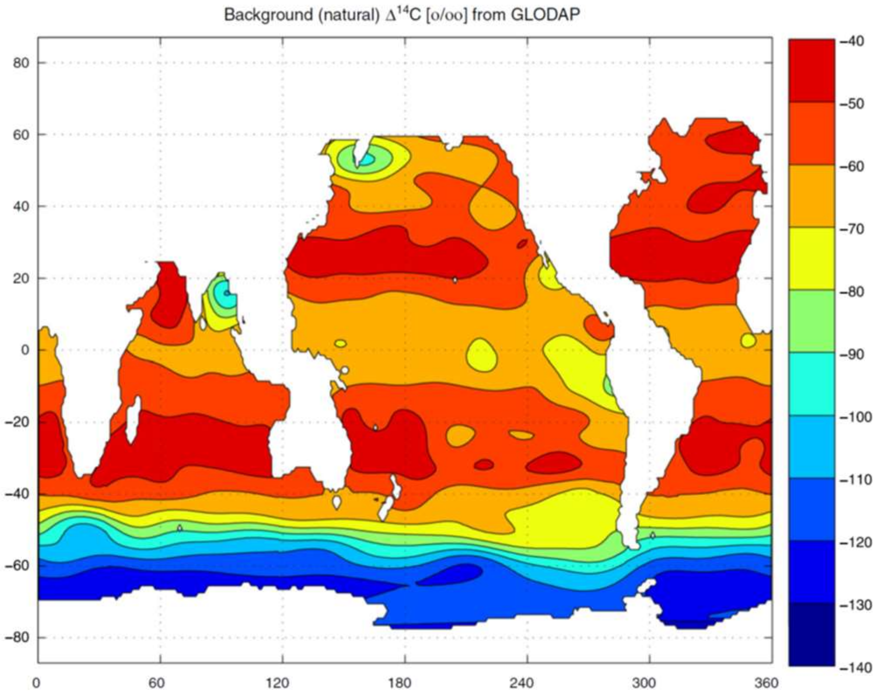




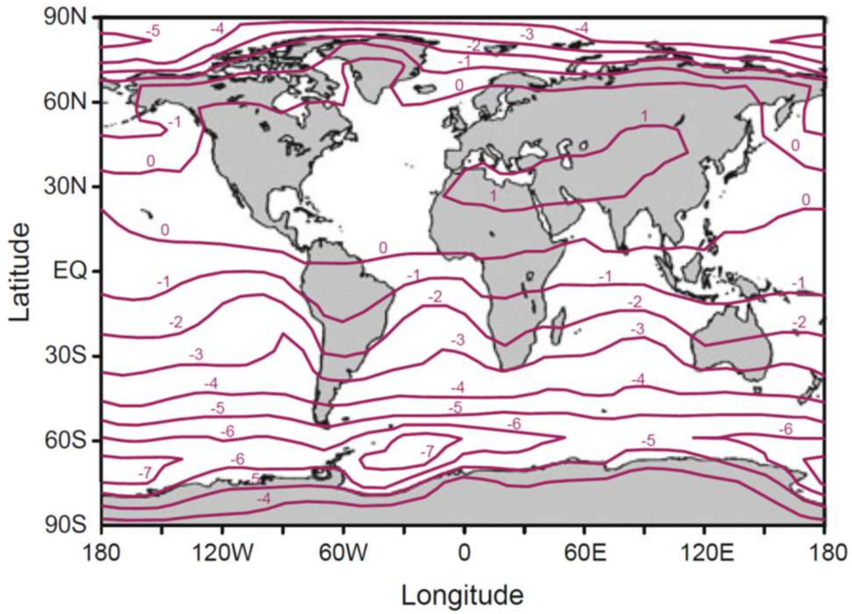
Key et al. 2004; figure from Khatiwala et al. 2012

Preindustrial ocean-atmosphere exchange resulted in higher Δ^{14}C in the Northern Hemisphere, as observed in several studies of tree rings

Preindustrial ocean Δ^{14}C



Preindustrial atmospheric Δ^{14}C (model)



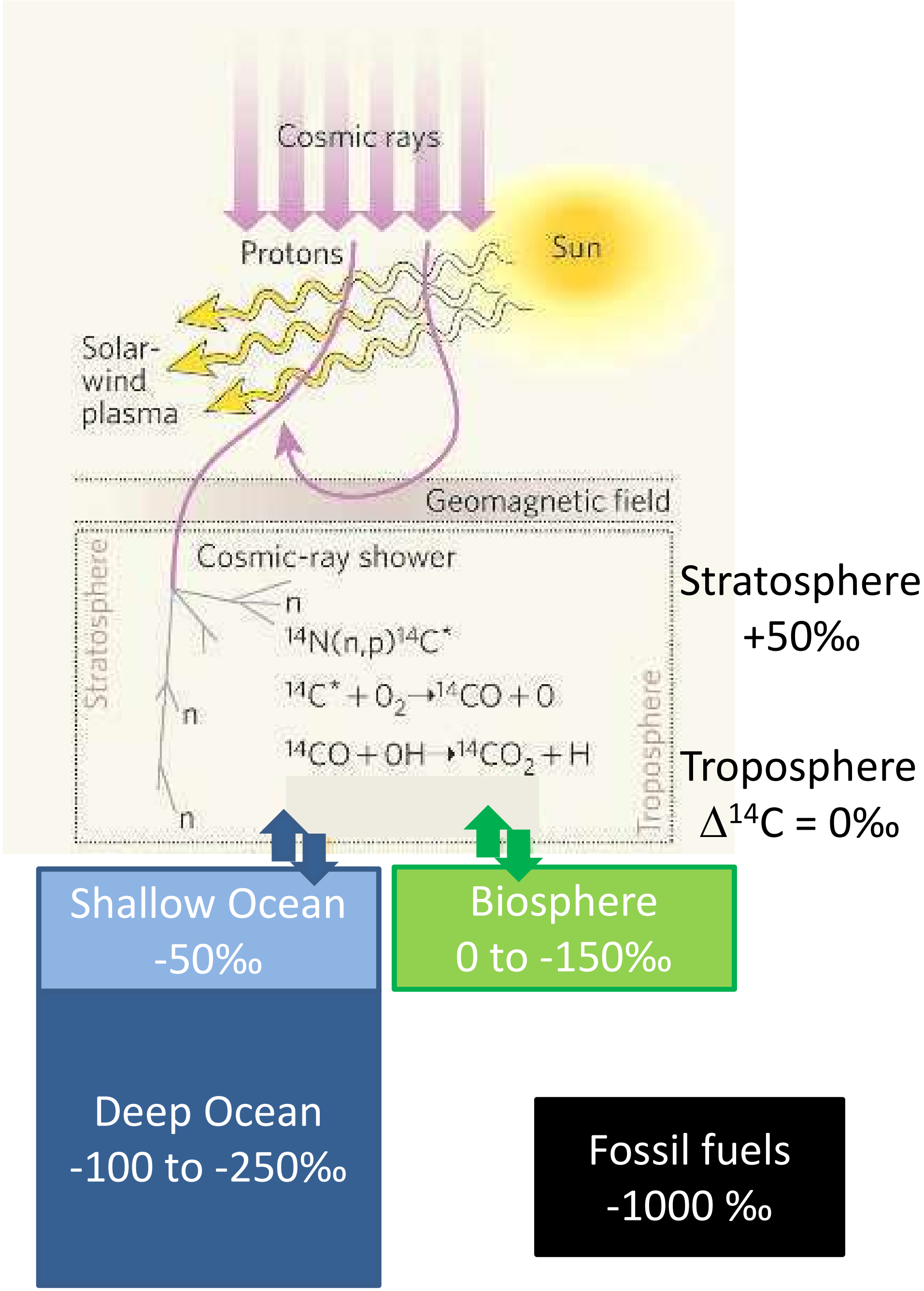
Relative to Washington, USA

Key et al. 2004; figure from Khatiwala et al. 2012

Braziunas et al. 1995

Early industrial period

- ^{14}C is produced naturally in the atmosphere
- ^{14}C participates in the carbon cycle
- Biospheric and oceanic carbon are depleted in ^{14}C due to radioactive decay
- $\Delta^{14}C$ standard defined to be similar to preindustrial troposphere
- Begin to introduce fossil carbon to the atmosphere through fossil fuel combustion



Jockel and Brenninkmeijer, 2005

“The Suess Effect” – Dilution of atmospheric $^{14}CO_2$ by fossil fuel-derived ^{14}C-free CO_2

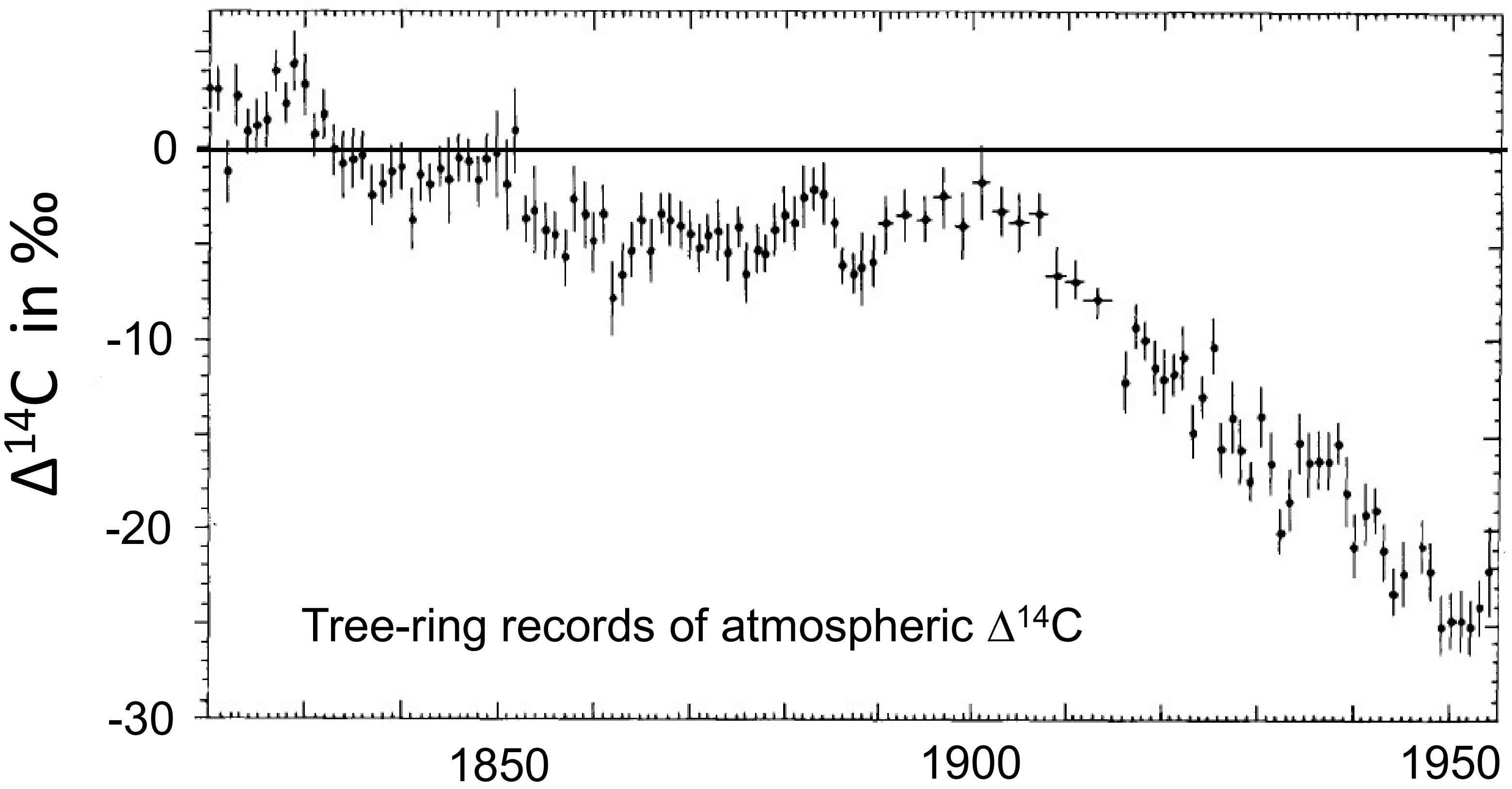
Radiocarbon Concentration in Modern Wood

Redeterminations of the absolute C^{14} concentration in wood carbon and of its variations since the industrial revolution became widespread in the late 19th century were carried out (*1*) by means of proportional counting of acetylene as described previously (*2*). No direct determination of the counting efficiency was made, but the counting rates were compared with those from material containing a mass-spectrometrically determined amount of artificial C^{14}, obtained from the National Bureau of Standards through the courtesy of H. H. Seliger.

Indications of a decrease in the specific C^{14} activity of wood at time of growth during the past 50 yr had been found previously (*3*). The decrease amounted to about 3.4 percent in two trees from the east coast of the United States. A third tree, from Alaska, investigated at that time, showed a smaller effect. The decrease can be attributed to the introduction of a certain amount of C^{14}-free CO_2 into the atmosphere by artificial coal and oil combustion and to the rate of isotopic exchange between atmospheric CO_2 and the bicarbonate dissolved in the oceans.

Hans Suess, Science, 1955

Early industrial or “Suess” period – “The Suess Effect” in the global carbon cycle



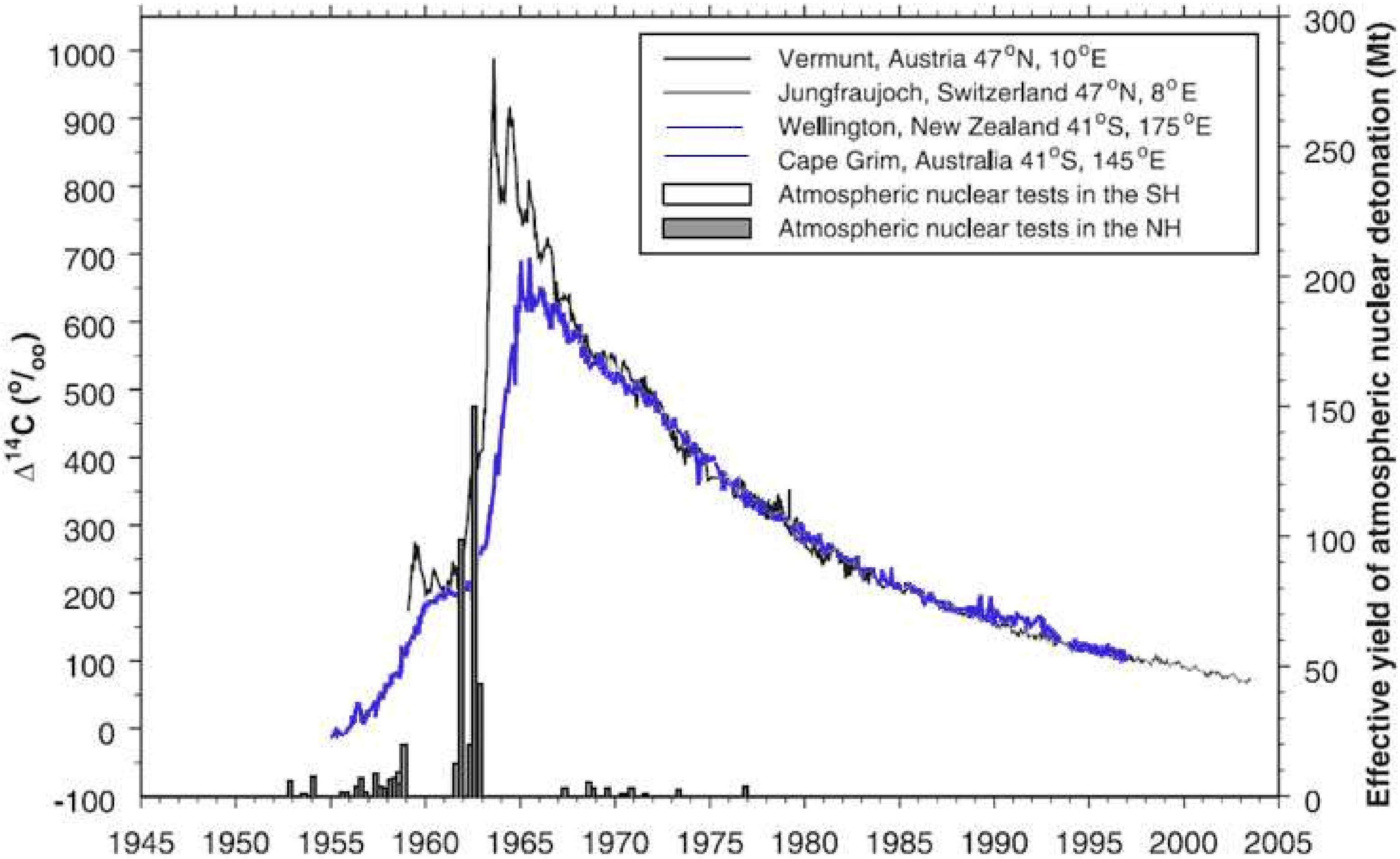
Stuiver and Quay, 1981

"Atom Bomb Effect"—Recent Increase of Carbon-14 Content of the Atmosphere and Biosphere

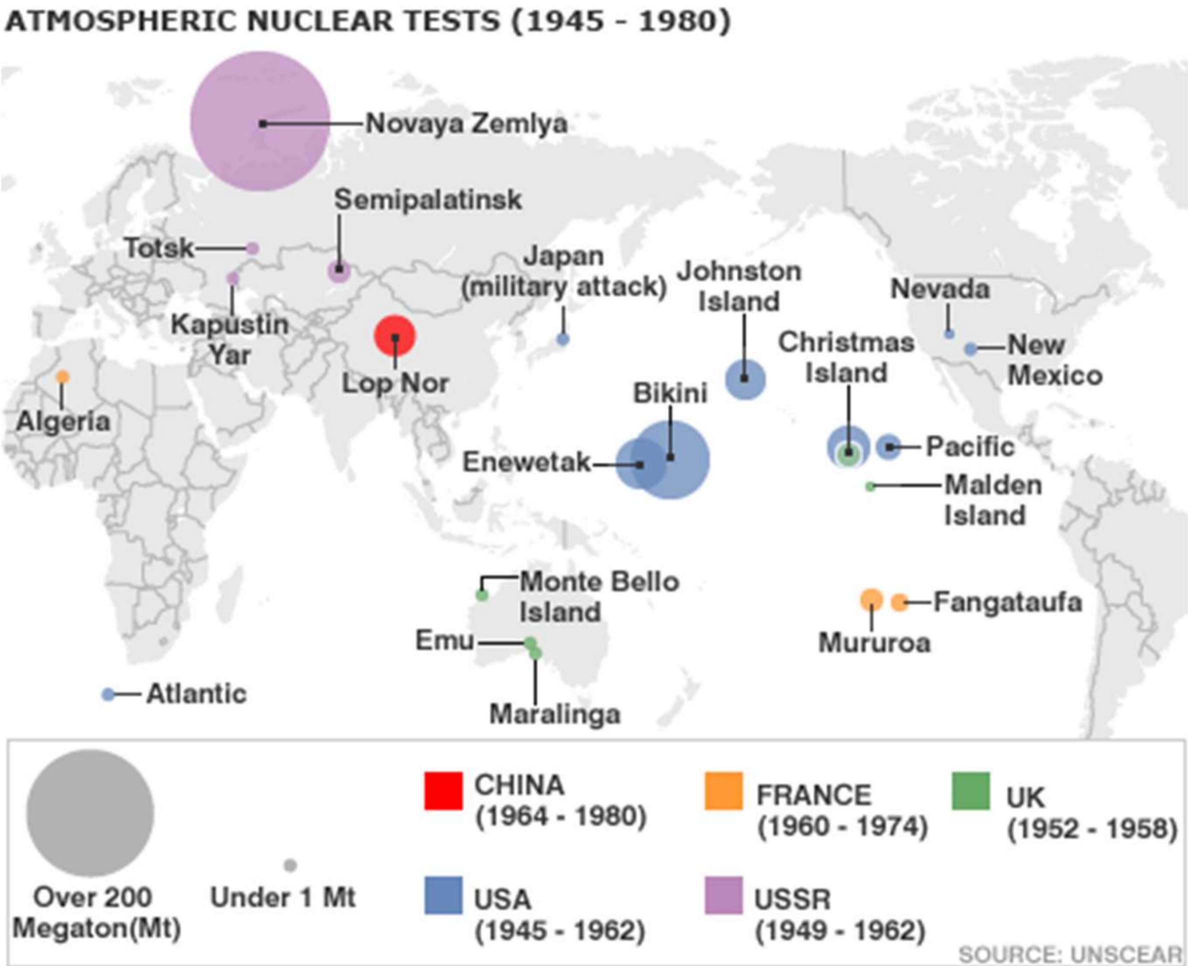
In a study of C^{14} variations in nature (*1*), collection of atmospheric carbon dioxide for the measurement of its C^{14} activity commenced in New Zealand on 24 Nov. 1954. The first three results were reported (*1*) in 1955, and the mean value +3.73 percent with respect to the New Zealand wood standard apparently agreed well with Craig's predicted value of +3.68 percent (*2*). There was, however, in these results an indication that the C^{14} content of the atmosphere was increasing; hence, the sampling program was continued to check whether or not there was a seasonal variation or a C^{14} enrichment of the atmosphere by atomic explosions. Nine samples of air have been assayed since 3 June 1955. The results indicate a steady increase in the C^{14} content of the atmosphere. Duplicate samples collected over the period 12 Dec. 1956 to 23 Mar. 1957 have assayed $+7.60 \pm 0.03$ percent and 8.05 ± 0.30 percent with respect to the New Zealand wood standard. Taking the average value +7.83 percent for the C^{14} enrichment of air for the first 3 months of 1957, these results show that the C^{14} content of the atmosphere of the Southern Hemisphere has increased by 4.10 ± 0.5 percent since February 1955.

Rafter and Fergusson, Science, 1957

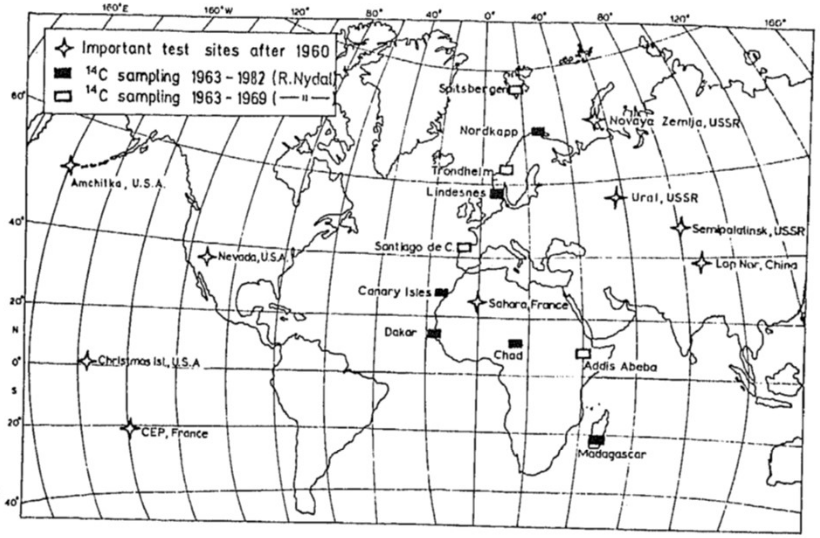
“Bomb Period”: Nuclear weapons testing caused intense production of ^{14}C in the atmosphere

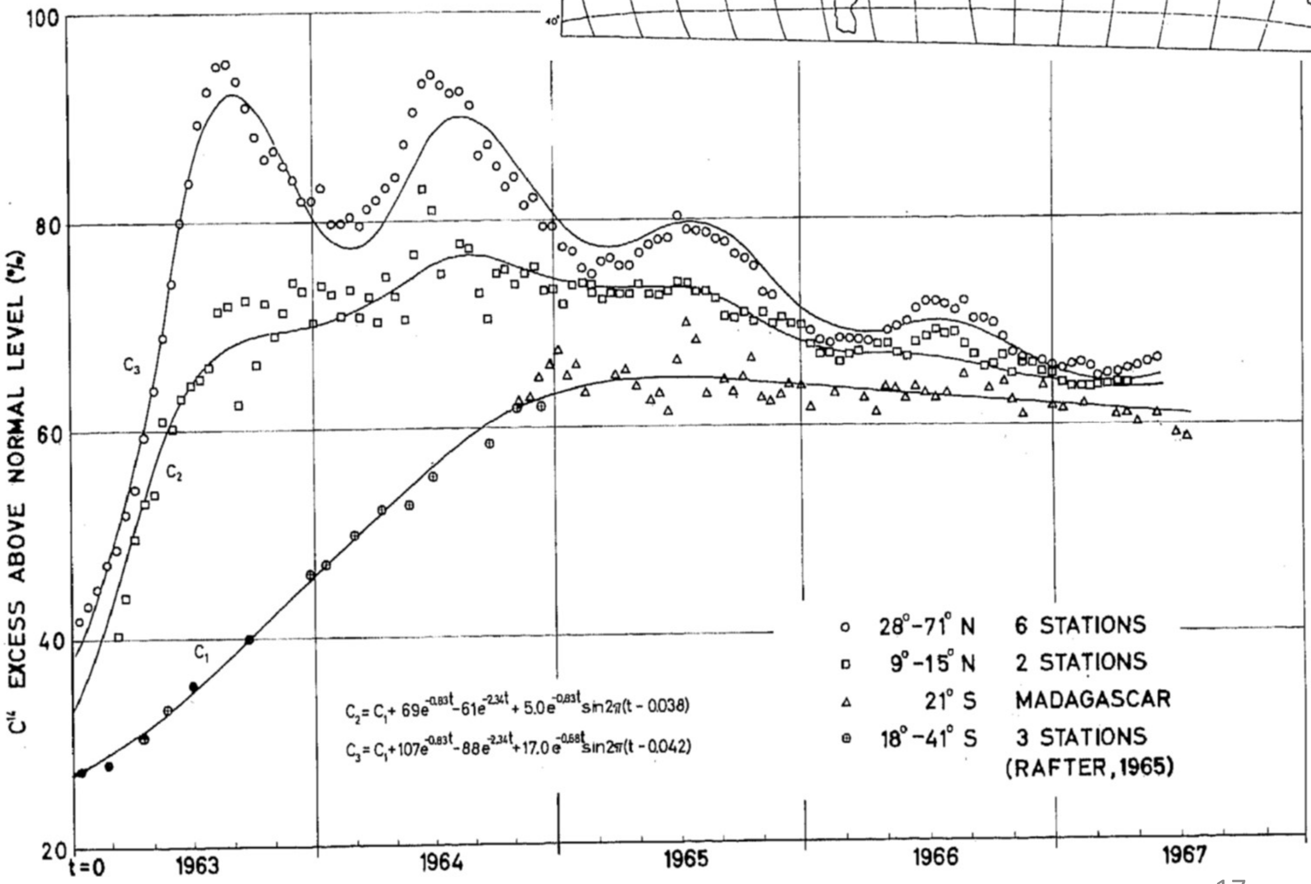


Hua & Barbetti, 2007; Manning et al., 1990; Levin and Kromer, 2004


ATMOSPHERIC NUCLEAR TESTS (1945 - 1980)
Novaya Zemlya
Semipalatinsk
Totsk
Kapustin Yar
Japan (military attack)
Johnston Island
Nevada
New Mexico
Christmas Island
Lop Nor
Algeria
Bikini
Enewetak
Pacific
Malden Island
Monte Bello Island
Emu
Maralinga
Fangataufa
Mururoa
Atlantic
Over 200 Megaton(Mt)
Under 1 Mt
CHINA (1964 - 1980)
FRANCE (1960 - 1974)
UK (1952 - 1958)
USA (1945 - 1962)
USSR (1949 - 1962)
SOURCE: UNSCEAR

Global network of observations captured atmospheric $\Delta^{14}C$ gradients in the bomb period

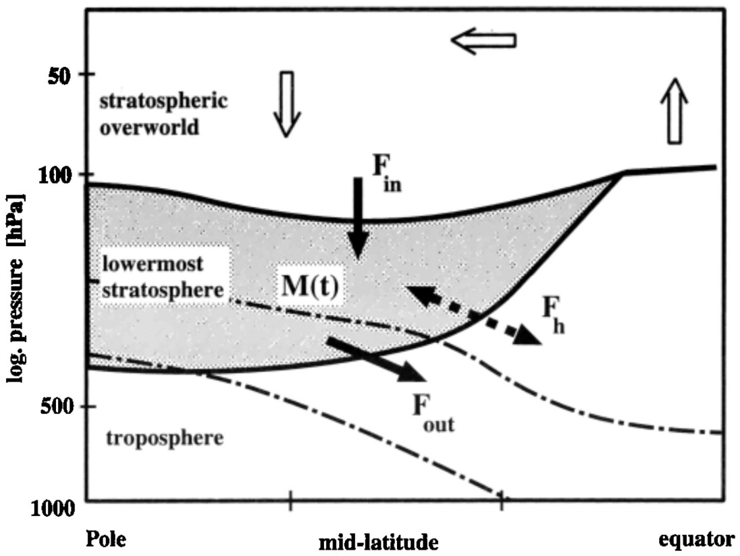


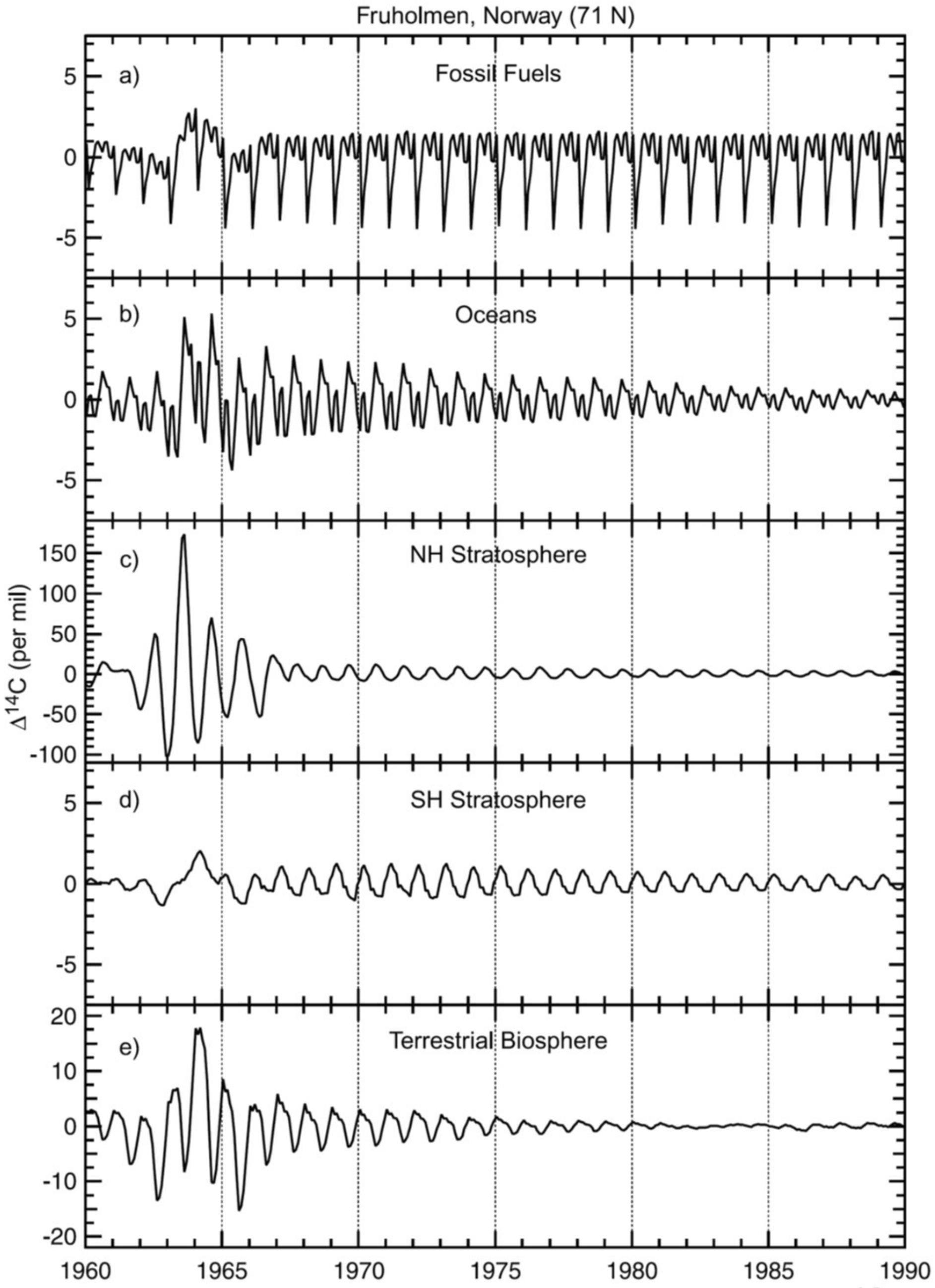


Nydal and Lövseth, 1983

Stratosphere-troposphere exchange caused large Northern Hemisphere seasonal cycles in the early bomb period

Cycles damped slightly by biosphere-atmosphere exchange

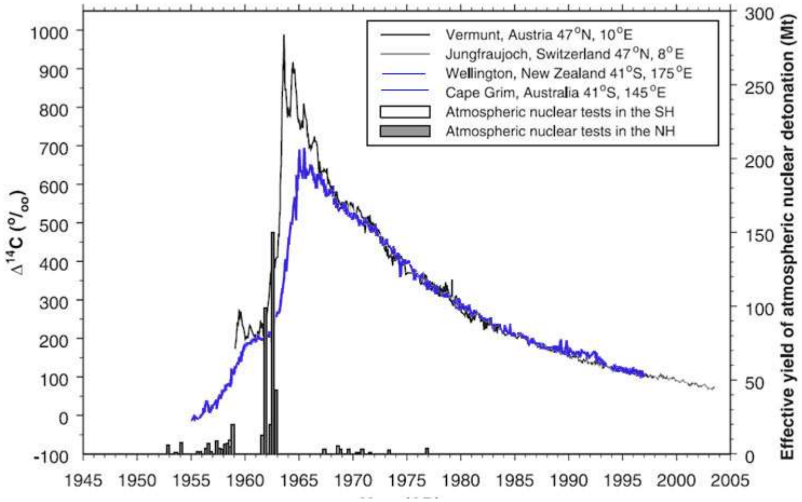




Randerson et al. 2002
Appenzeller et al. 1996

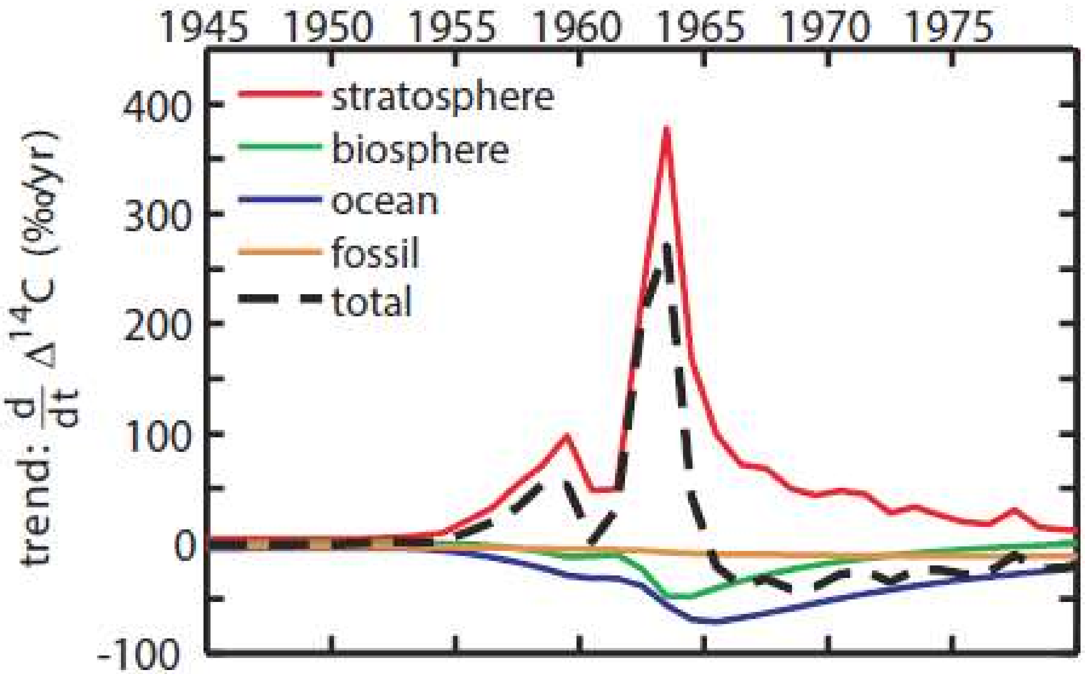
Influences on the $\Delta^{14}CO_2$ trend during the bomb period

$\Delta m - \Delta bg = Cs/Cm\ (\Delta s - \Delta bg)$



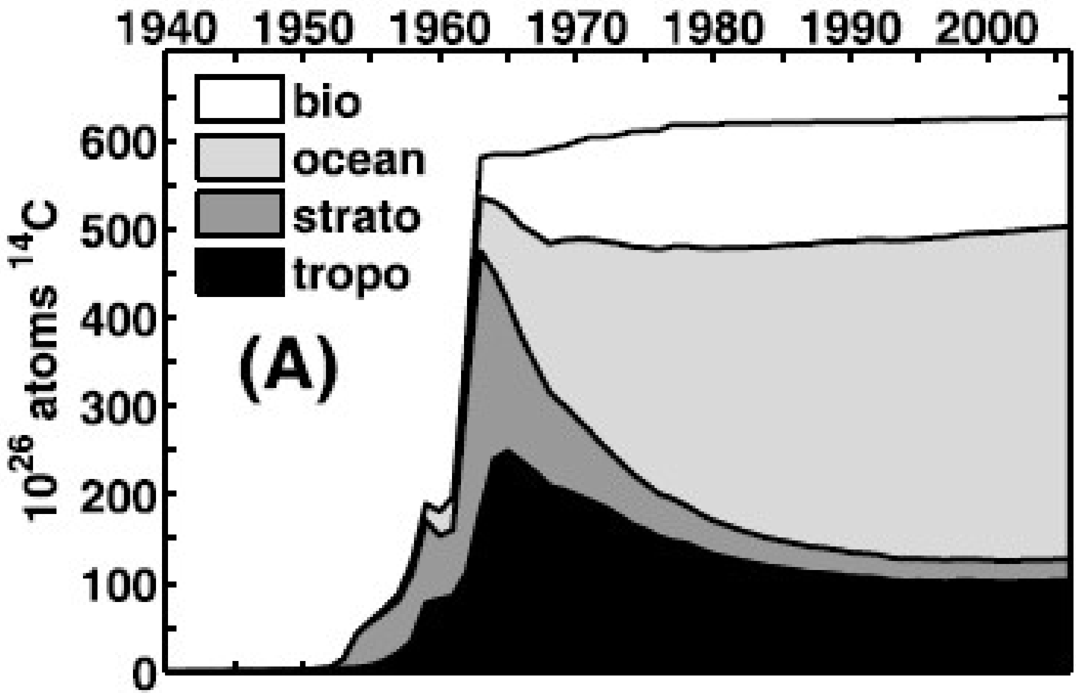
Ocean and biosphere exchange with the atmosphere introduced carbon with lower $\Delta^{14}C$ into the atmosphere

Ocean and biosphere exchanges counteracted and then overpowered the stratospheric influence after the majority of testing ceased



Levin et al. 2010

Inventory of bomb ^{14}C in different reservoirs evolved over time as a result of carbon cycling



Estimates of total 14C produced by bomb strength and number

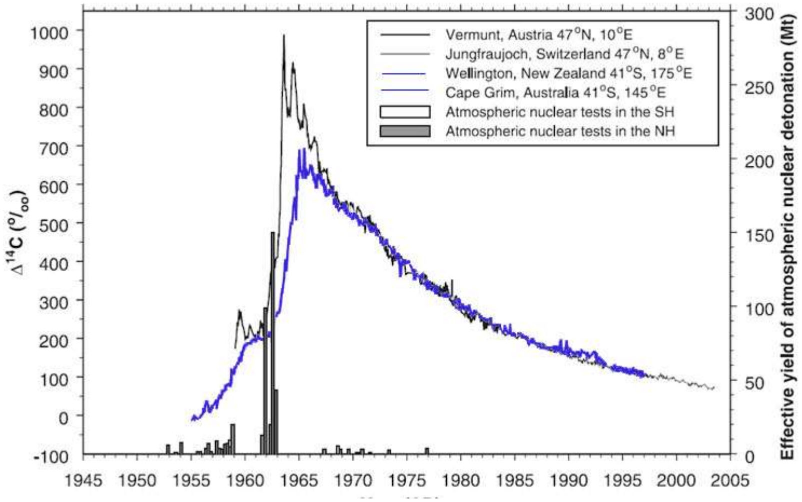
Some ^{14}C is continually emitted from nuclear power plants (about 10% of natural production rate)

Ocean and atmosphere bomb ^{14}C inventories constrained by observations

Biospheric inventory by residual

Naegler and Levin 2009

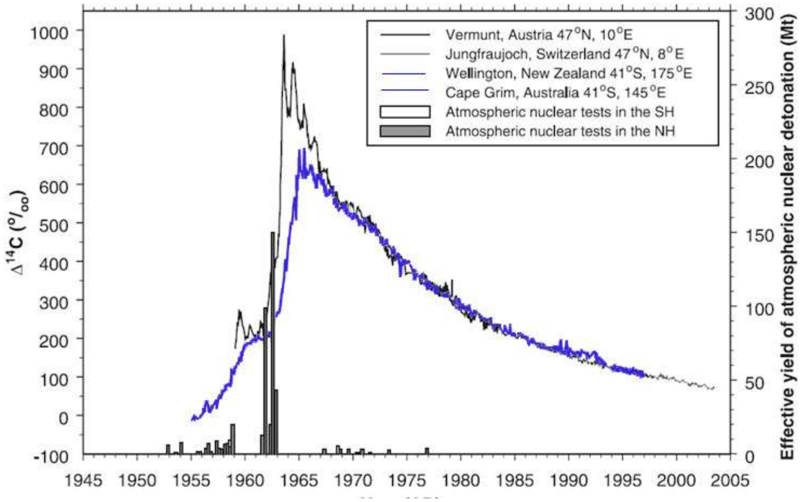
Discussion



What do you think the atmospheric $\Delta^{14}C$ history would have looked like if:

the exchange between the stratosphere and the troposphere was much slower?

Discussion



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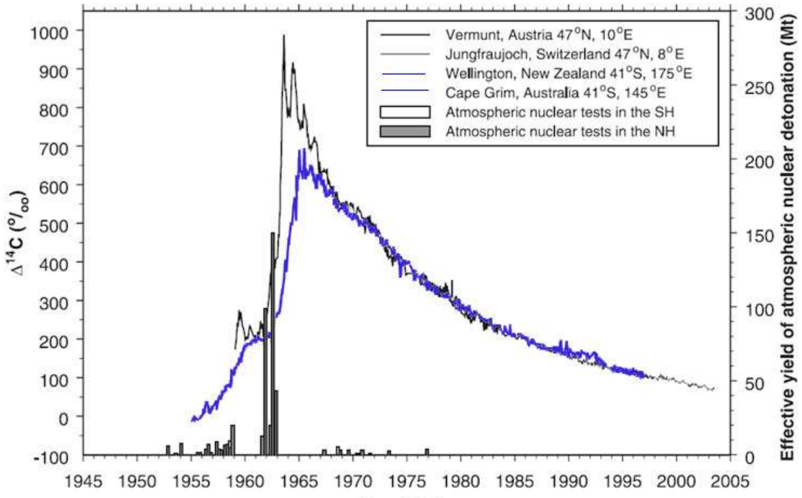
the exchange between the atmosphere and the ocean was much faster?

Discussion



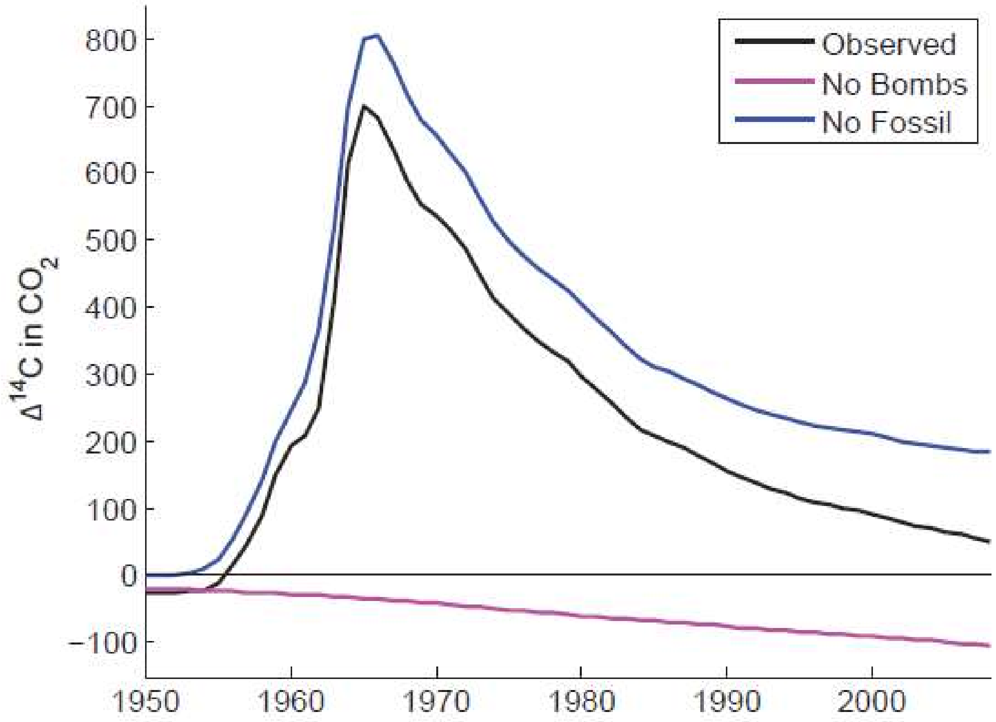
What do you think the atmospheric $\Delta^{14}C$ history would have looked like if:

the exchange between the stratosphere and the troposphere was much slower?

the exchange between the atmosphere and the ocean was much faster?

there were no fossil fuel emissions, but there were nuclear weapons tests?

Imagine if there were no fossil fuel emissions, but there were nuclear weapons tests - what do you think the atmospheric $\Delta^{14}C$ history would have looked like?



Simulations using simple carbon cycle model

Scenario w/o fossil fuel emissions:

- Similar to observed trend through 1980s
- Weaker trend than observed after 1990

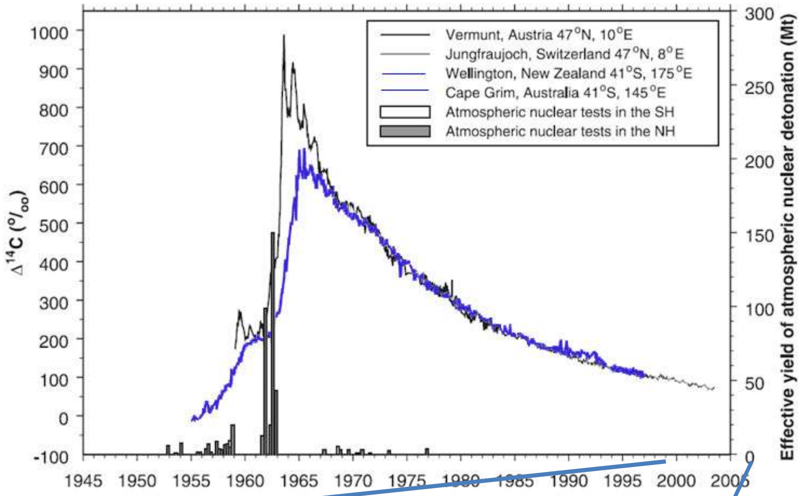
Scenario w/o bomb testing:

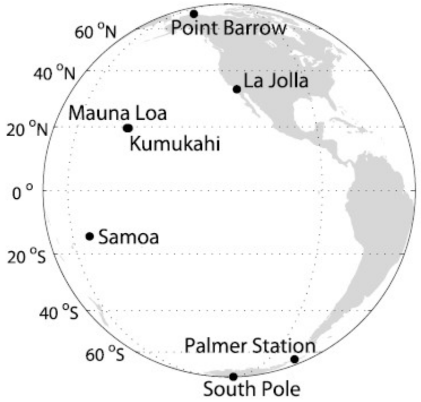
- Indicates atmospheric $\Delta^{14}C$ would now have been -100 ‰ due to fossil fuel emissions

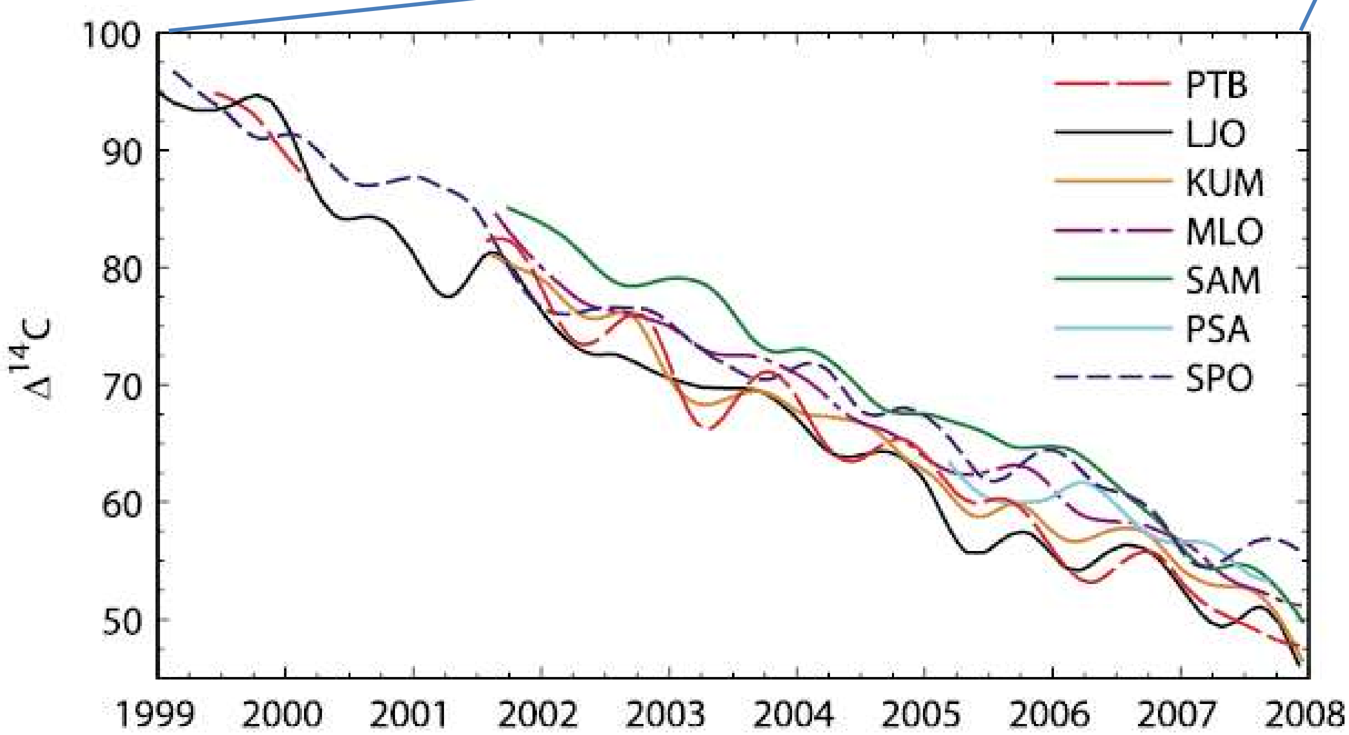
Graven, unpublished

Observations show that $\Delta^{14}C$ is continuing to decline

Decreasing by ~5 ‰ per year
Smaller gradients and cycles

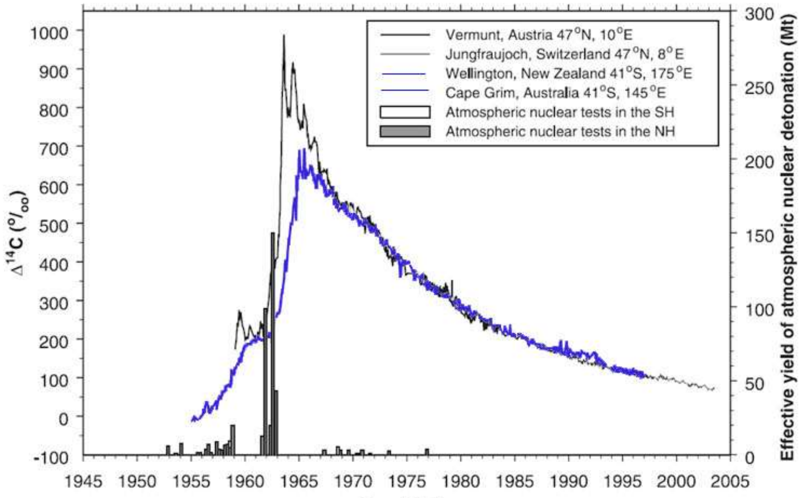






Graven et al. 2012

Influences on the $\Delta^{14}CO_2$ trend during the post-bomb period

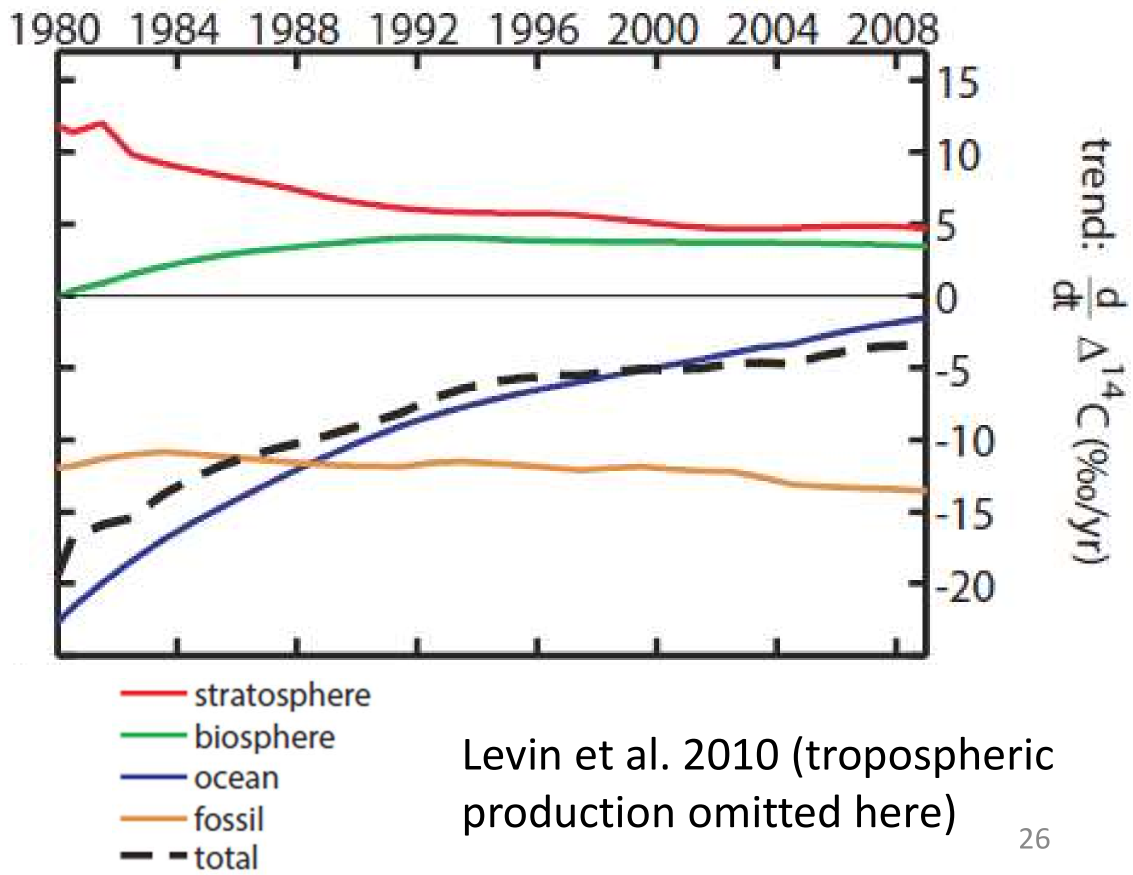


$\Delta m - \Delta bg = Cs/Cm\ (\Delta s - \Delta bg)$

Stratosphere and biosphere exchange have recently had relatively steady positive influences

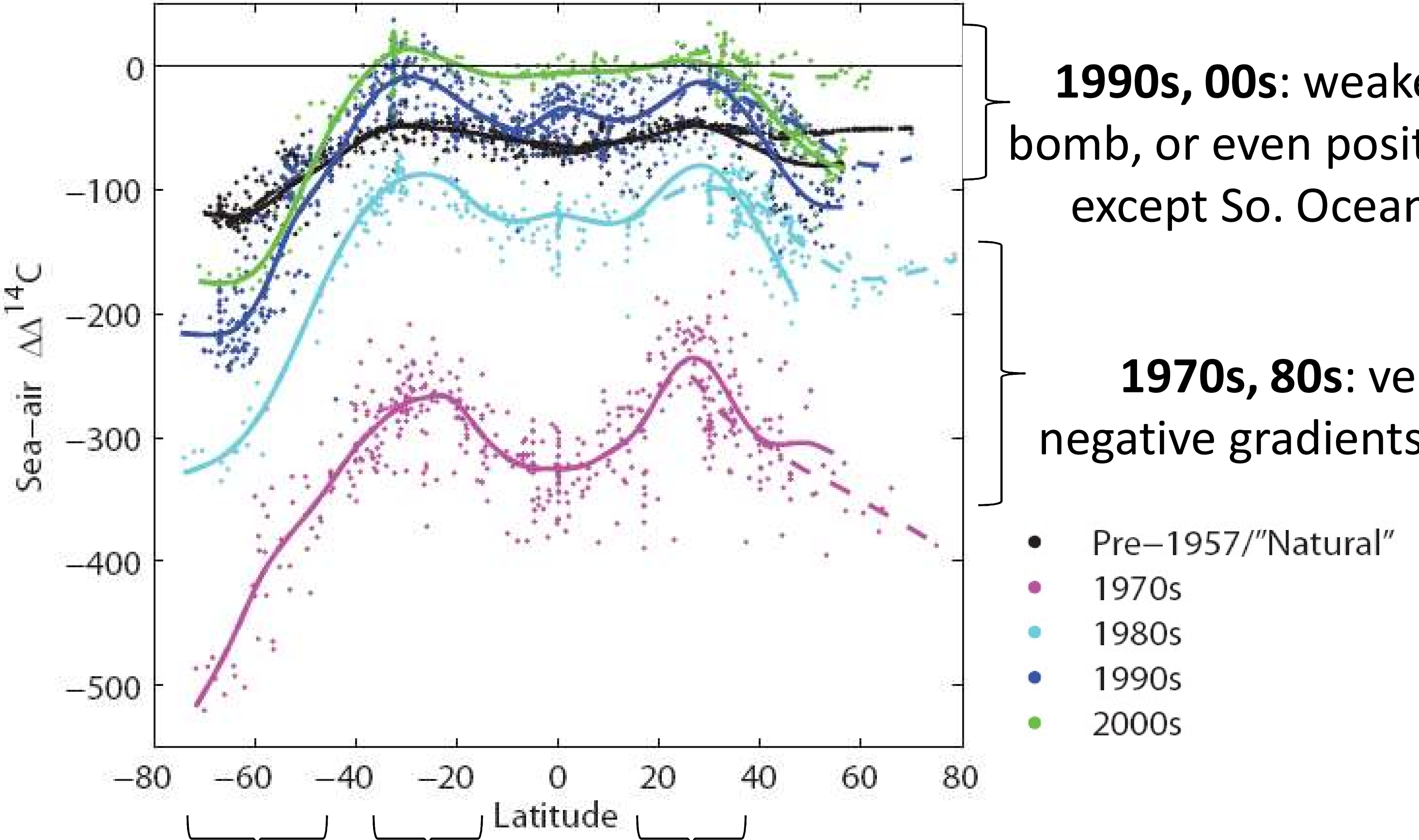
Ocean exchange strongly weakened

Fossil fuel emissions are now the largest influence on the $\Delta^{14}CO_2$ trend



Levin et al. 2010 (tropospheric production omitted here)

Observed sea-air $\Delta^{14}C$ gradients show switch to ^{14}C outgassing



1990s, 00s: weaker than pre-bomb, or even positive gradients, except So. Ocean, N. Pacific

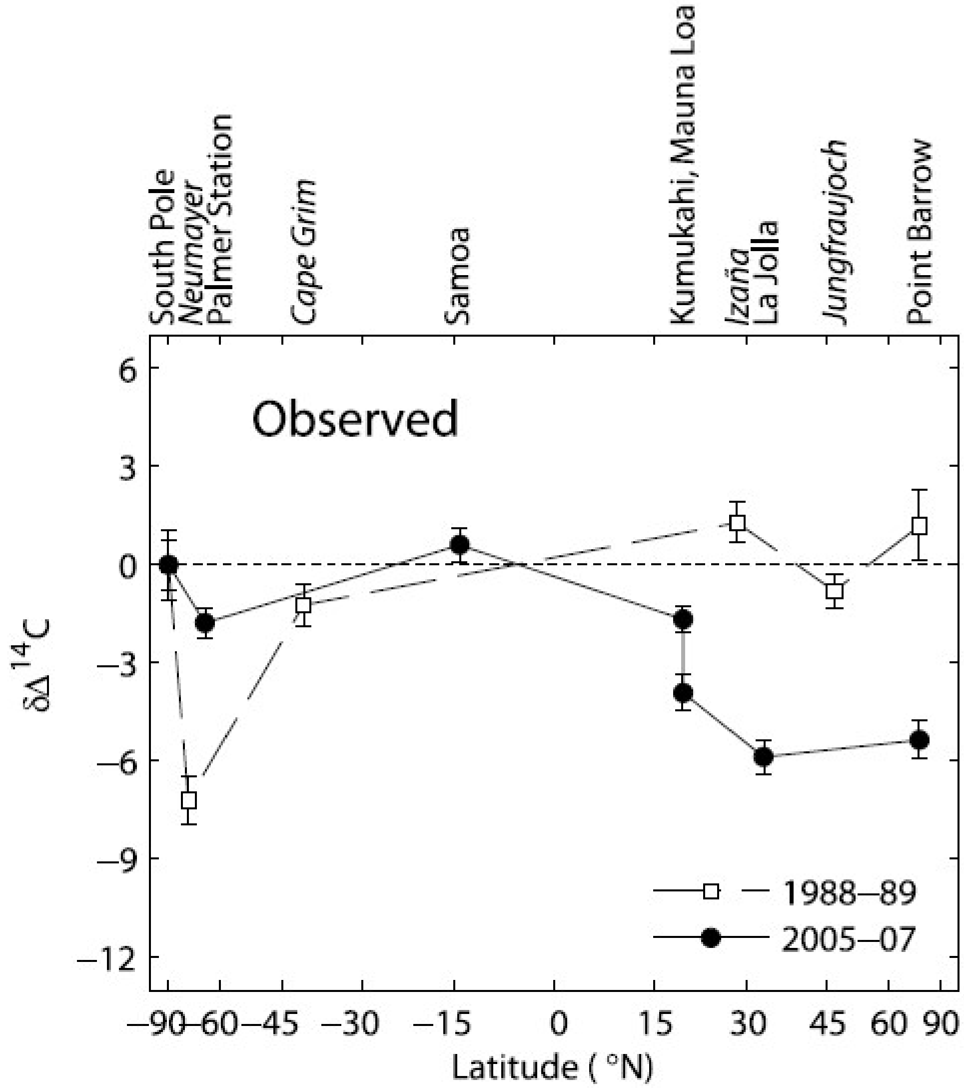
1970s, 80s: very strong negative gradients everywhere

Southern Ocean
Strong mixing with deep, old water

Subtropical Gyres
Weak mixing with deep water

Linick 1978; Fairhall 1985; Broecker 1985; Nydal 1998; Key 2004 and many others

Observed recent shift in North-South gradient



Δ^{14}C relative to the South Pole in 1988-89 and 2005-07

Main influences on gradient:

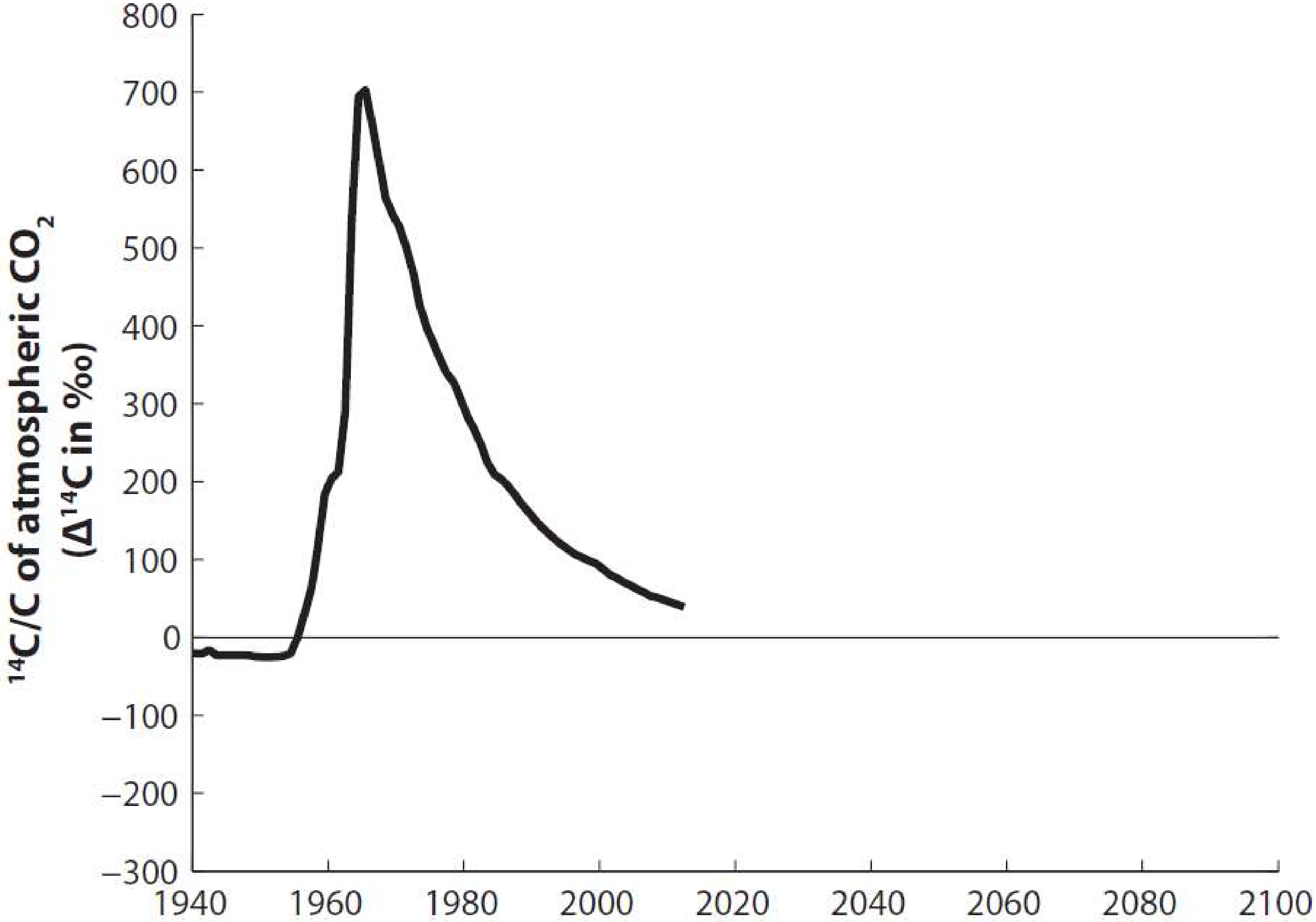
- **Fossil fuel emissions in the North**
- **Ocean exchange in the South**

Shift indicates change in relative strength of fossil fuel and ocean influences

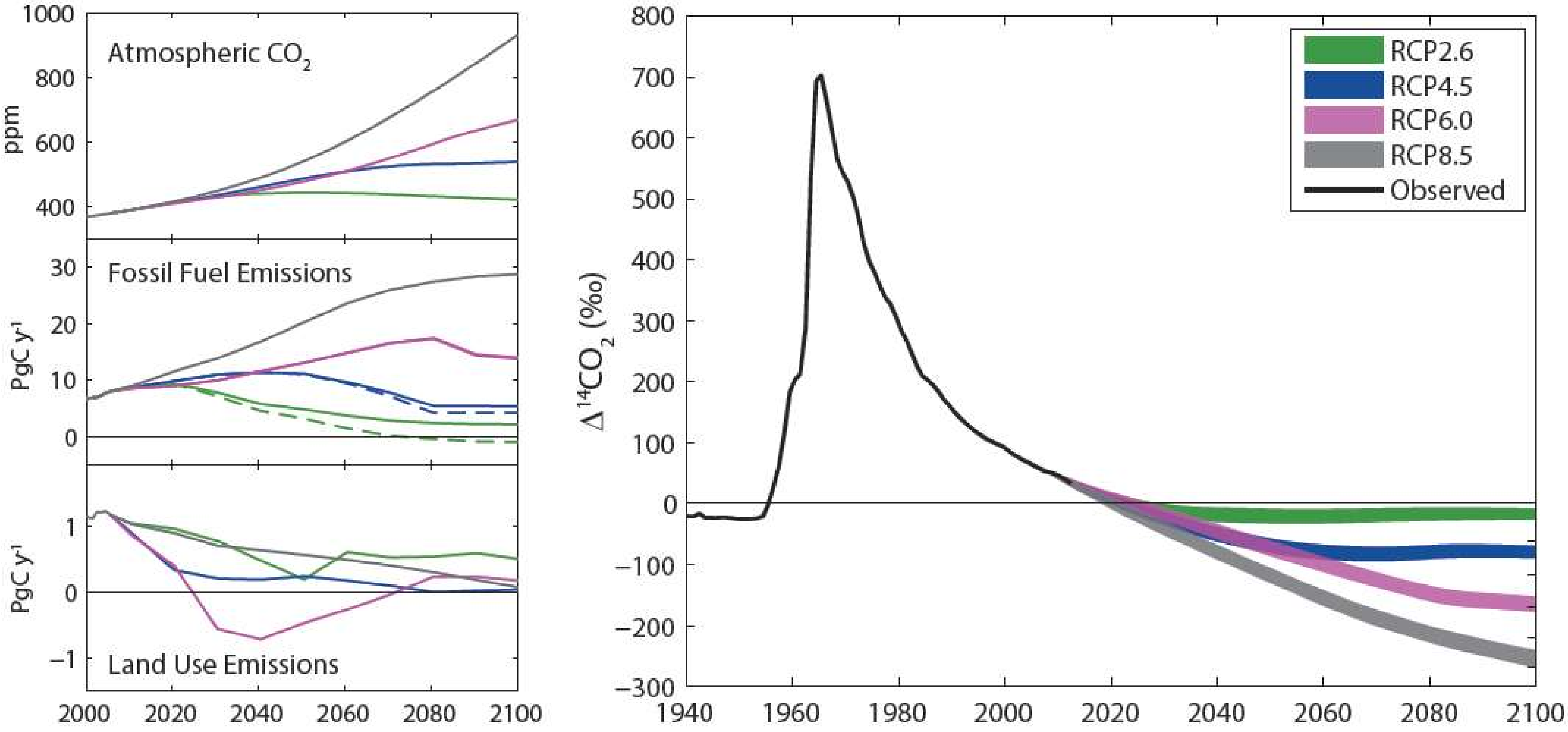
Gradient is now opposite to preindustrial gradient

Graven et al. 2012bc; Levin et al. 2010; Meijer et al. 2006

How will atmospheric $\Delta^{14}C$ evolve over this century?



A wide range of $\Delta^{14}CO_2$ trajectories are simulated using the RCPs and a simple carbon cycle model



Range of emission scenarios – Representative Concentration Pathways (RCPs) from IPCC AR5

Graven, PNAS, 2015

Model simulations of future ocean $\Delta^{14}C$

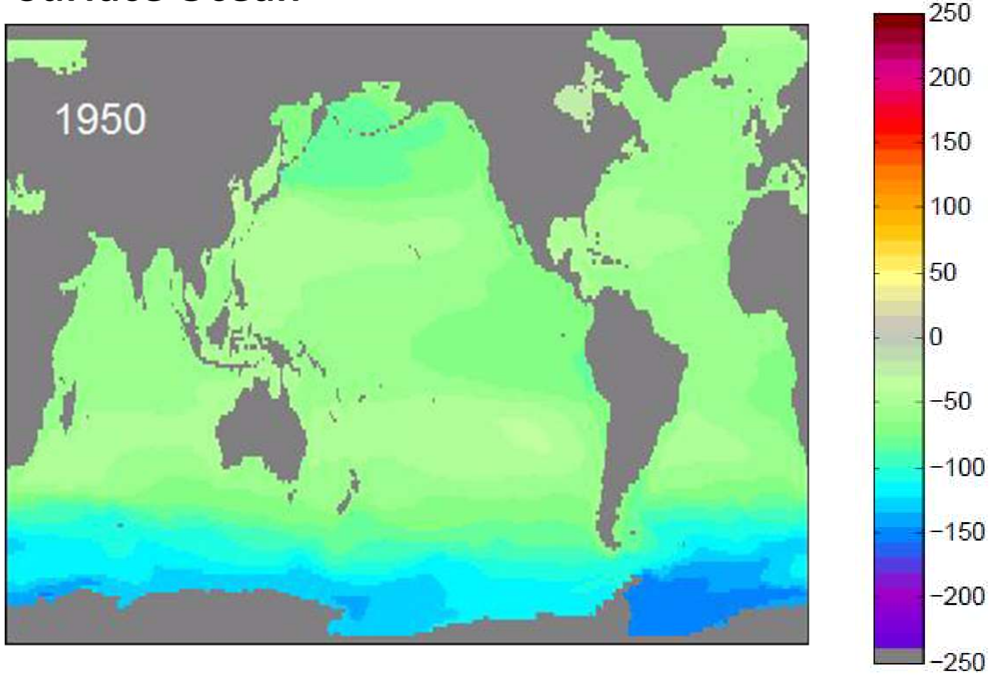
ECCO ocean model forced with atmospheric $\Delta^{14}C$ observations and projections (Khatiwala 2007; Graven et al. 2012)

Constant ocean circulation

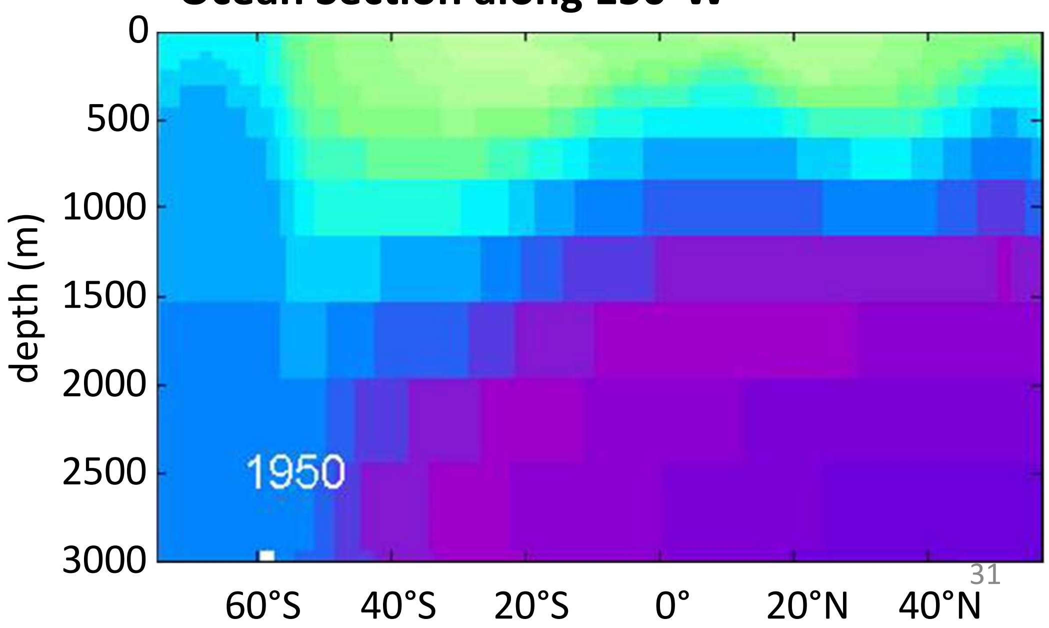
Gas exchange velocity of 15 cm/hr (Sweeney et al. 2007)

Khatiwala et al., in rev.

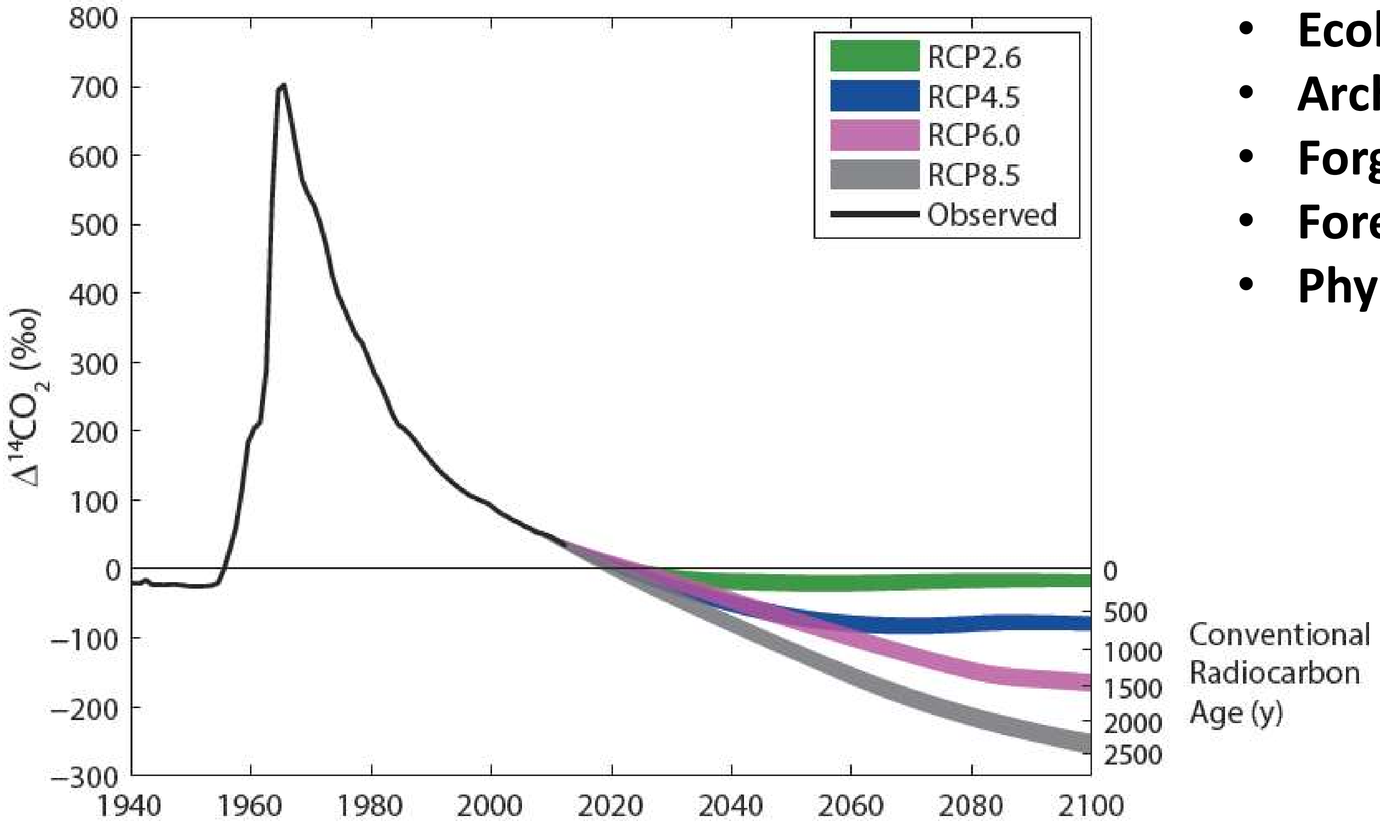
Surface Ocean



Ocean Section along 150°W



Forthcoming changes in $\Delta^{14}CO_2$ have implications for various applications of ^{14}C



- **Ecology**
- **Archaeology**
- **Forgery detection**
- **Forensics**
- **Physiology**

Graven, PNAS, 2015

Summary

Nuclear and fossil fuel perturbations have caused dramatic changes to $\Delta^{14}C$ in atmospheric CO_2

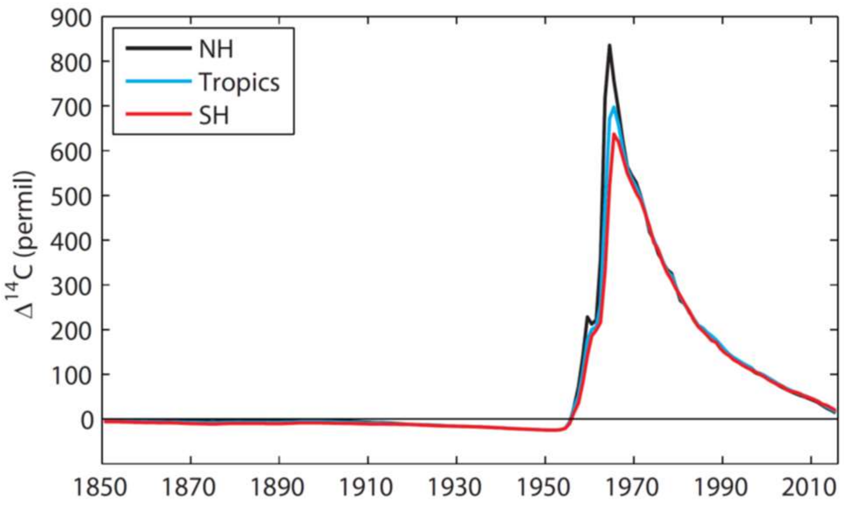
Disequilibria with the stratosphere, ocean and biosphere have evolved over time and, in some cases, changed sign

Now that several decades have past since the bomb tests, fossil fuel emissions are becoming more and more dominant in driving $\Delta^{14}C$ trends and gradients

$\Delta^{14}C$ in atmospheric CO_2 will reach 0 ‰ in the next couple of years, and depending on fossil fuel emissions, may decline to -250 ‰ by the end of the century, impacting various ^{14}C applications

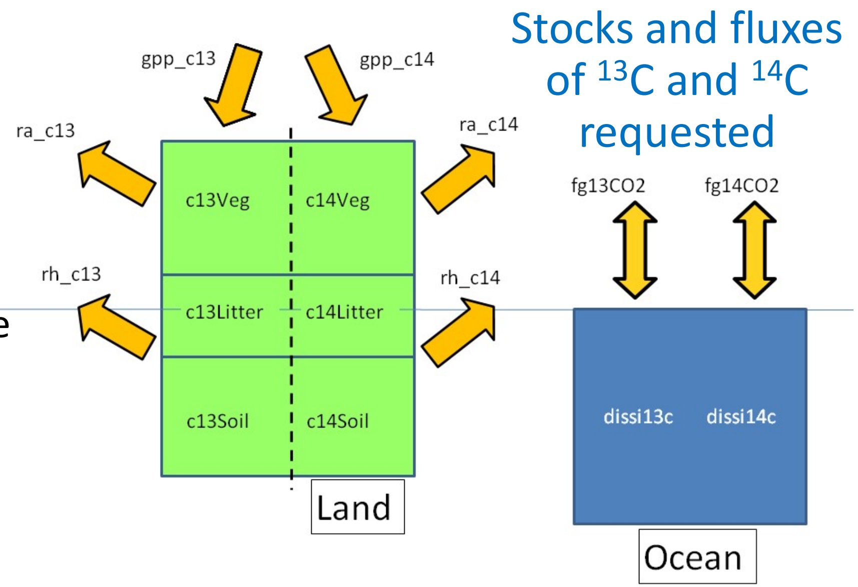
Carbon isotopes in CMIP6

Motivation:
- **Improved understanding of carbon cycle processes**
- **Model evaluation**



We are providing historical and future forcing data for atmospheric $\delta^{13}CO_2$ and $\Delta^{14}CO_2$

Data available at
https://esgf-node.llnl.gov/search/input4mips/
Graven et al. GMDD 2017



Ocean-only simulations: OMIP (Ocean MIP)
Historical ocean-only simulations with observed climate
Orr et al. GMD 2017

Land and Ocean simulations:
C4MIP (Coupled Climate-Carbon Cycle MIP)
Historical and future simulations with coupled climate
Jones et al. GMD 2016