UNIVERSITÄT BERN

OESCHGER CENTRE CLIMATE CHANGE RESEARCH

Radiocarbon (^{14}C)-based source apportionment of atmospheric aerosols

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Aerosol pollutions

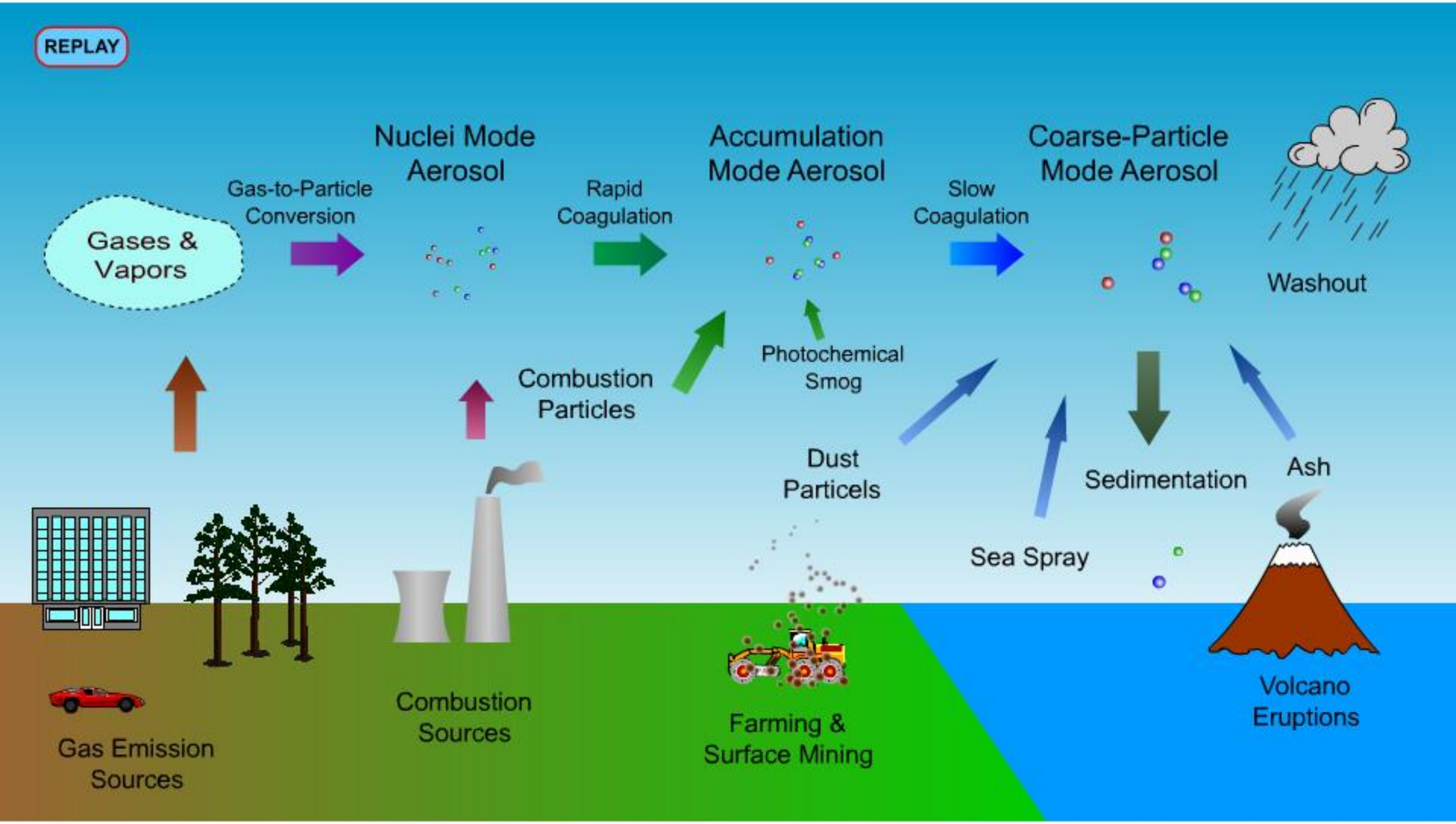
The smog in London 1952

The smog in London 2009

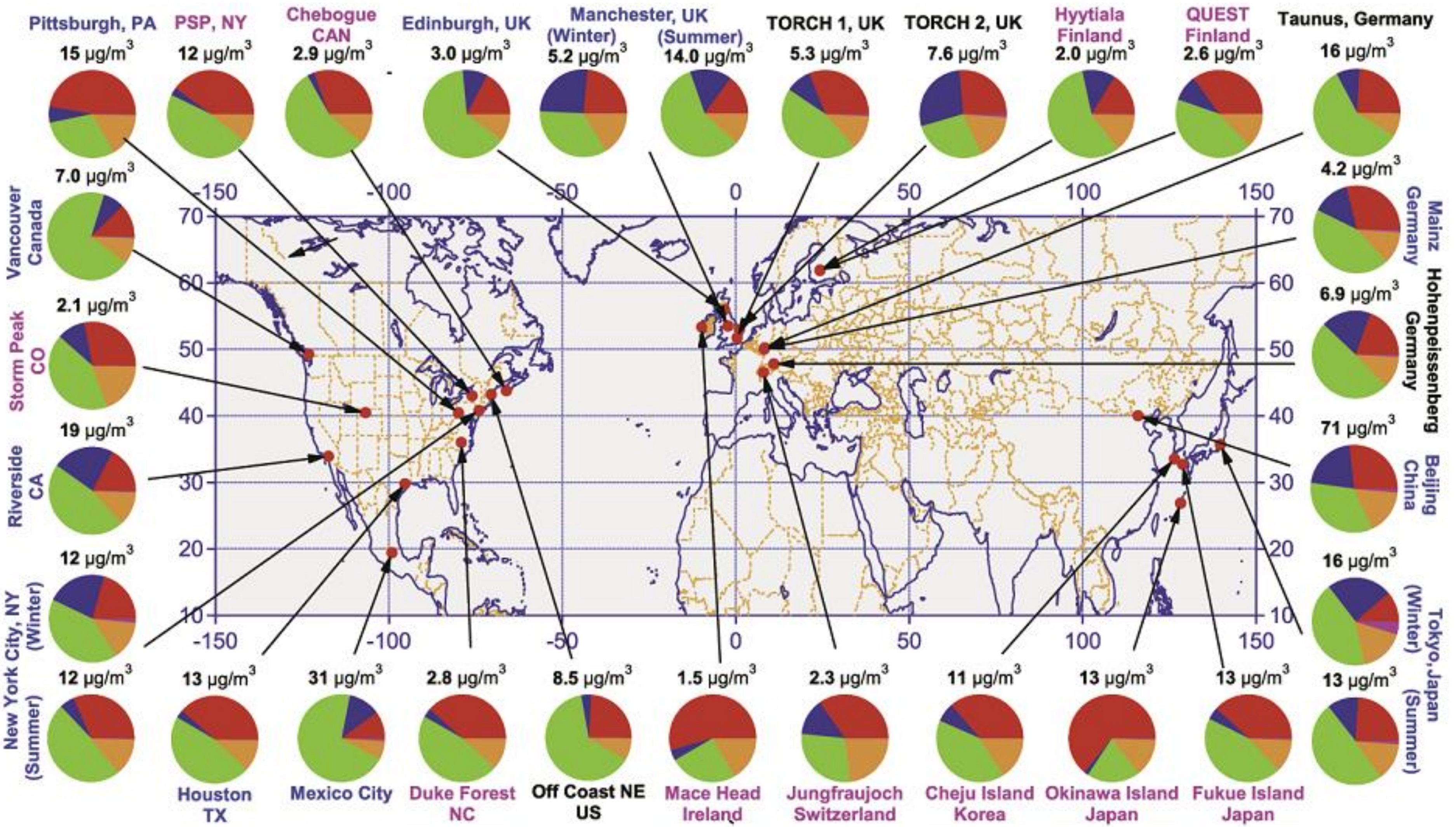
The smog in China 2013

The smog in Arctic

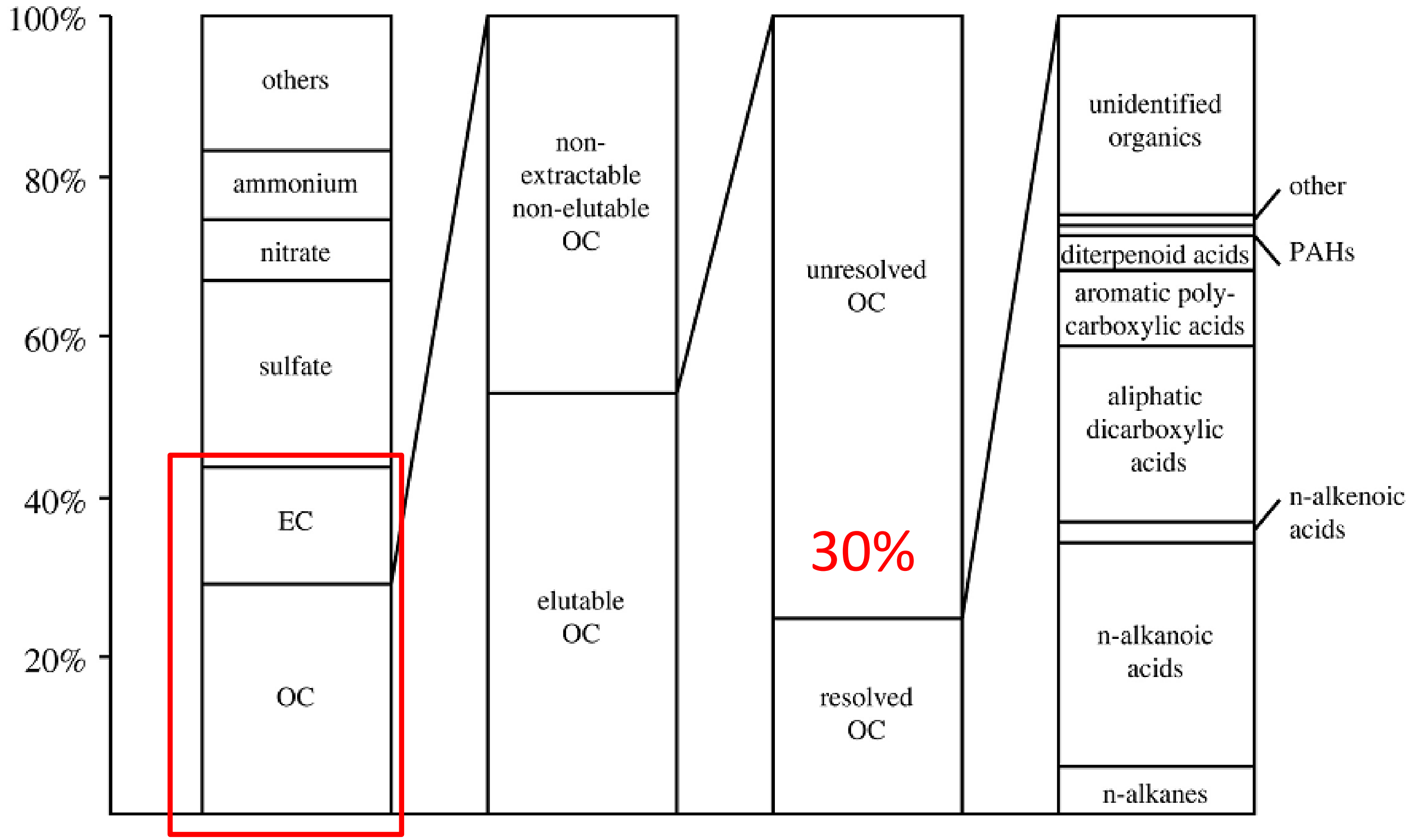
Radiocarbon (14C)-based source apportionment of atmospheric aerosols



Chemical composition of PM1 measured by AMS (Zhang et al., 2007).

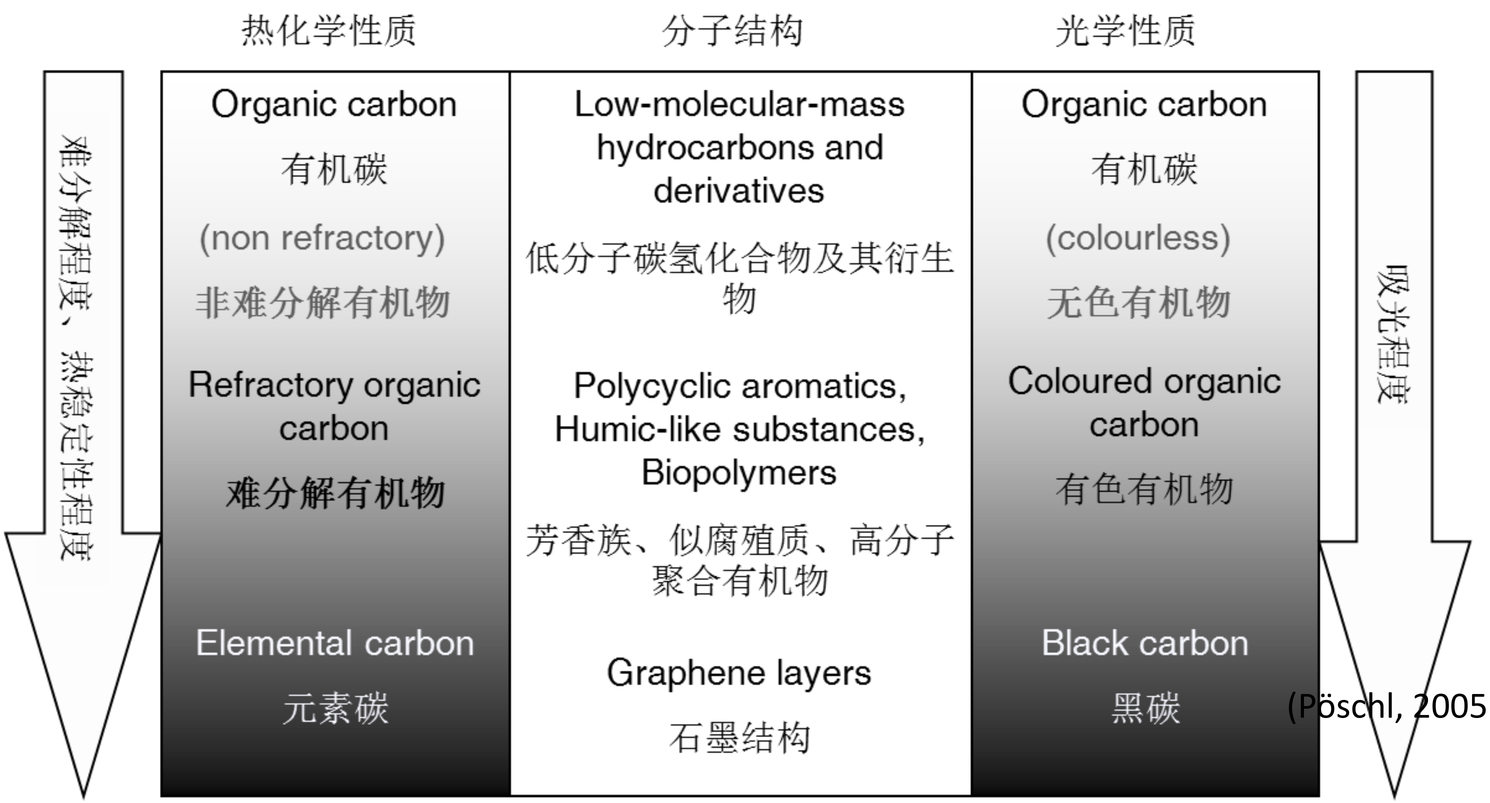


Typical chemical composition in aerosols by offline analytic method method

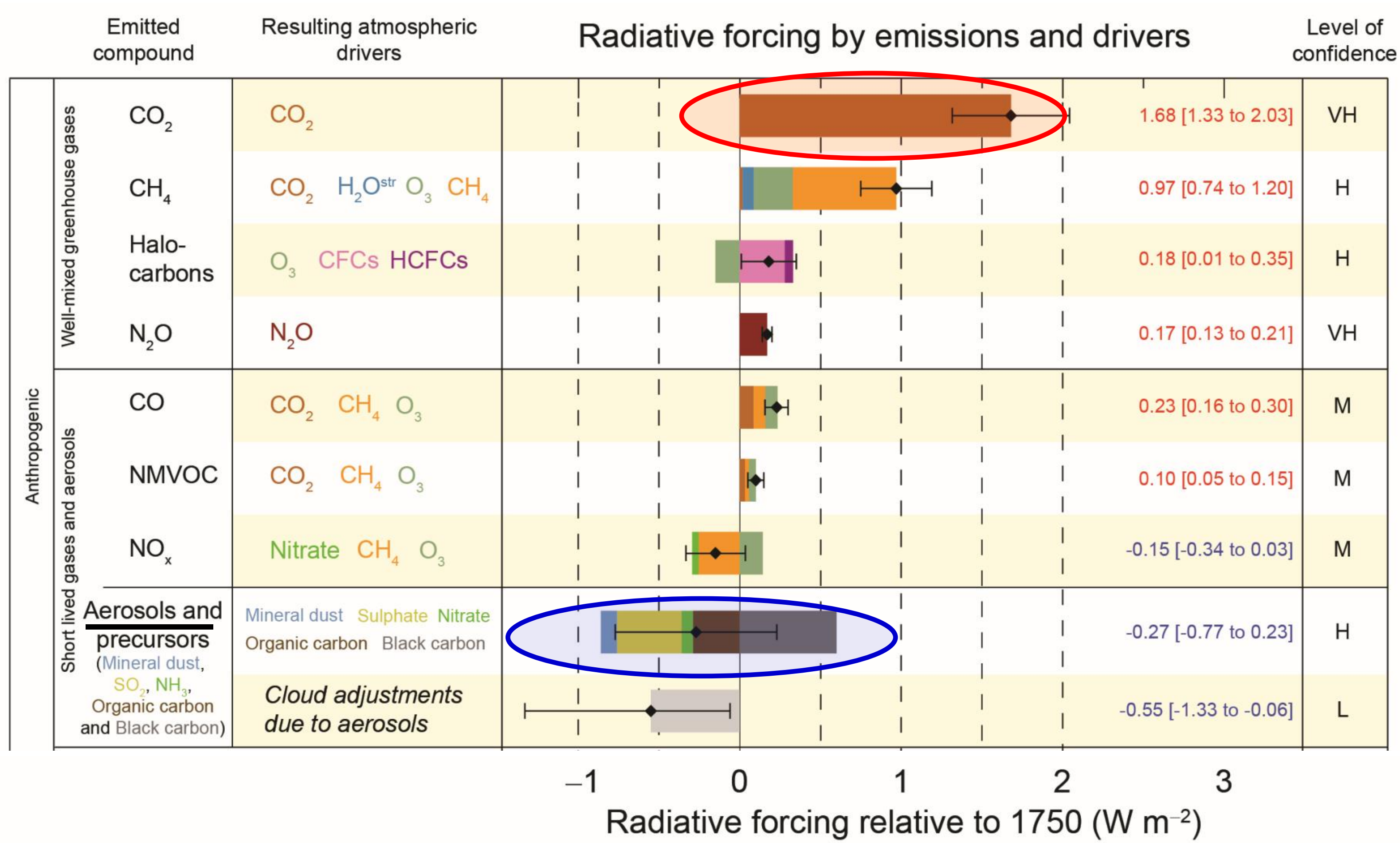


Rogge et al., 1993

OC and EC

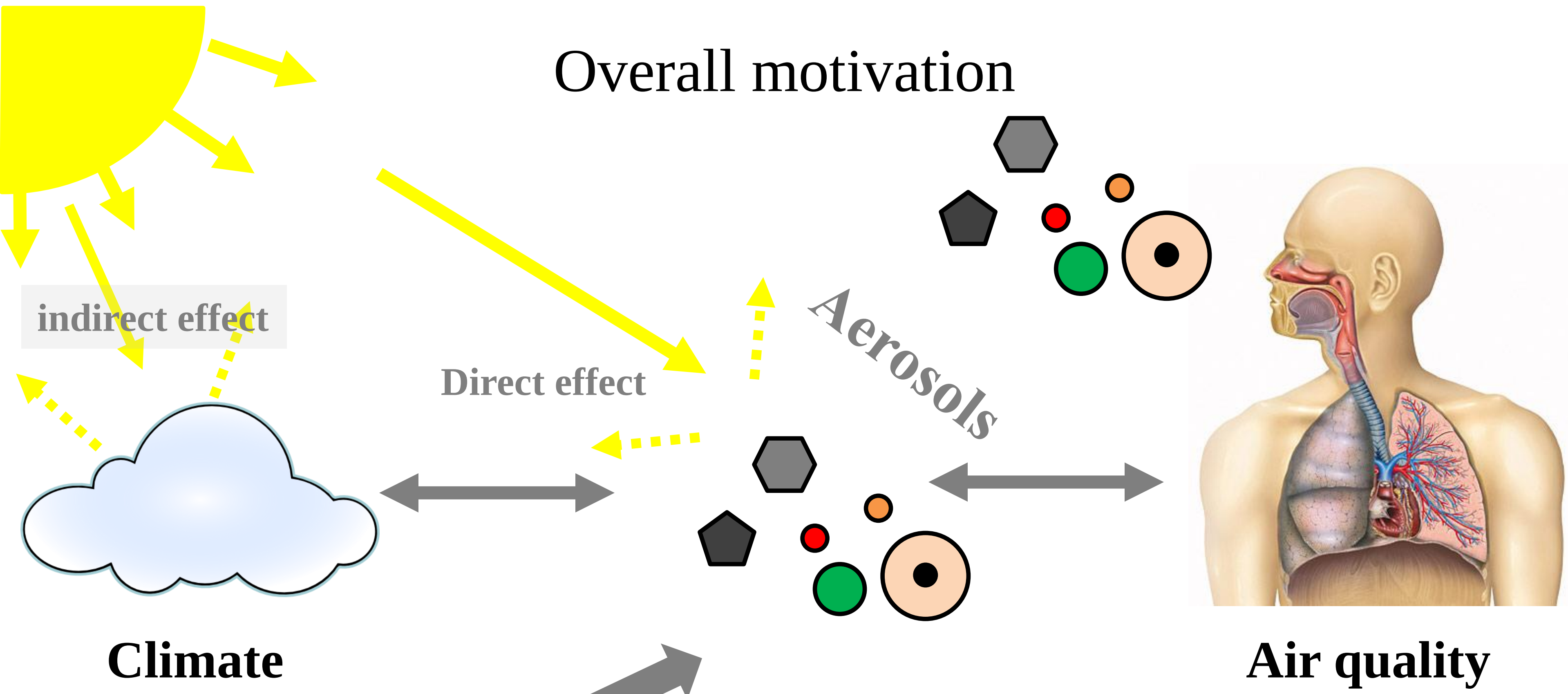

热化学性质
分子结构
光学性质
难分解程度、热稳定性程度
Organic carbon
有机碳
(non refractory)
非难分解有机物
Refractory organic carbon
难分解有机物
Elemental carbon
元素碳
Low-molecular-mass hydrocarbons and derivatives
低分子碳氢化合物及其衍生物
Polycyclic aromatics, Humic-like substances, Biopolymers
芳香族、似腐殖质、高分子聚合有机物
Graphene layers
石墨结构
Organic carbon
有机碳
(colourless)
无色有机物
Coloured organic carbon
有色有机物
Black carbon
黑碳
吸光程度
(Pöschl, 2005)

Climate effect of aerosols



Intergovernmental Panel on Climate Change (IPCC), 2014

Overall motivation



The characterization of sources of aerosols is a key step for a better implementation of effective abatement and mitigation strategies.

^{14}C: 判断化石和生物来源的最有效的工具

Brown Clouds over South Asia: Biomass or Fossil Fuel Combustion?

Science

SCIENCE VOL 323 23 JANUARY 2009

Örjan Gustafsson,[1*] Martin Kruså,[1] Zdenek Zencak,[1] Rebecca J. Sheesley,[1] Lennart Granat,[2] Erik Engström,[2] P. S. Praveen,[3] P. S. P. Rao,[4] Caroline Leck,[2] Henning Rodhe[2]

Carbonace
important
understood
emissions,
We used ra
Ocean to
aerosols, a
constraints
burning) a
quality.

ATMOSPHERE

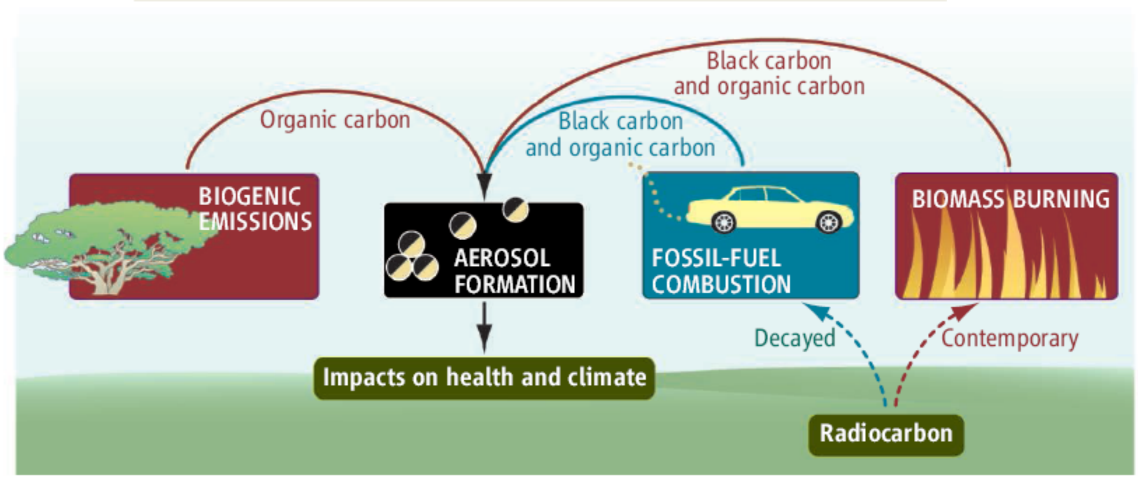
Sources of Asian Haze

Sönke Szidat

Radiocarbon analysis elucidates the sources of the pollutants responsible for the "brown clouds" over South Asia.

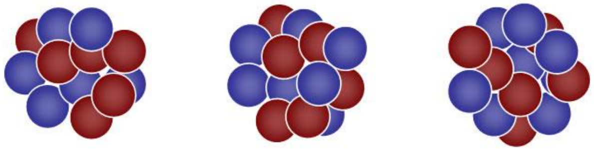
23 JANUARY 2009 VOL 323 SCIENCE

Carbonaceous aerosols—that is, the carbon-containing aerosol fraction, such as soot—can affect both climate and human health, especially in regions where the atmosphere contains high levels of such particles. Yet, knowledge of the sources of the aerosols is limited. On page 495 of this issue, Gustafsson *et al.* (*1*) use radiocarbon (^{14}C) analysis as an atmospheric tracer to quantify biomass and fossil-fuel contributions to the atmospheric "brown clouds" (*2*) over South Asia, a persistent and large-scale pollution layer of haze. The results resolve a discrepancy between measurements of other atmospheric tracers and calculations of emission-based inventories for carbonaceous aerosols.



Source apportionment. Radiocarbon analysis allows fossil and nonfossil sources of black carbon and organic carbon to be identified. [Adapted from (*12*)]

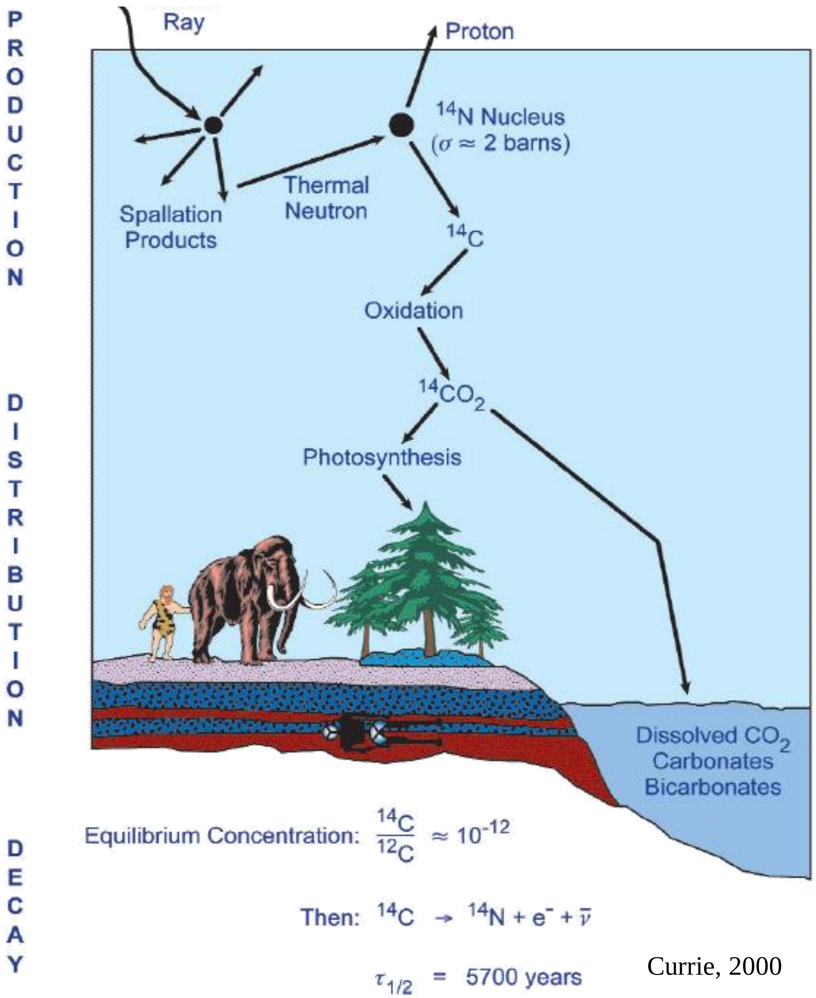
Radiocarbon



carbon-12 98.9% 6 protons 6 neutrons

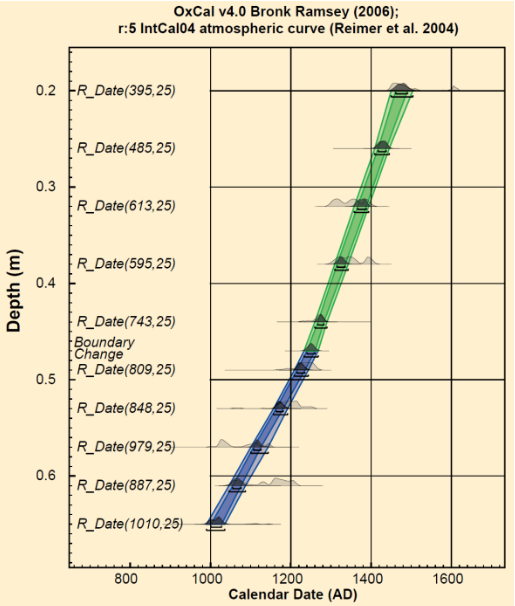
carbon-13 1.1% 6 protons 7 neutrons

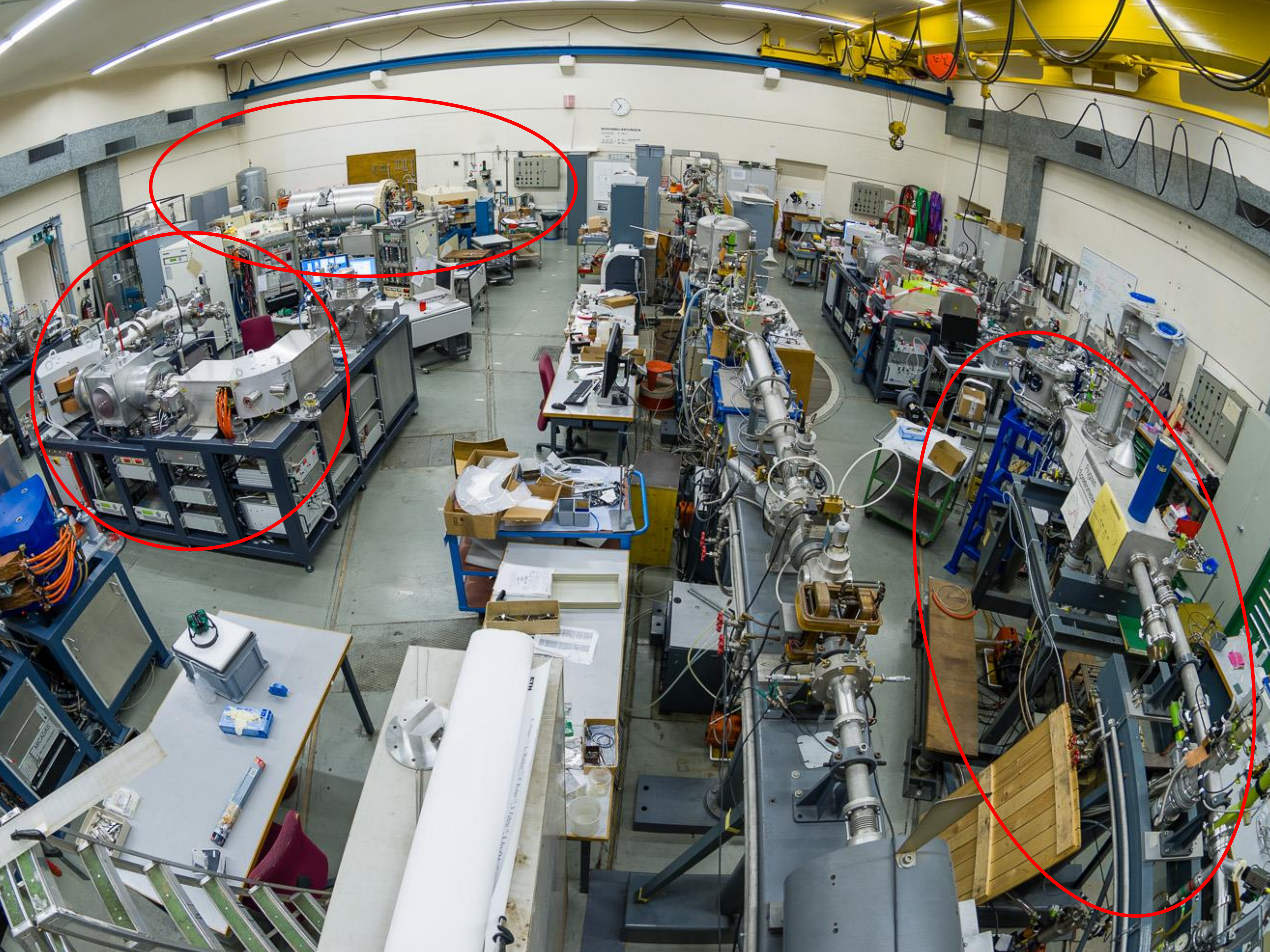
carbon-14 <0.1% 6 protons 8 neutrons



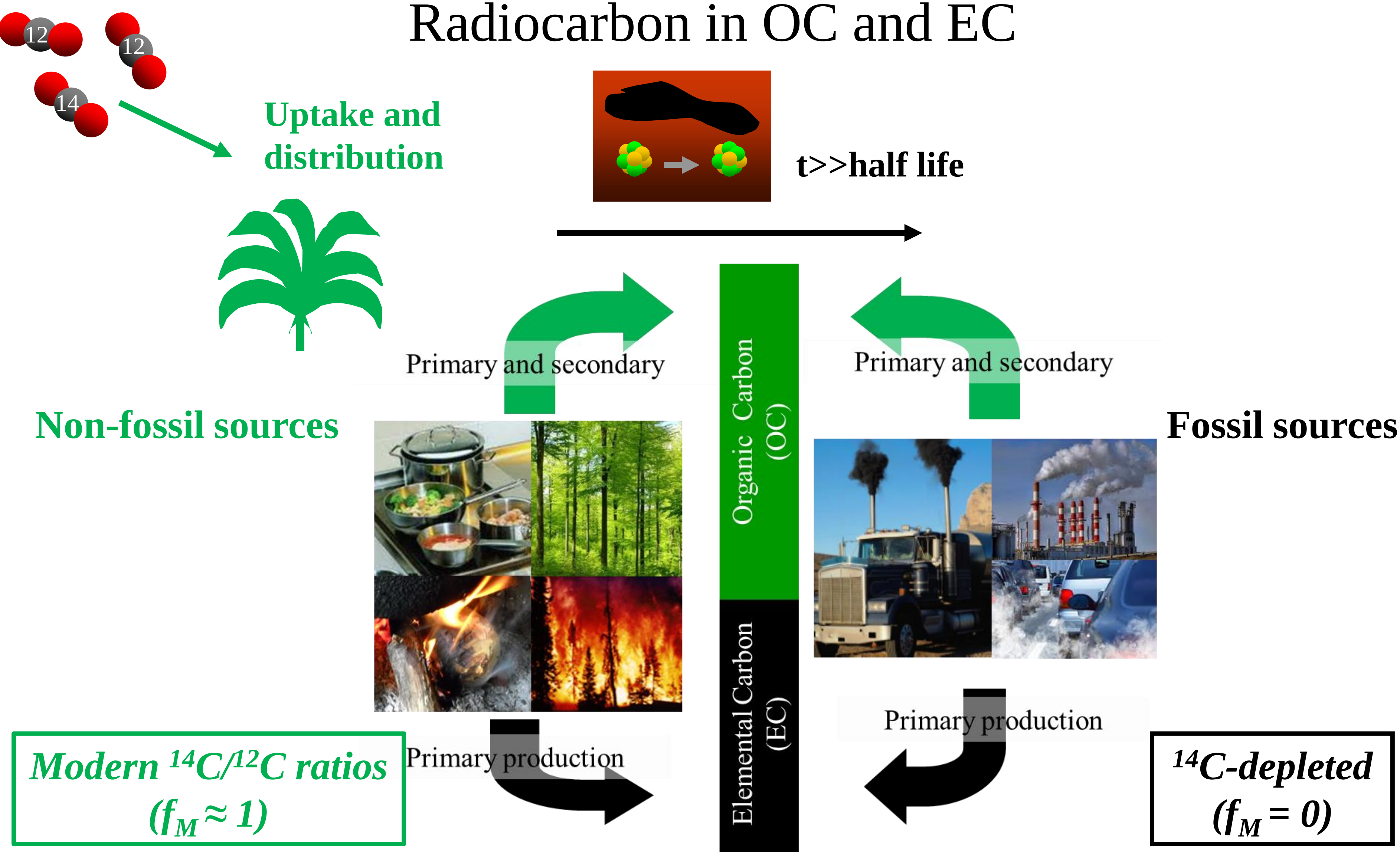
Currie, 2000

Willard Libby





Radiocarbon in OC and EC



Experimental flow

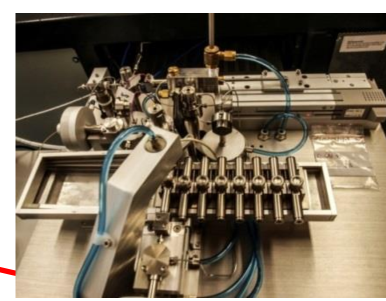
1 **Aerosol Sampling**

2 **OC/EC separation**

3 **CO_2 trapping**

4 **CO_2 ampoule sealing**

5 **Ampoule crack**



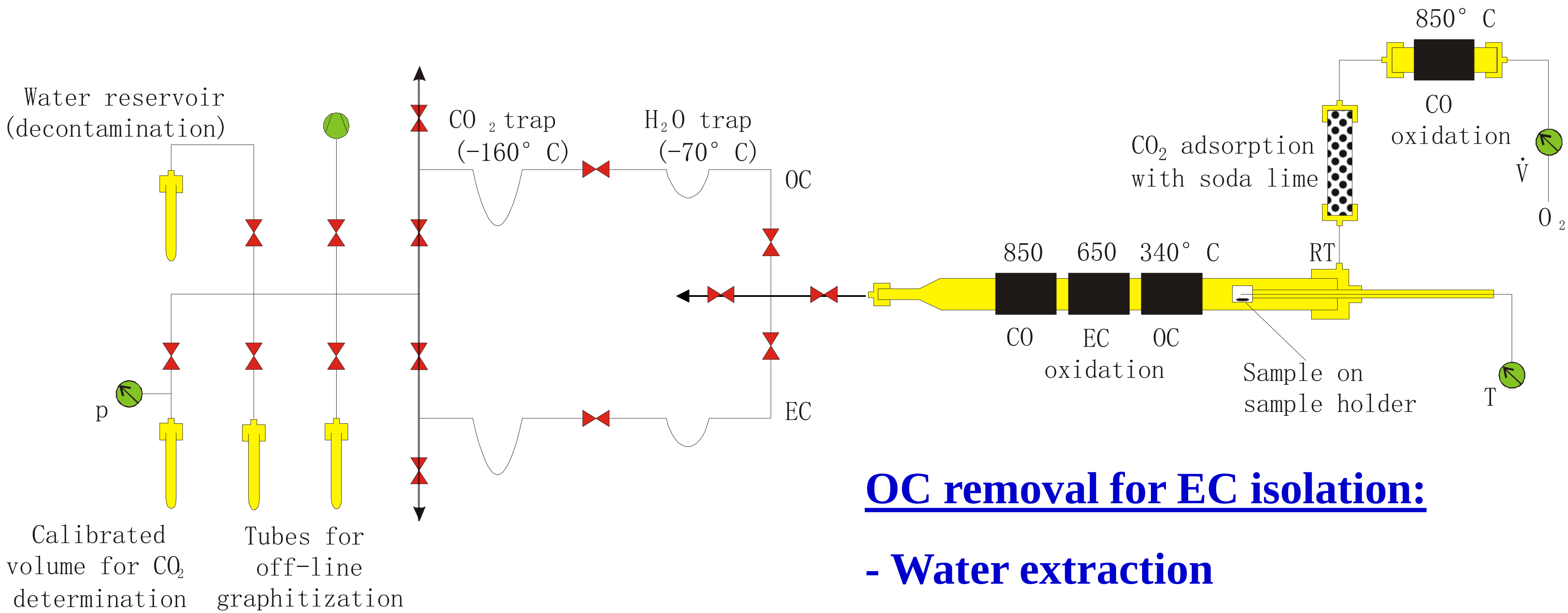
6 **^{14}C measurement**

Graphitization is not needed!

5-7 µgC sample is possible!

Previous method

Two-step combustion method

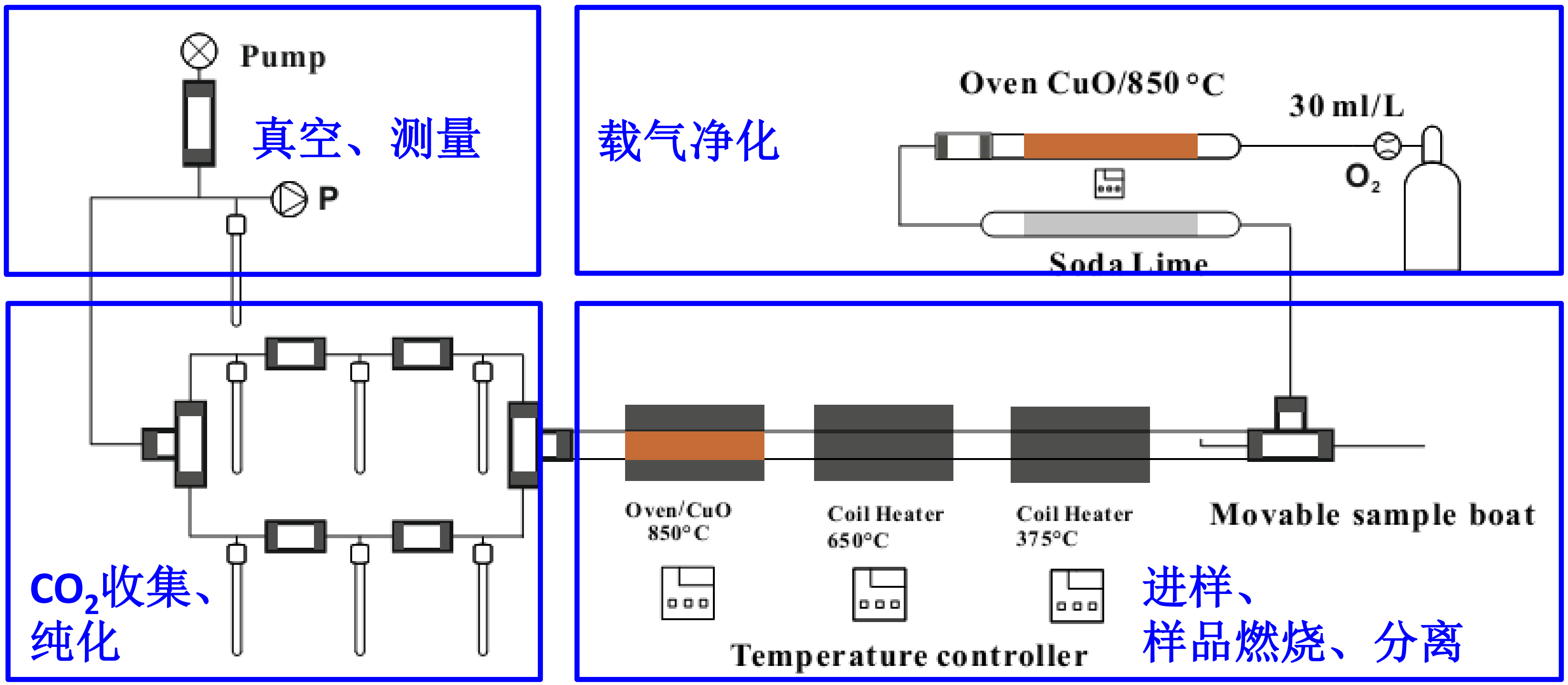


OC removal for EC isolation:

- **Water extraction**
- **4h @ 375-390°C**

Szidat et al., 2004

Layout of experimental line



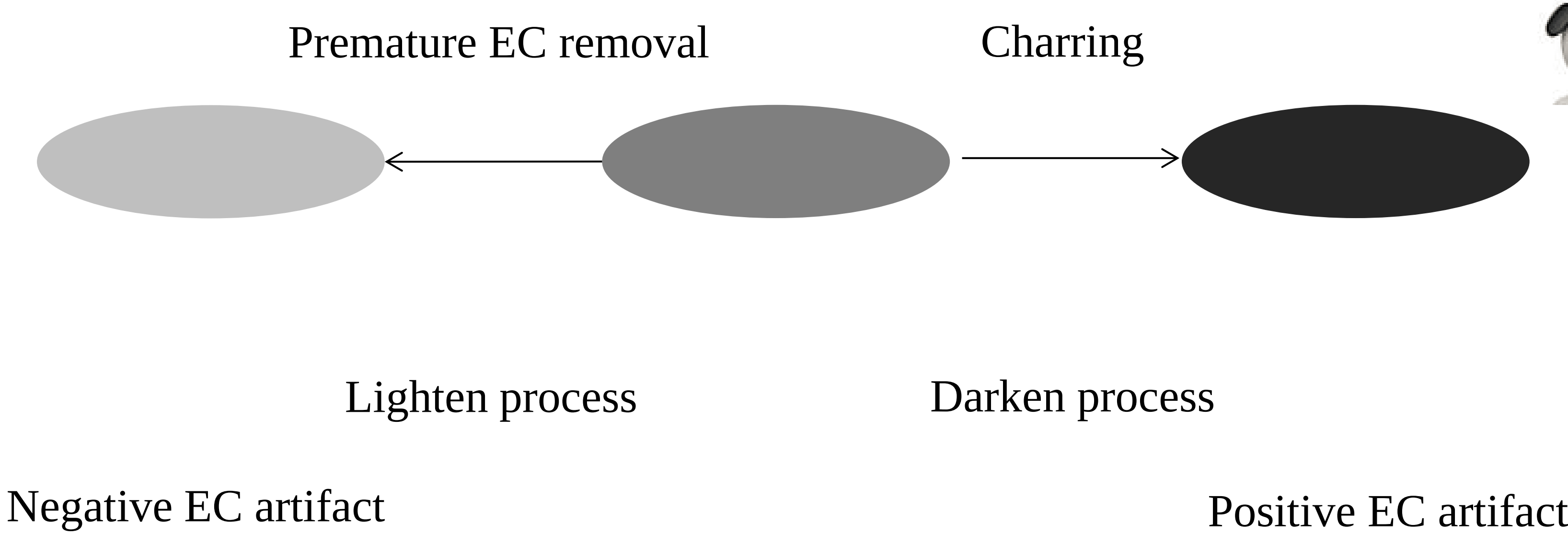
Zhang et al., 2010

Aerosol-^{14}C lab in GIG, CAS

⇧

气溶胶的^{14}C分析在线制样系统实物装置图 ⇨

The limitations of the current of method



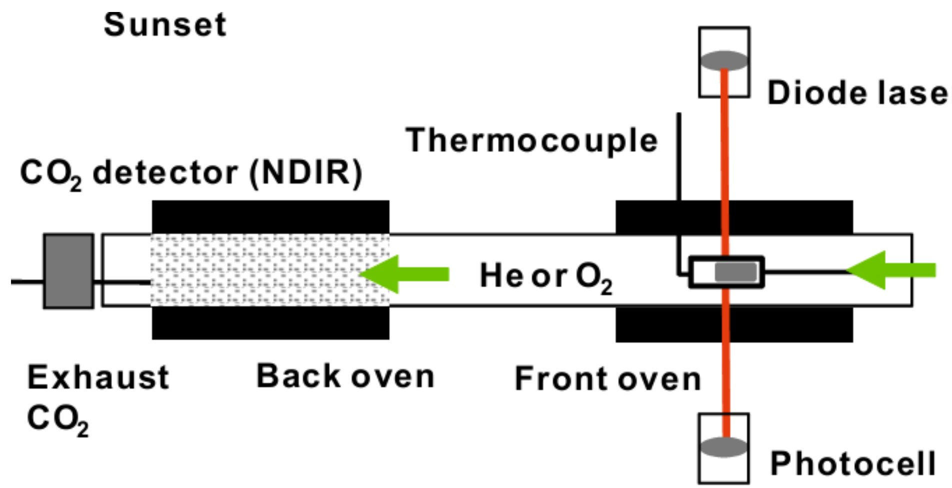
- ✓ The method is blind to OC charring and early removal of EC.
- ✓ It biases ^{14}C analysis of EC, because OC and EC have very different ^{14}C content.

Can we do better?

1. Minimizing of charring
2. Complete OC removal
3. As much EC as possible

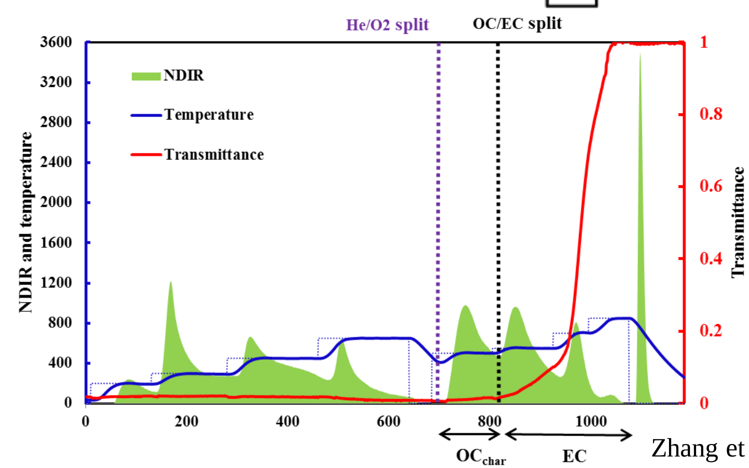
OC/EC analyzer: Sunset

Thermo-optical control of charring, OC removal and EC losses



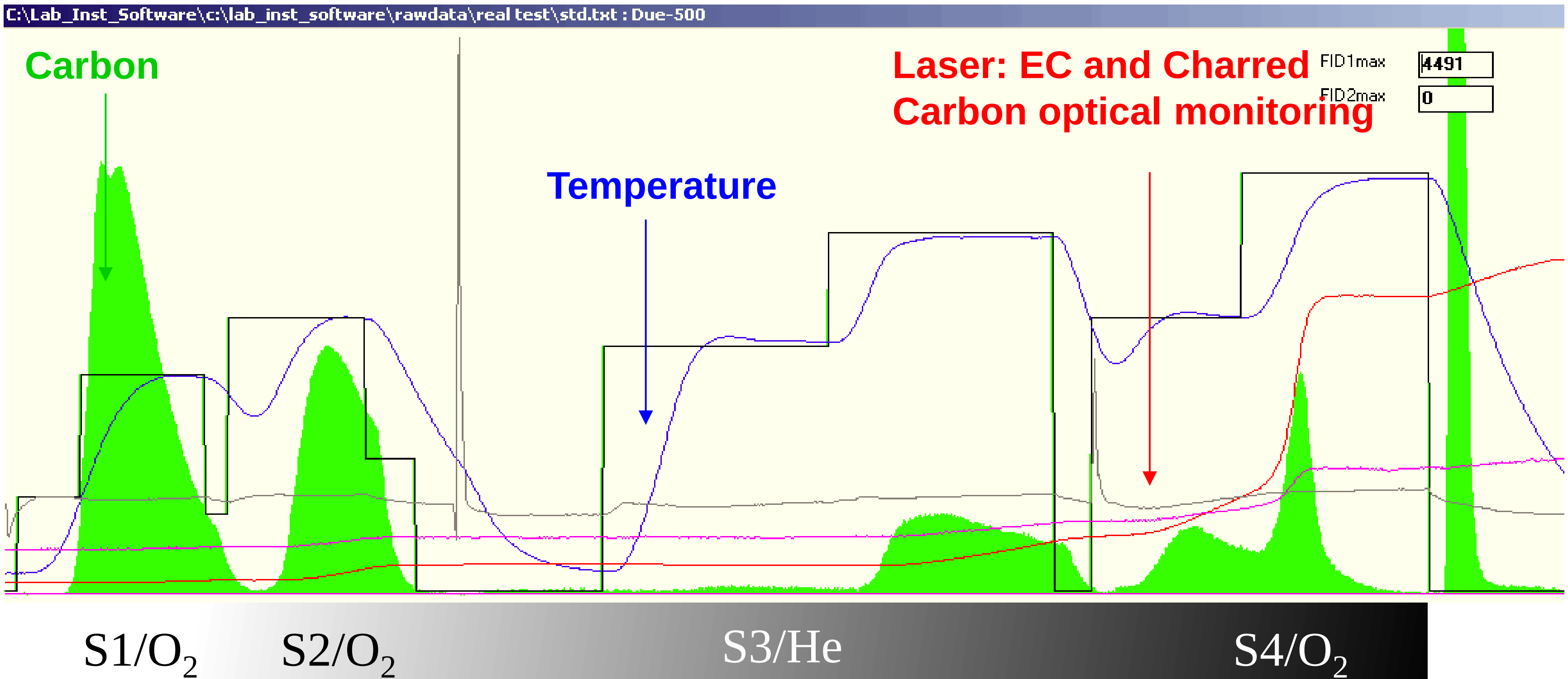
Specified Sunset instrument:

- ✓ NDIR detector to detect CO_2
- ✓ Multiple gas supply He, He/O_2, O_2
- ✓ Programmed oven temperature
- ✓ Continuous optical monitoring



Zhang et al., 2012

Swiss_4S protocol



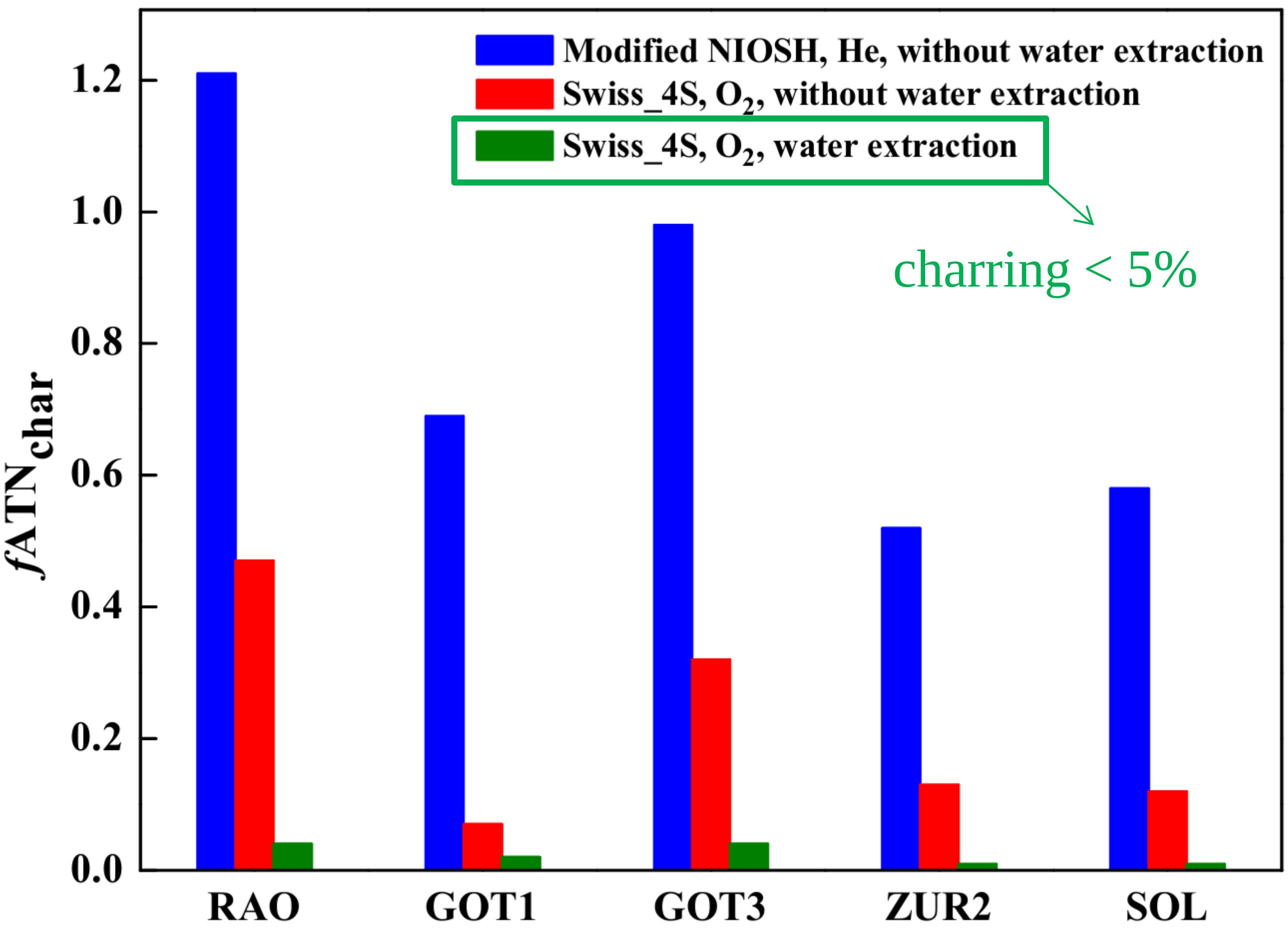
Pure OC	Mixture of OC and EC	Pure EC

Aims at recovering OC for ^{14}C measurement

Aims at recovering OC for ^{14}C measurement

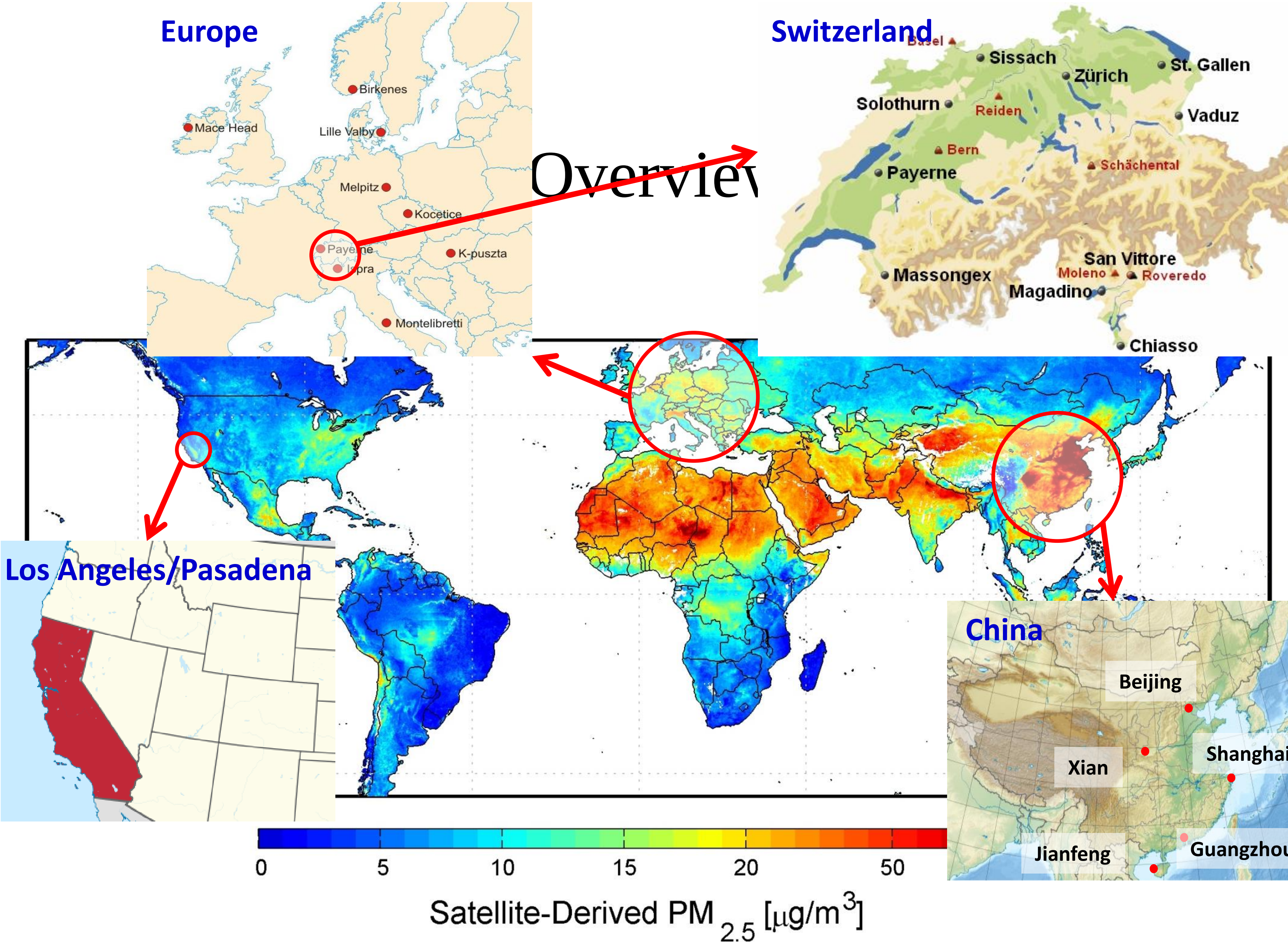
Zhang et al., 2012

Minimization of charring



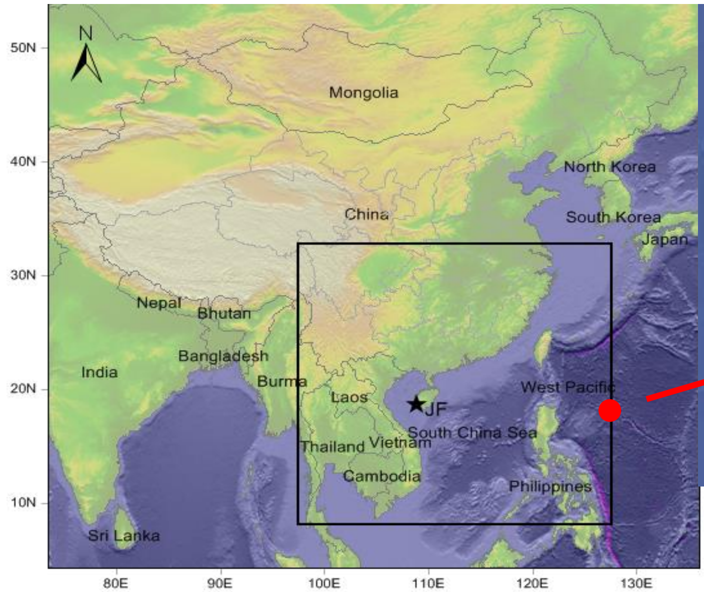
Zhang et al., 2012

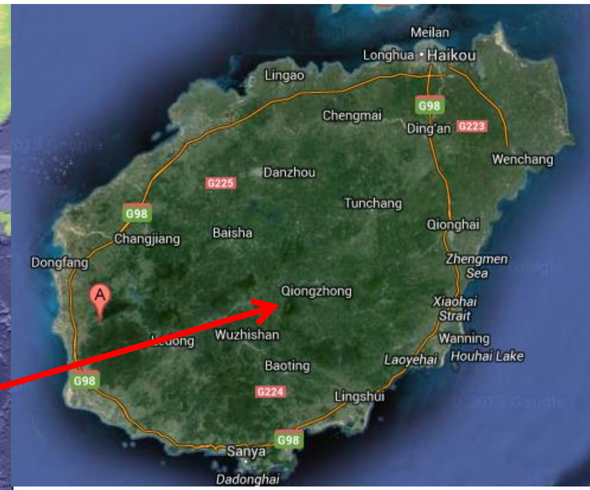
Overview



Europe
Birkenes
Mace Head
Lille Valby
Melpitz
Kocetice
Payerne
Ispra
K-puszta
Montelibretti
Switzerland
Basel
Sissach
Zürich
St. Gallen
Solothurn
Reiden
Vaduz
Bern
Schächental
Payerne
Massongex
San Vittore
Moleno
Roveredo
Magadino
Chiasso
Los Angeles/Pasadena
China
Beijing
Xian
Shanghai
Jianfeng
Guangzhou
0
5
10
15
20
50
Satellite-Derived PM 2.5 [μg/m3]

Jianfeng (JF), Hainan island

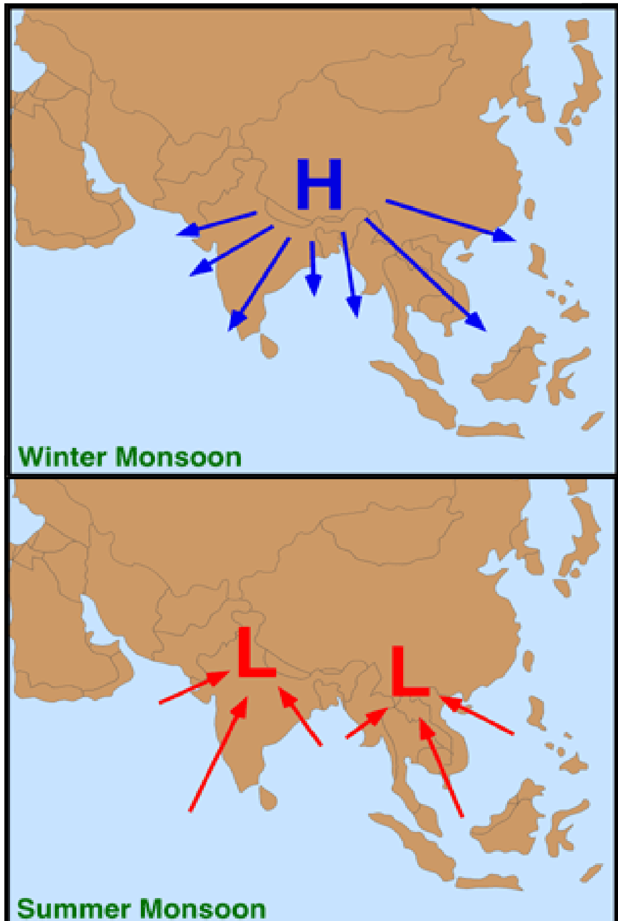


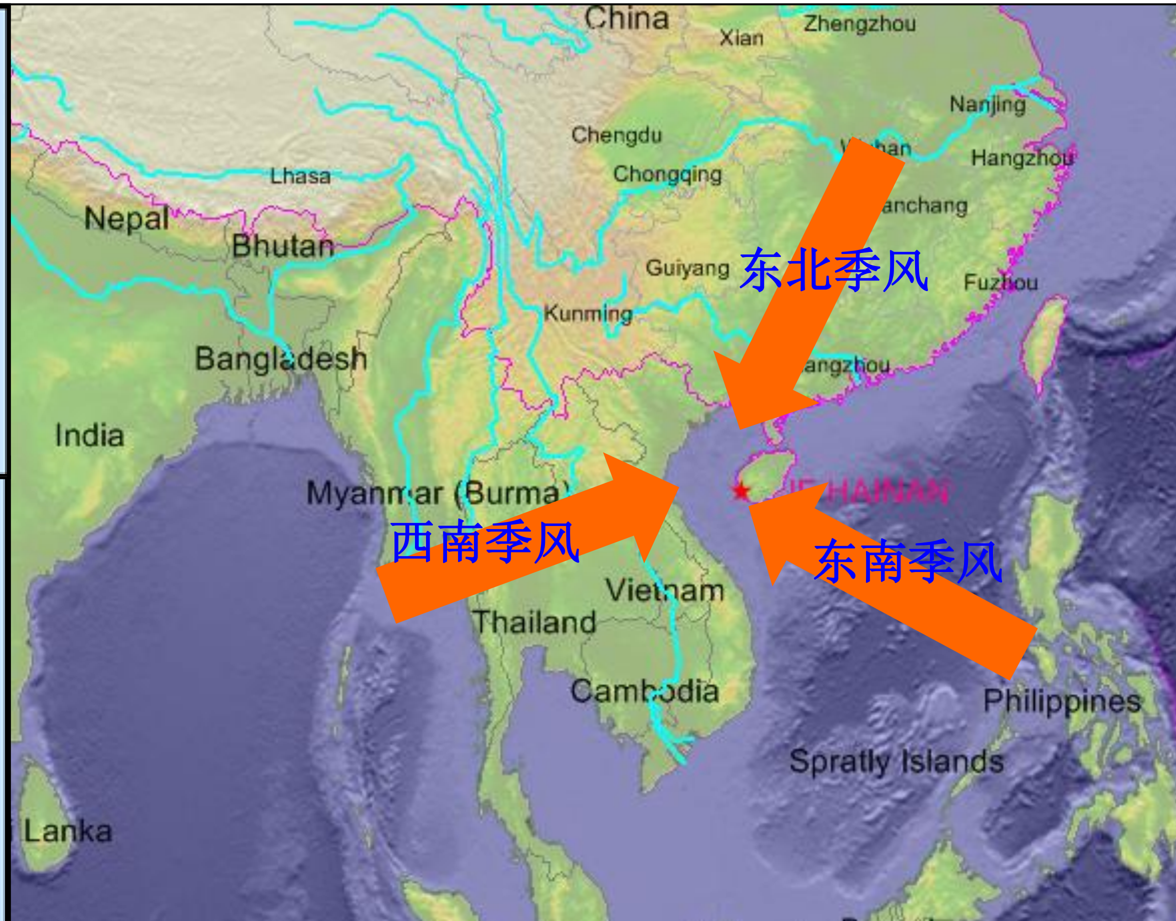


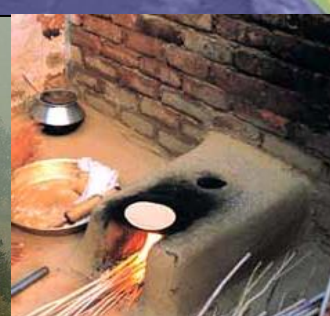
- 18° 40’ N, 108° 49‘ E
- Tropical rain forest
- National Reserve Park
- 115 km from city of Sanya
- Altitude: 820m asl.

PM2.5 24-h samples
May 2005 – Aug 2006

Asia monsoon system and potential aerosol sources

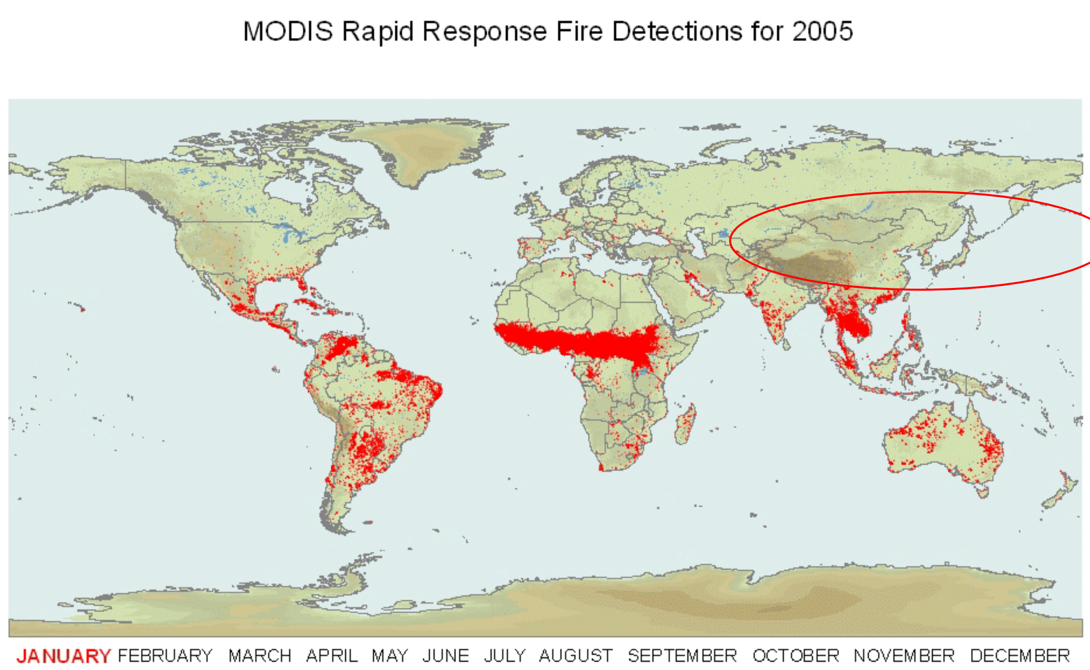






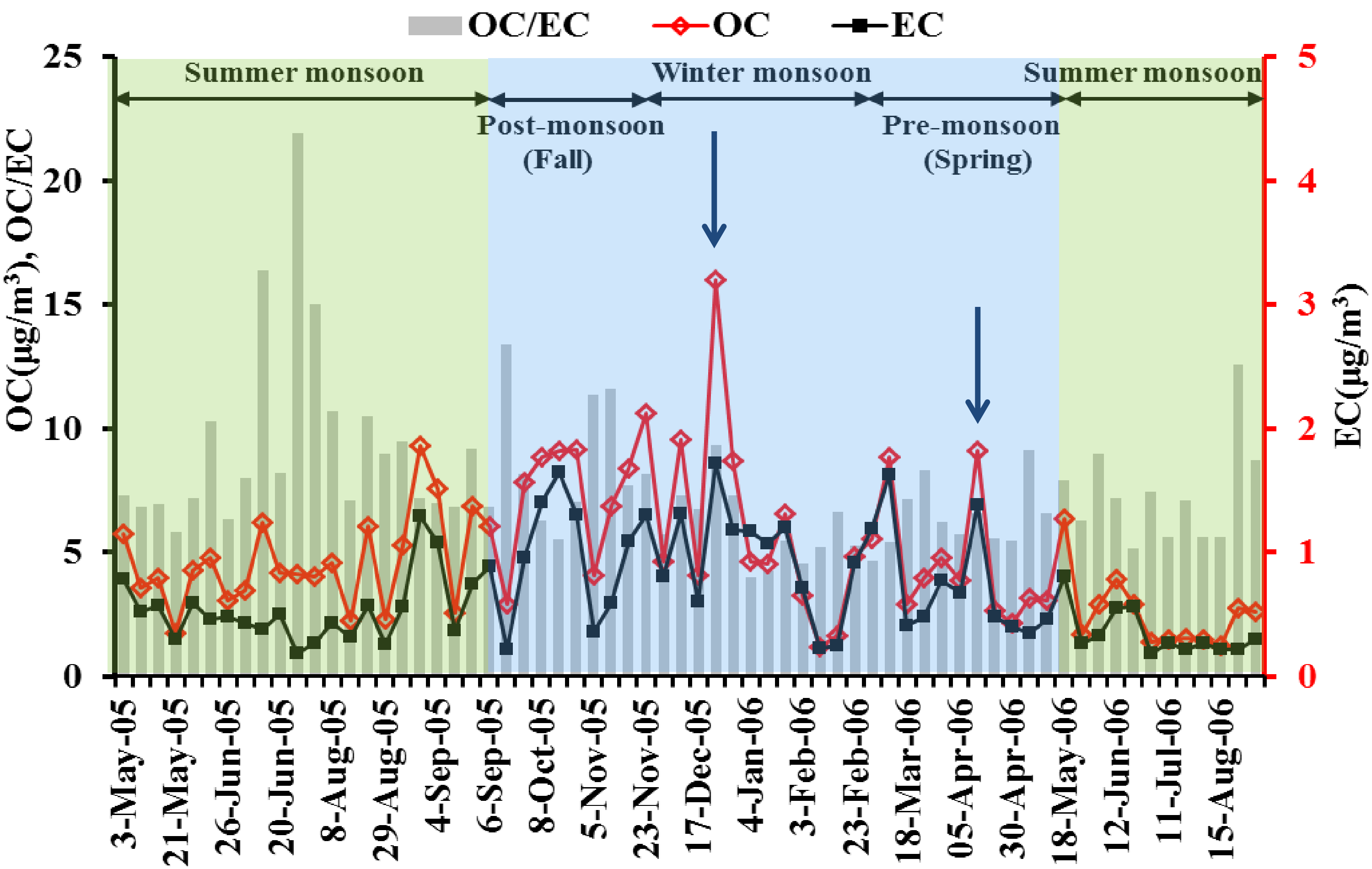
An important contributor: open biomass burning

Biomass (including residential biofuels) burning is an important contributor to air pollution in Asia (Streets et al., 2003).



Can open biomass-burning activities can influence on the air quality of this background site?

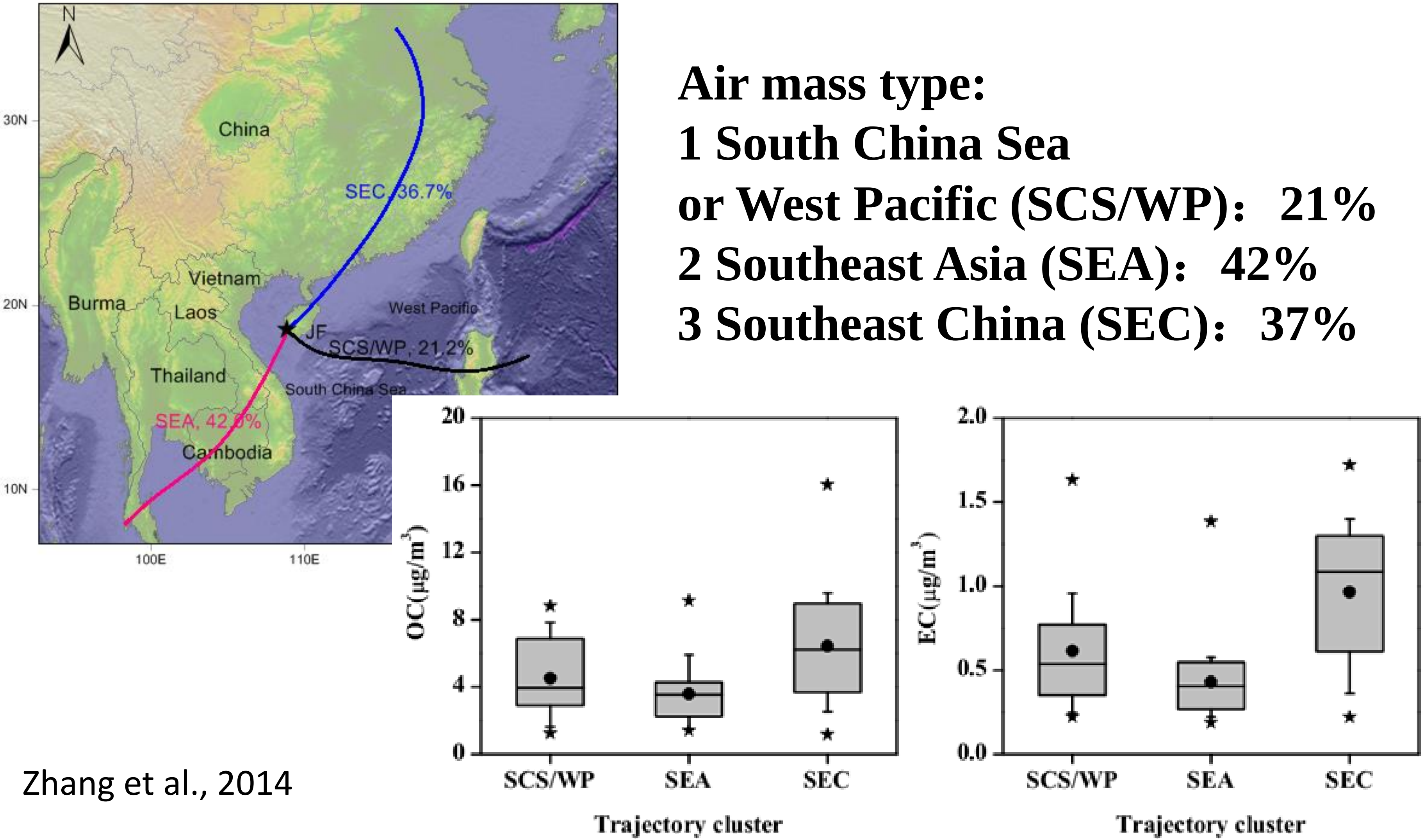
Temporal variations



Zhang et al., 2014

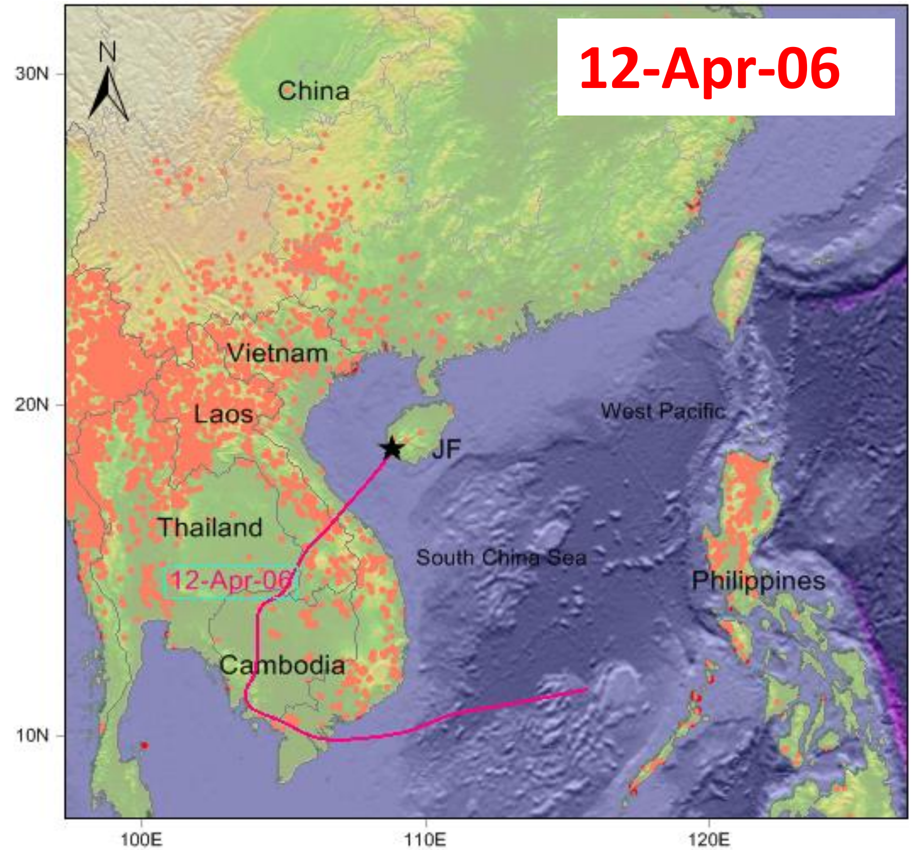
Backward trajectory analysis

Air mass type:
1 South China Sea or West Pacific (SCS/WP)：21%
2 Southeast Asia (SEA)：42%
3 Southeast China (SEC)：37%



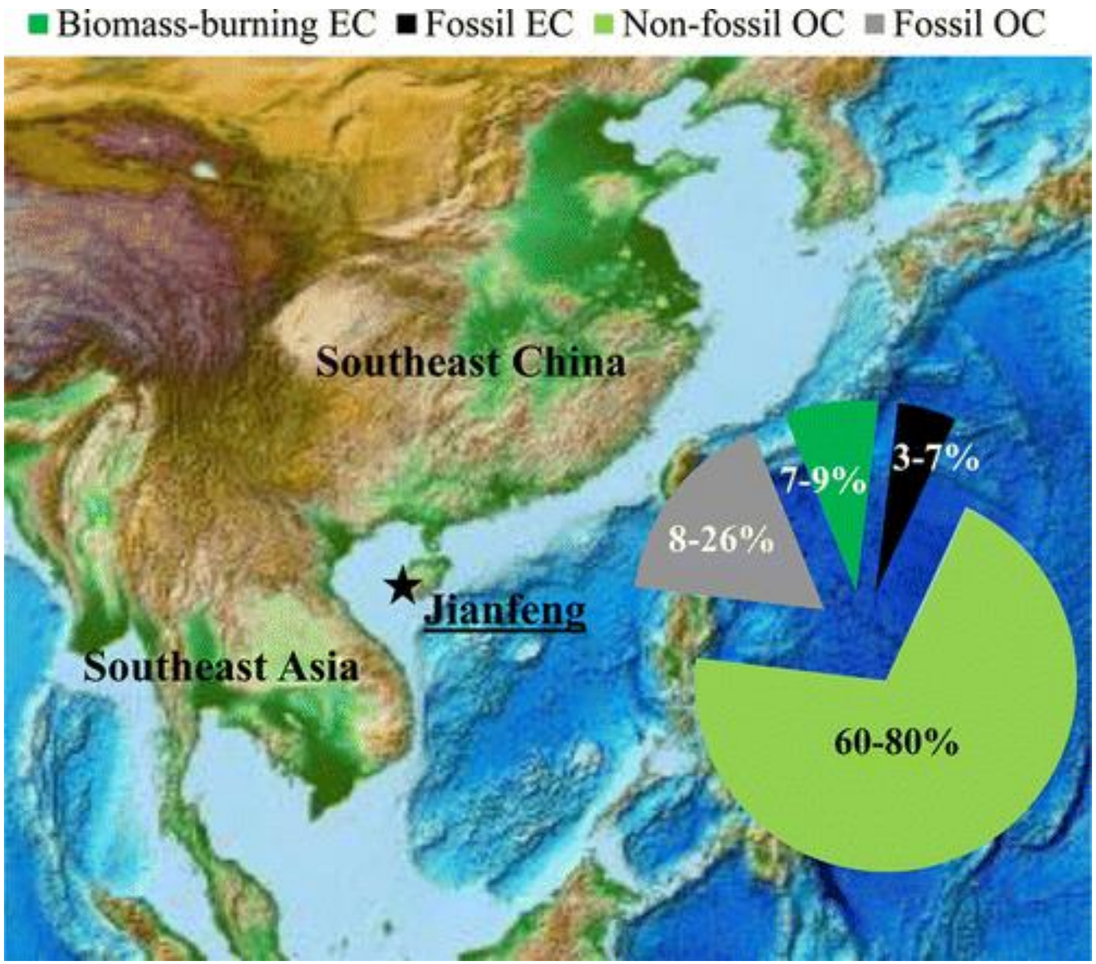
Zhang et al., 2014

Backward trajectory and fire counts map



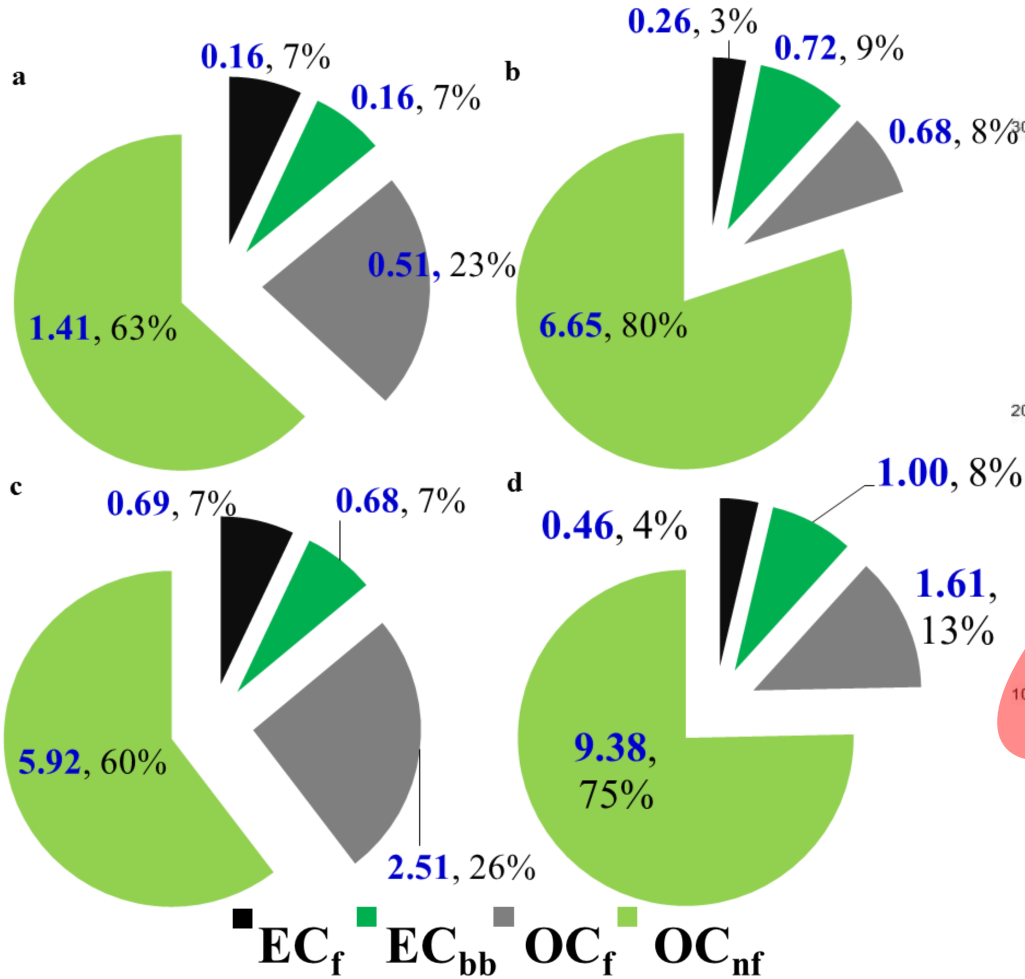
- **High concentrations were associated to intensive open biomass-burning activities.**

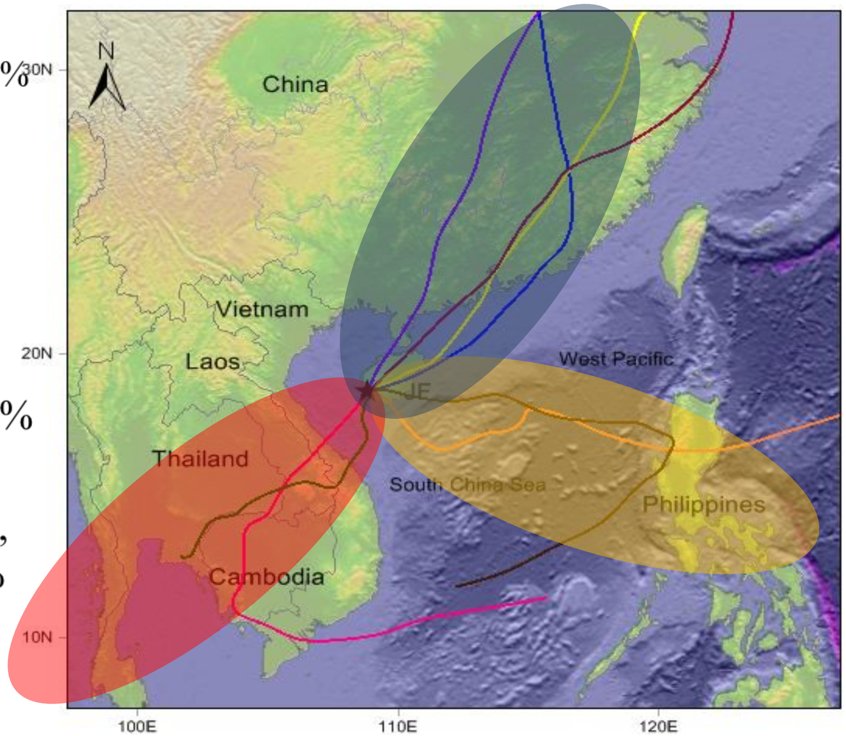
^{14}C-based source apportionment



Zhang et al., 2014

^{14}C Source apportionment for different source regions





Numbers: mass concentration (μg/m^3)

Percentages: relative contribution to TC

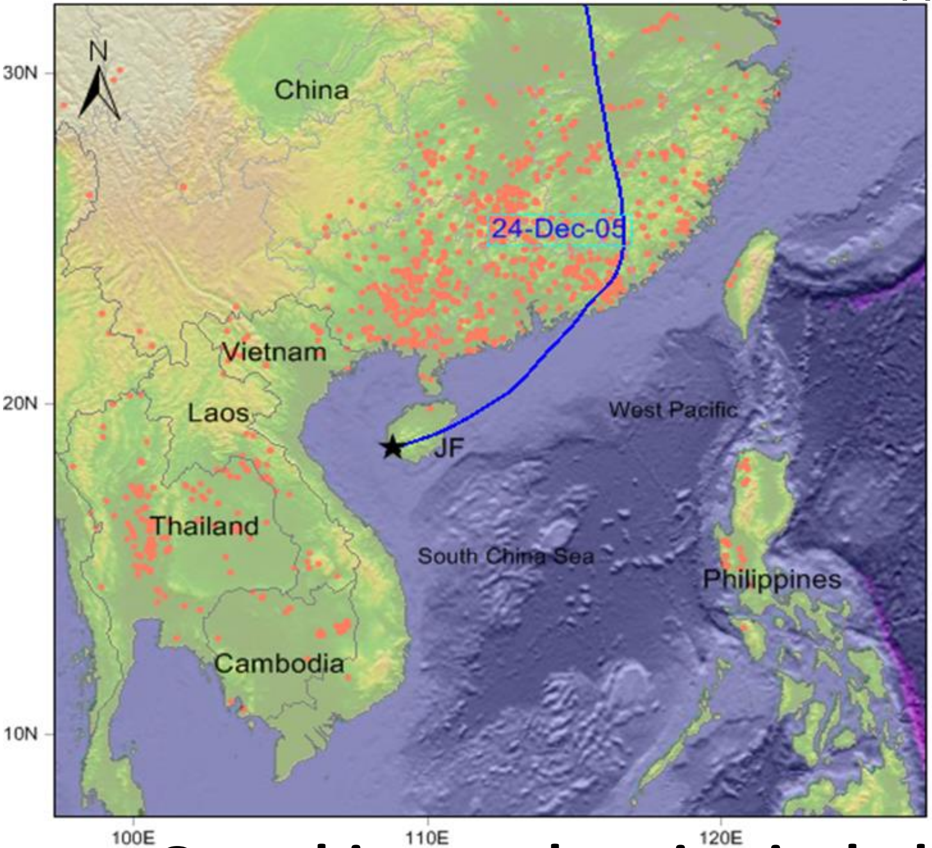
(a) South China Sea (SCS)

(b) Southeast Asia (SEA)

(c) Southeast China (SEC) low fire

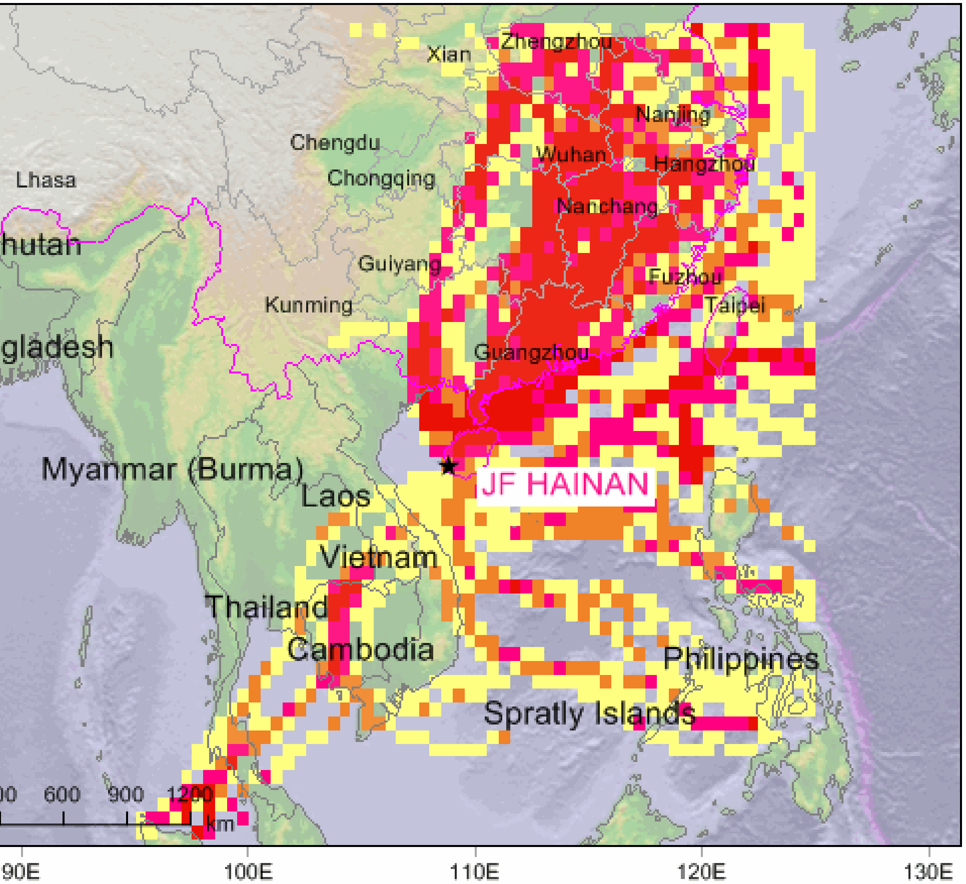
(d) Southeast China (SEC), high fire

Why non-fossil contributions are high in winter when air mass travelling from SEC?

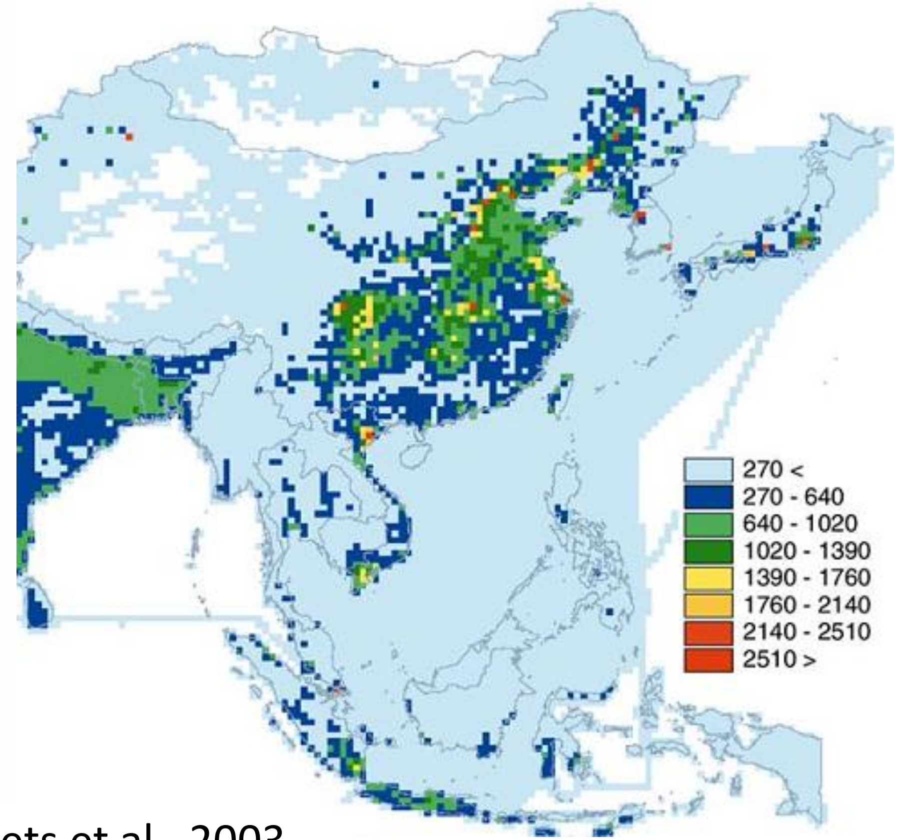


- **Open biomass burning including agricultural residue combustion and forest fires**
- **Biofuel usage for heating in winter as well as cooking**

Potential source contribution function (PSCF)



2000年排放清单



Streets et al., 2003

Overview of ^{14}C studies in Asia

Percentages of non-fossil

Location	TC	EC	OC
Rural or remote site			
Maldives	~68	~70	
Sinhagad, India	~65	~62	
Cape Hedo, Japan		43-62	60-97
Yufa, China	~37	~26	
Jiuxianshan, China	55-87		
Jianfeng, China	~77	~62	~81
Sub-urban or urban site			
Maebashi, Japan	37±15		
Kisai, Japan	37±12		
Shanghai		~46	~63
Tokyo	33-45		
Beijing	32-50		
Lhasa, China	~49		
Xiamen, China	27-37		

TC: >60% (rural)
>30% (urban)
OC: >60%
EC: >40% (rural)
>20% (urban)

Urban modern carbon:

- Biogenic POA and SOA
- BB from cooking and heating
- Mixed with non-fossil aerosols from neighboring rural regions

Reference not shown here

Conclusions

1. ^{14}C analysis can provide an improved and unambiguous source apportionment of OC and EC.
2. This method has been successfully applied to different field studies.
3. Based on our studies, different PM control mitigation should be taken for different targeted regions.