Refined ^{14}C source apportionment of organic carbon

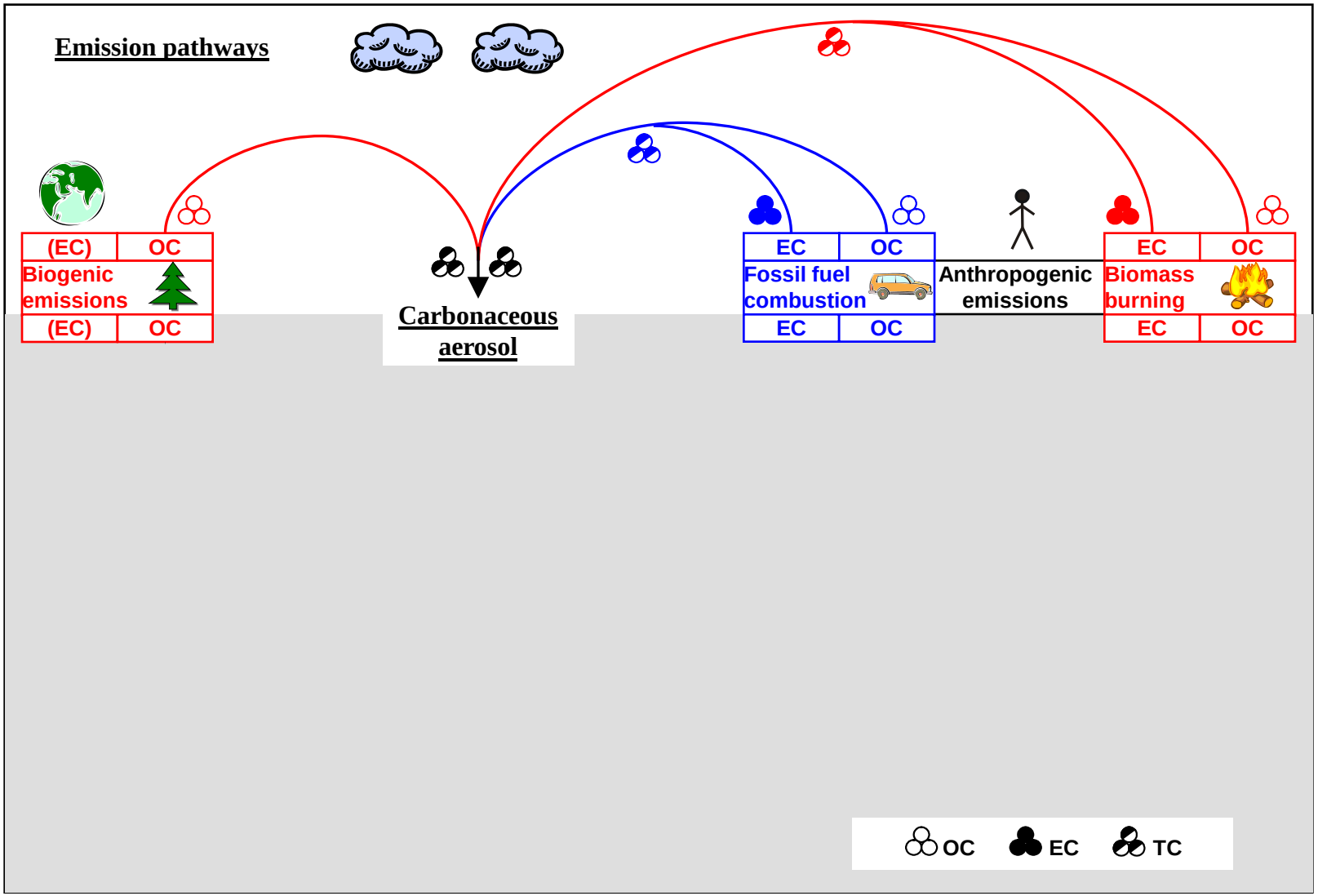
Sönke Szidat [1], Simon Fahrni [1], Urs Baltensperger [2],

Matthias Ruff [2], Lukas Wacker [3], Barbara Klatzer [4],

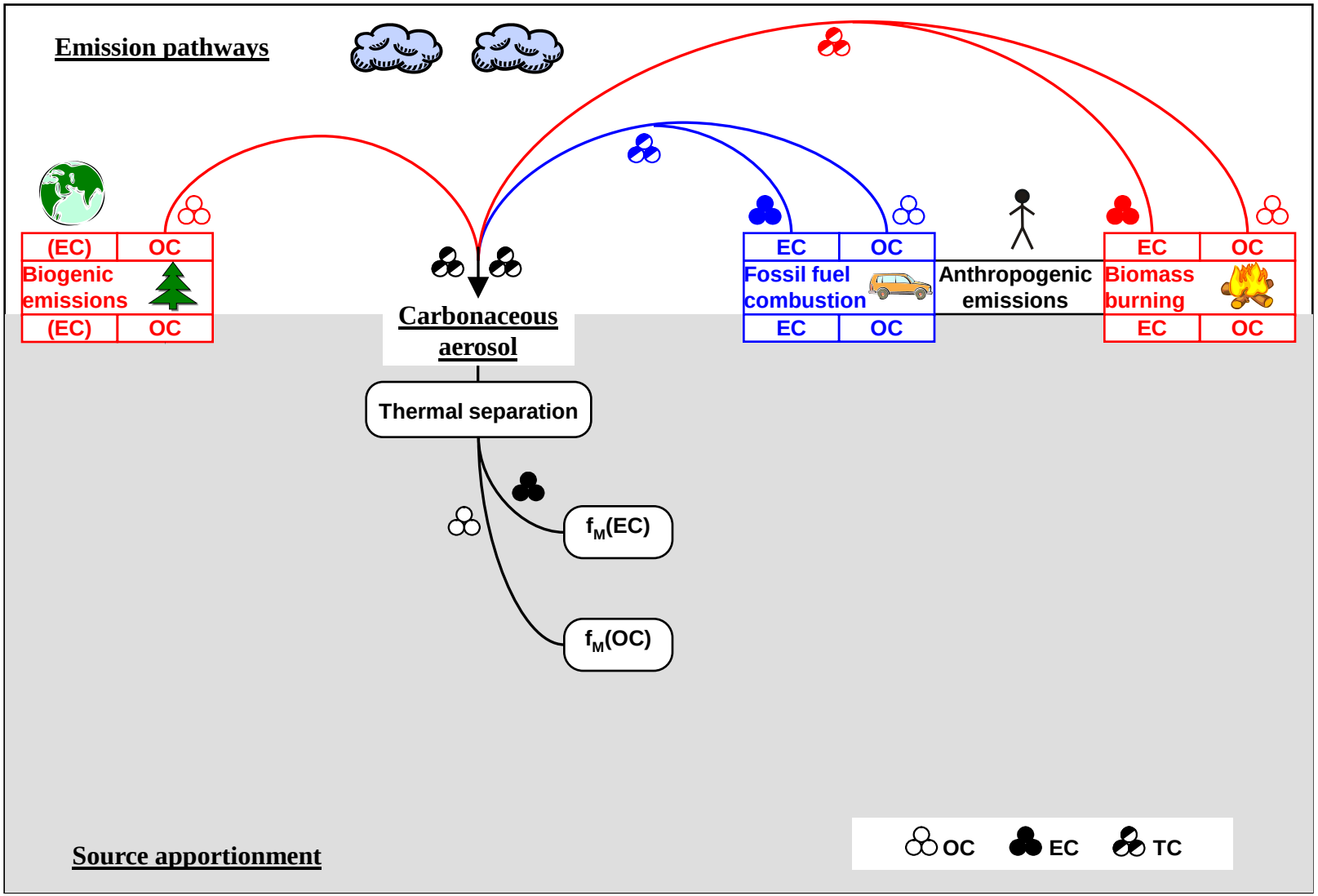
Hans Puxbaum [4], Emanuela Finessi [5], Stefano Decesari [5]

[1] Department of Chemistry and Biochemistry, University of Berne, Switzerland
[2] Paul Scherrer Institut (PSI), Villigen, Switzerland
[3] Institute for Particle Physics, ETH Hönggerberg, Zürich, Switzerland
[4] Technical University of Vienna, Austria
[5] Instituto ISAC – C.N.R., Bologna, Italy

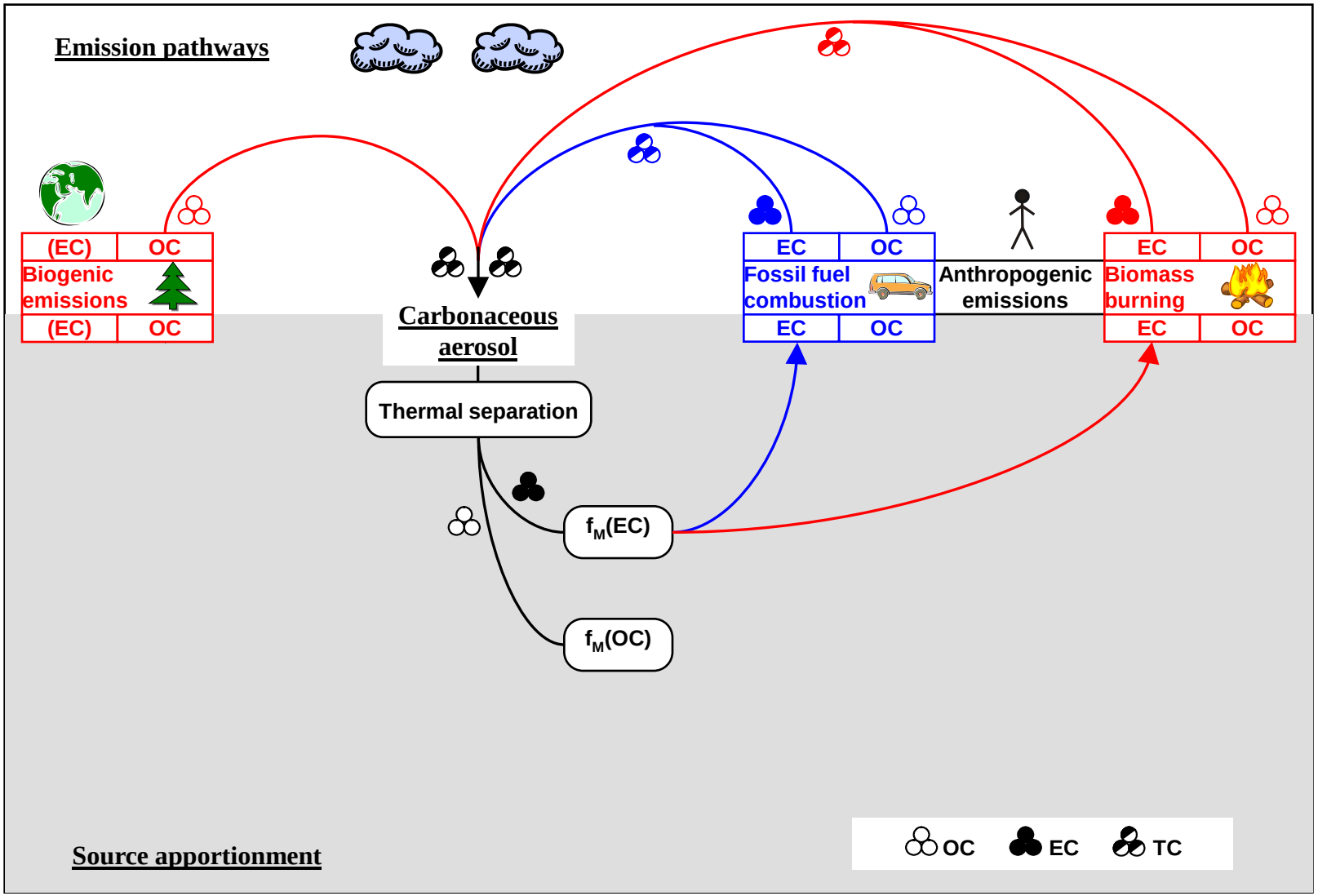
An advanced source apportionment model contemporary, fossil and mixed carbon



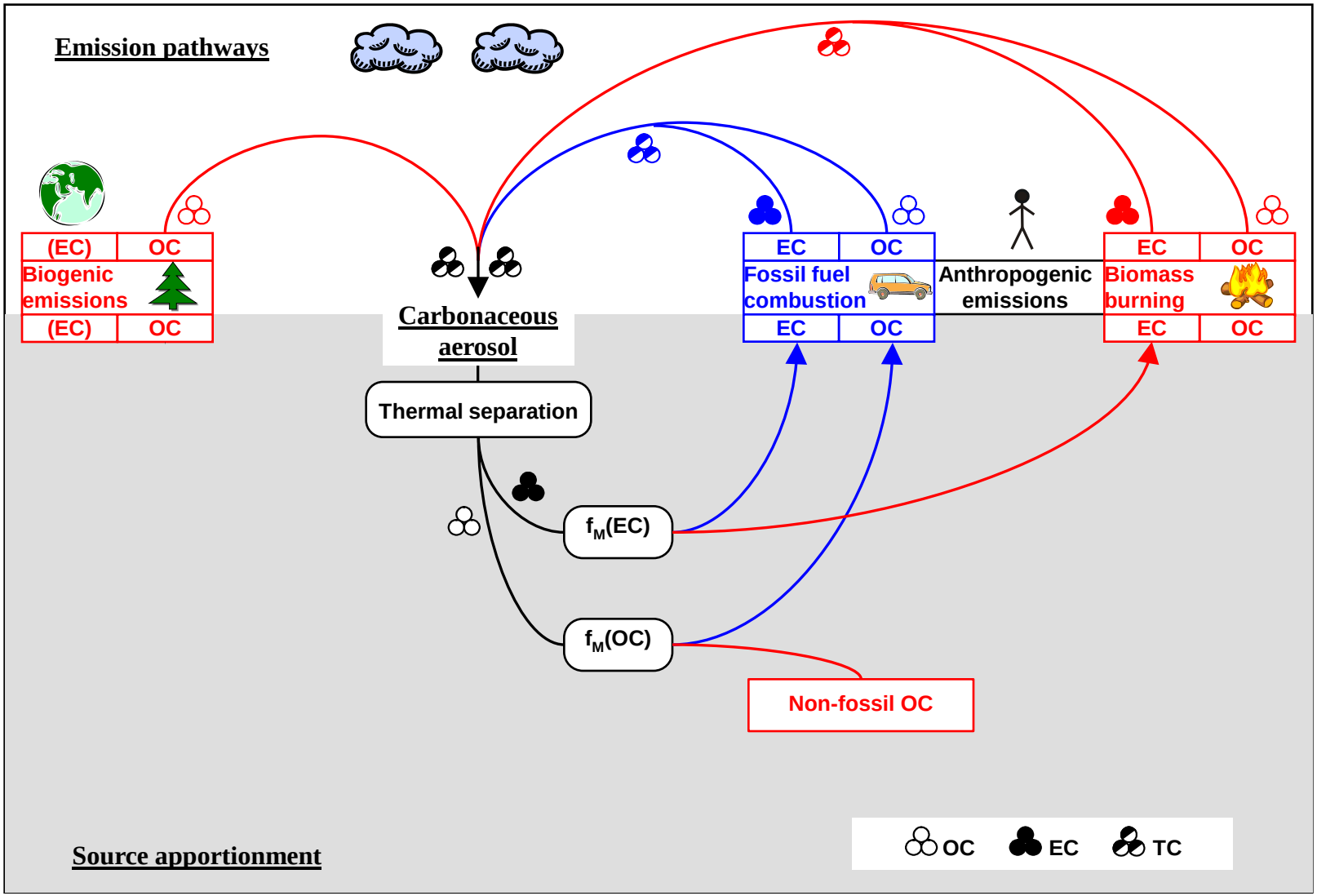
An advanced source apportionment model contemporary, fossil and mixed carbon



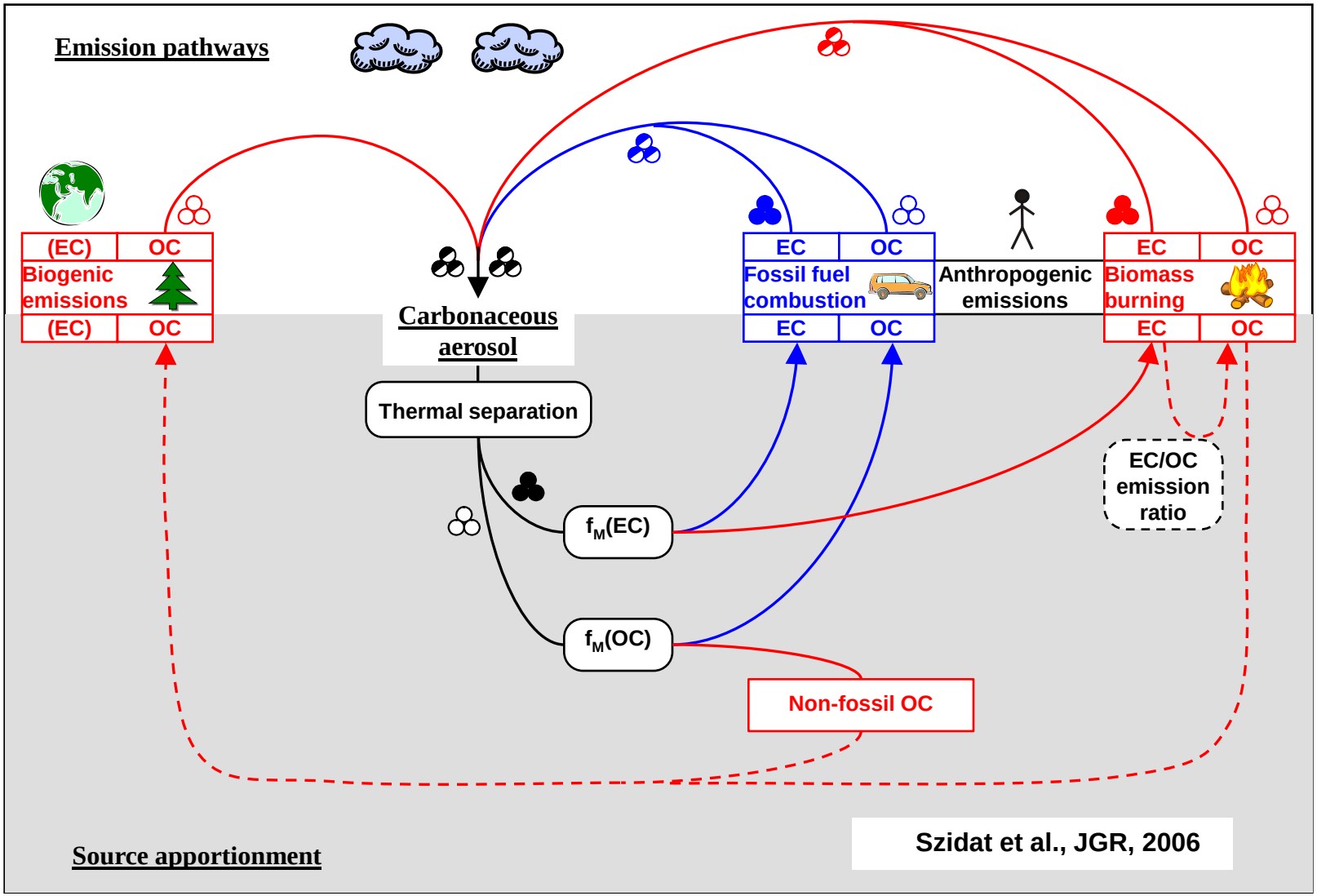
An advanced source apportionment model contemporary, fossil and mixed carbon



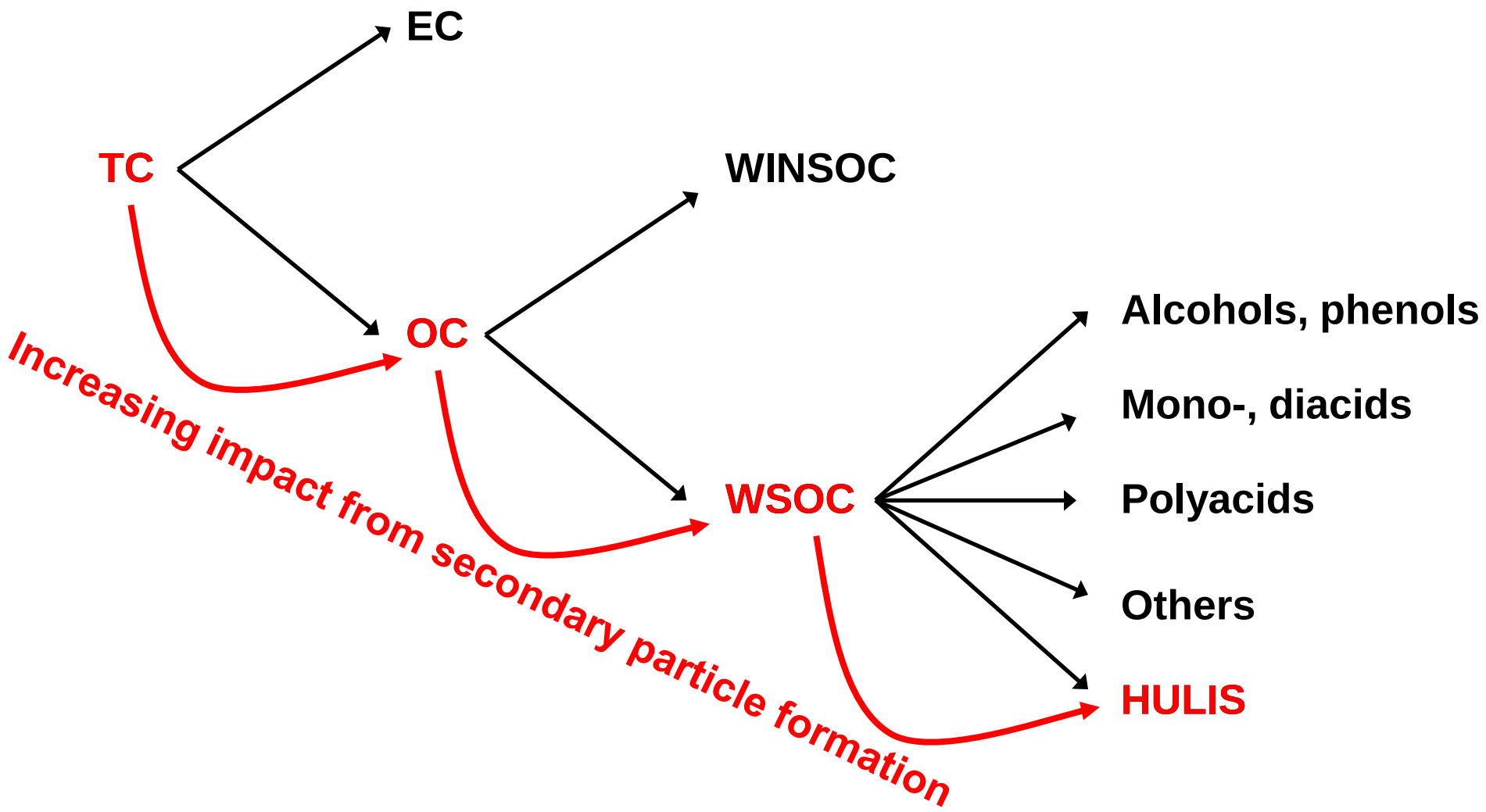
An advanced source apportionment model contemporary, fossil and mixed carbon



An advanced source apportionment model contemporary, fossil and mixed carbon



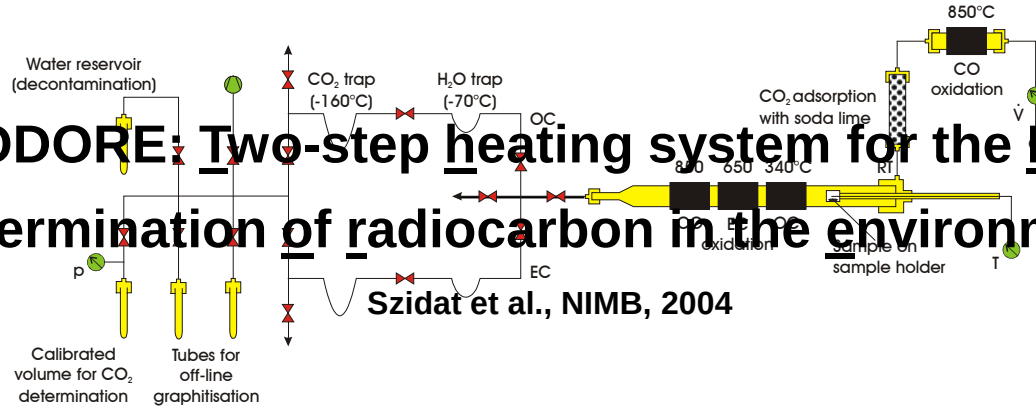
TC and its subfractions



Preparation of OC and EC for ^{14}C analysis

THEODORE: Two-step heating system for the EC/OC determination of radiocarbon in the environment

Szidat et al., NIMB, 2004

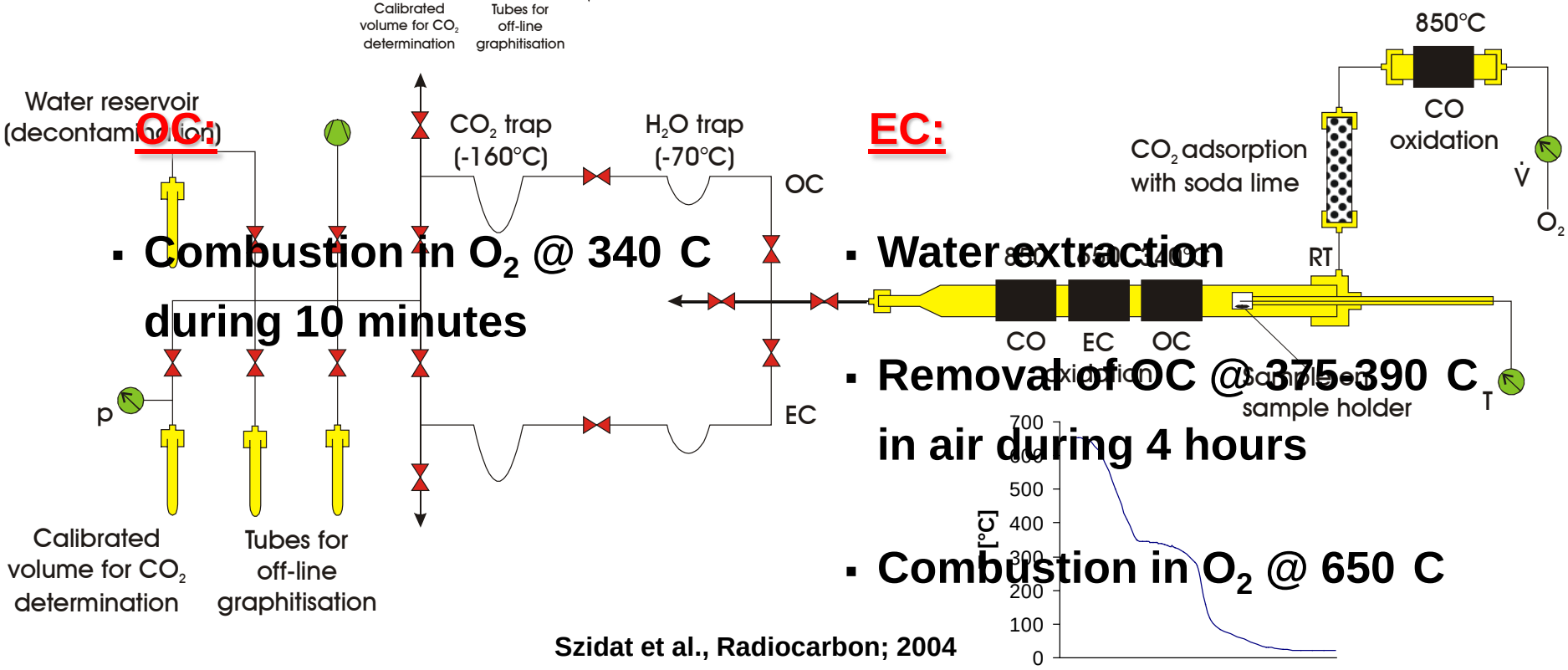


OC:

- Combustion in O_2 @ 340 C during 10 minutes

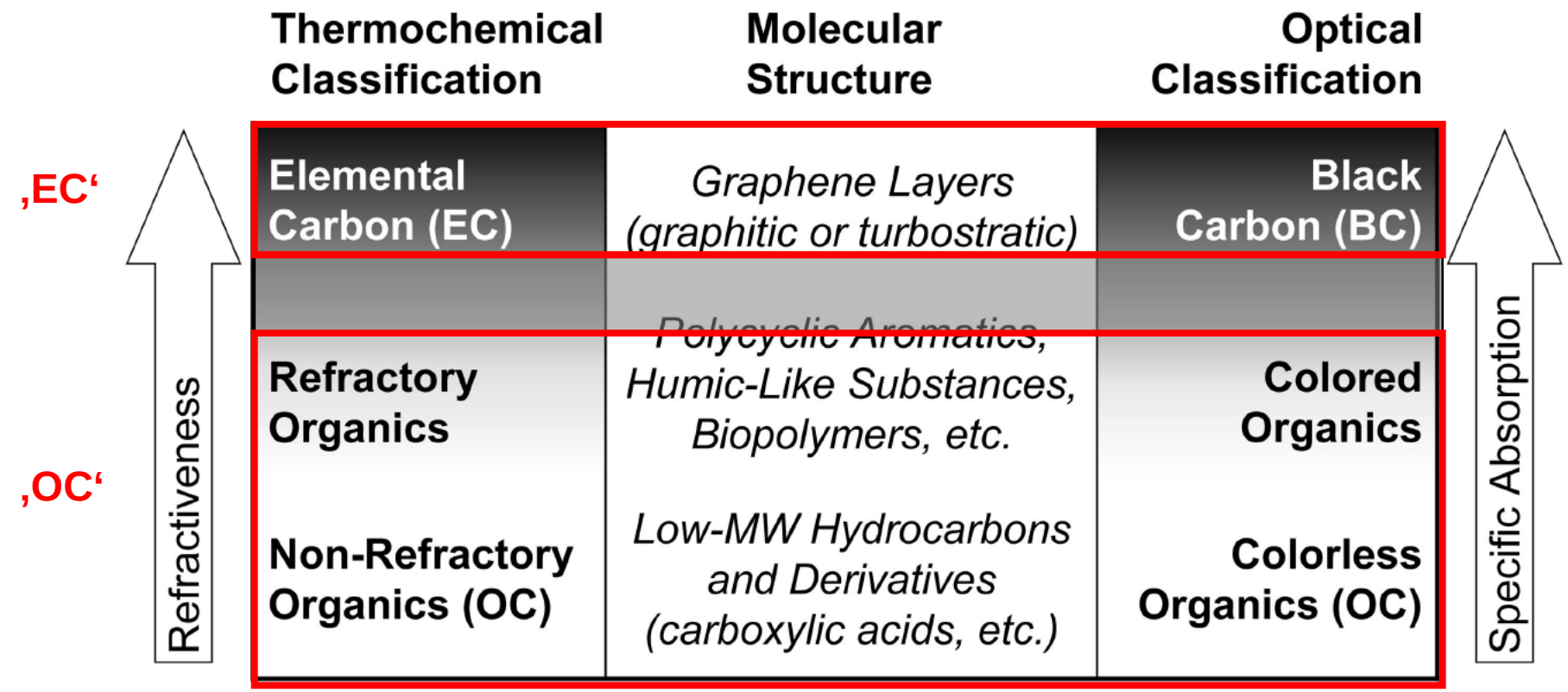
EC:

- Water extraction
- Removal of OC @ 375-390 C in air during 4 hours
- Combustion in O_2 @ 650 C



Szidat et al., Radiocarbon; 2004

Thus there is a ‚grey' zone between OC and EC in our C-14 measurements: overall yield for Σ(OC+EC) = 70-80%



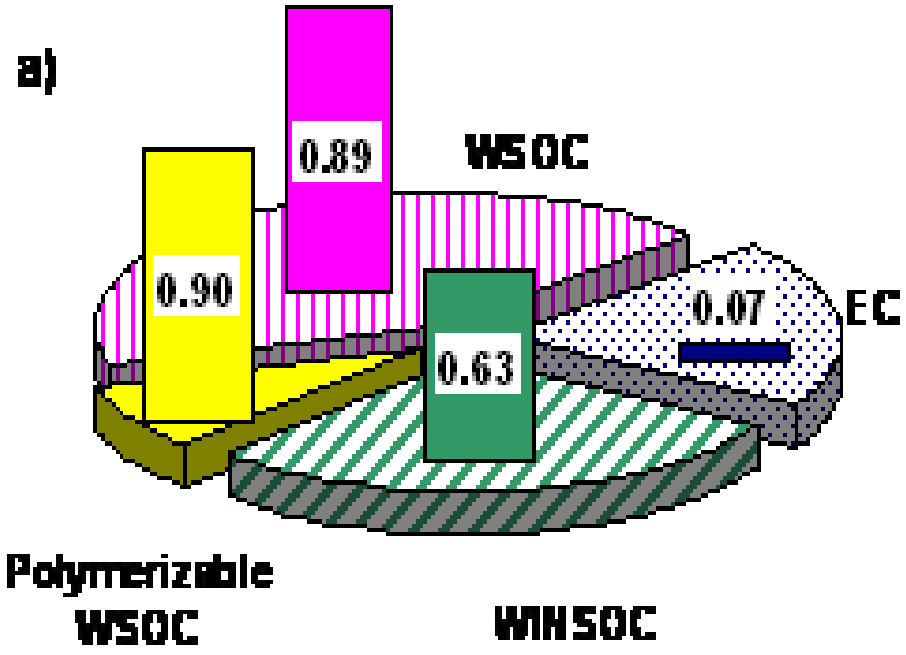
Andreae and Gelencser ACP 2006

^{14}C measurement @ ETH Zurich with Micadas, a small accelerator mass spectrometer (AMS)

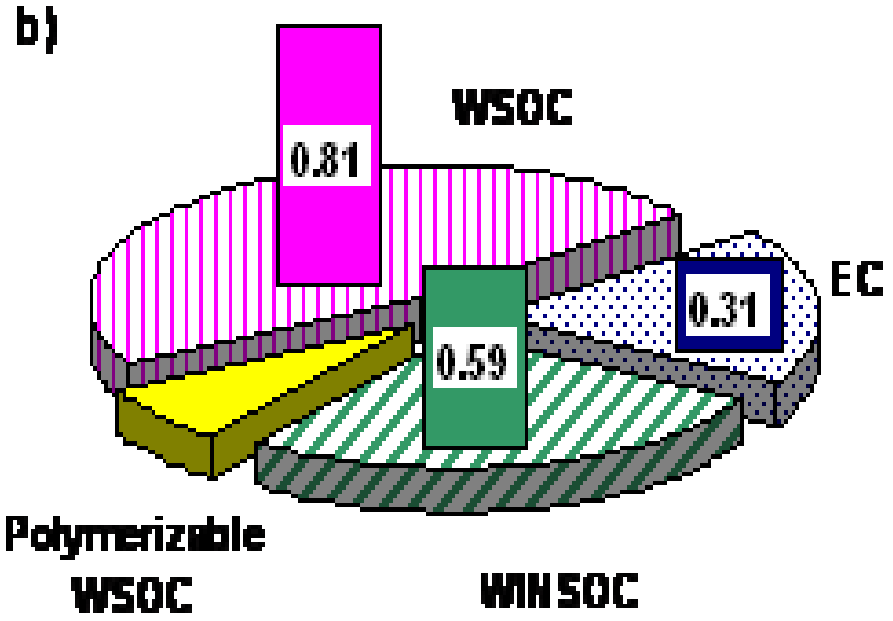
Synal et al., 2007

Fossil and non-fossil fractions of OC and EC for Zurich

Summer



Winter

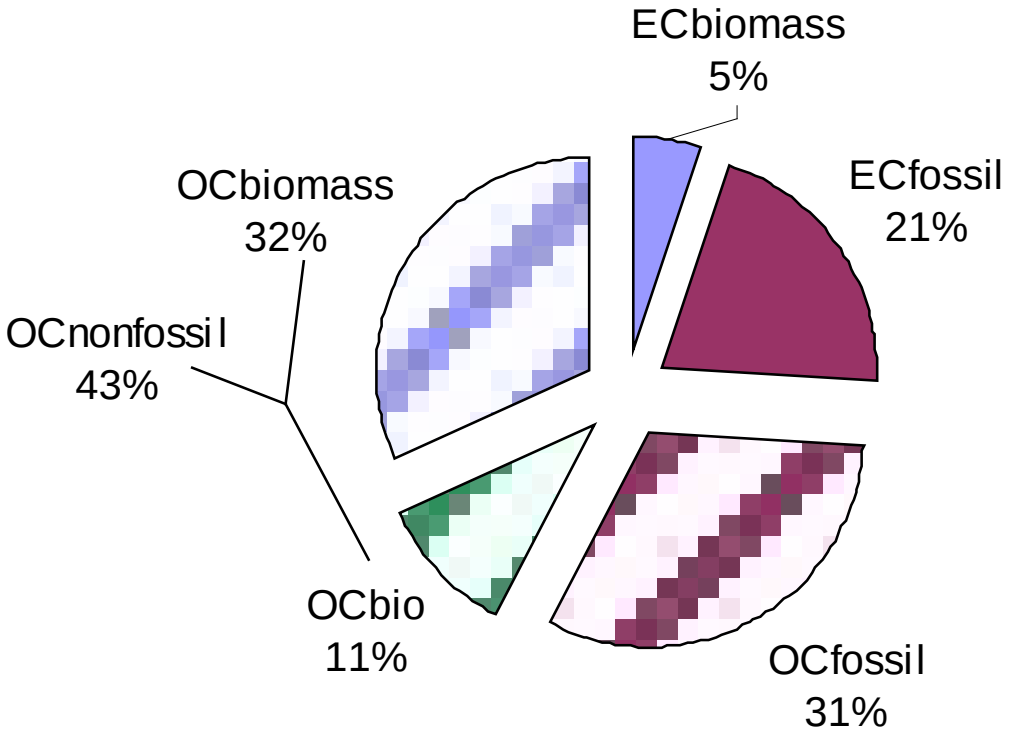


Szidat et al., JGR (2006)

PM10 sampling at San Pietro Capofiume, Italy

- **Rural station in an industrialized region**
- **3 daily HiVol samples were collected in July 2007**

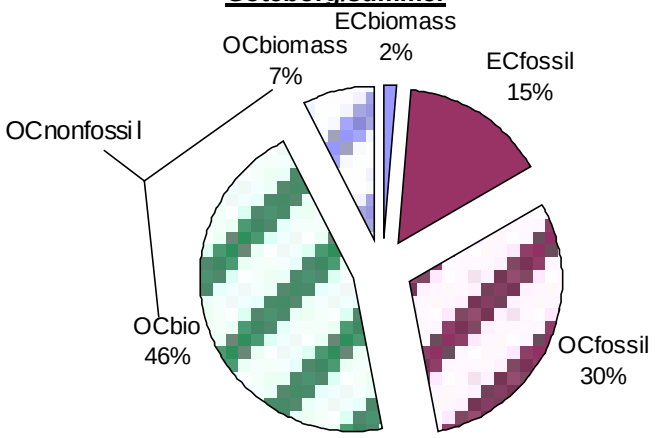
Fossil and non-fossil sources of OC and EC in San Pietro Capofiume



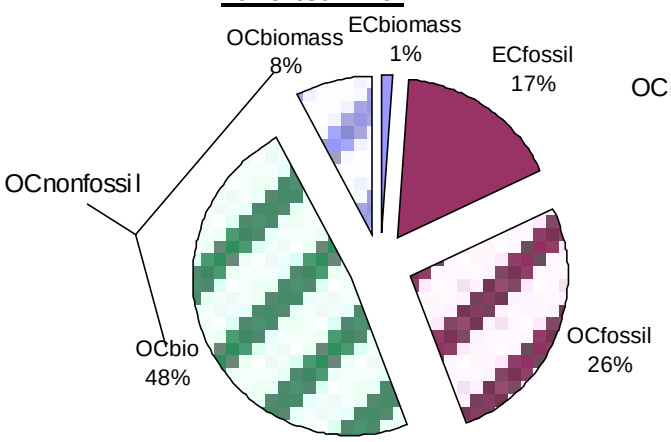
- **Average of the 3 daily HiVol samples**
- **TC: 4.0 µg/m^3**

^{14}C: Comparison of different sites

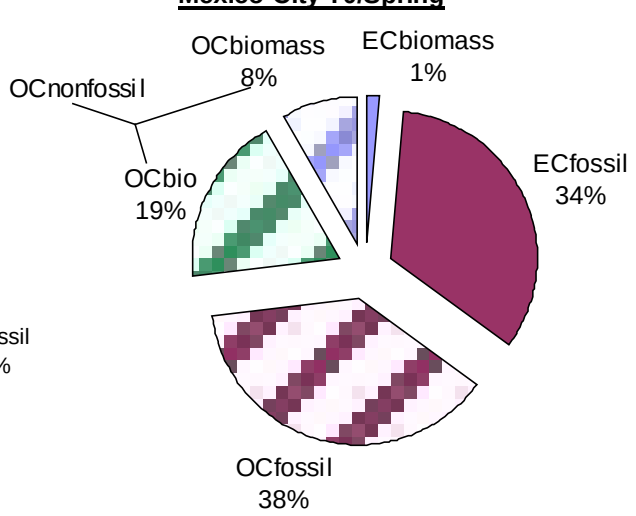
Göteborg/summer



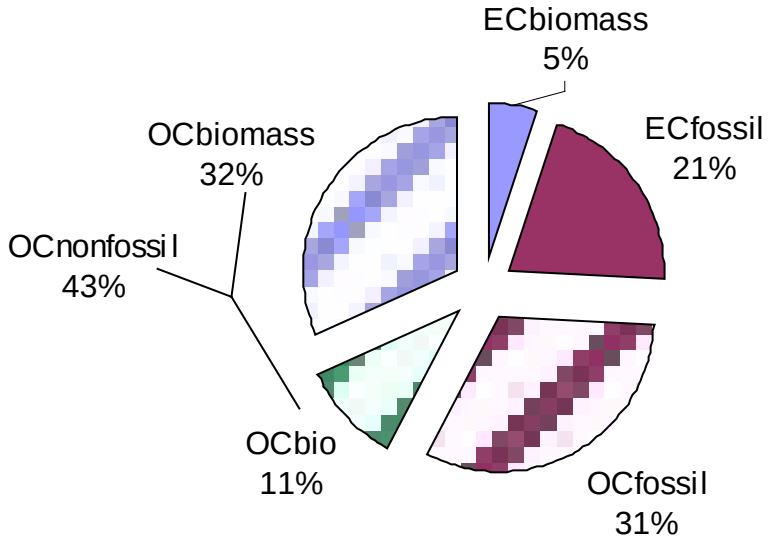
Zurich/summer



Mexico-City T0/Spring



San Pietro Capofiume/summer

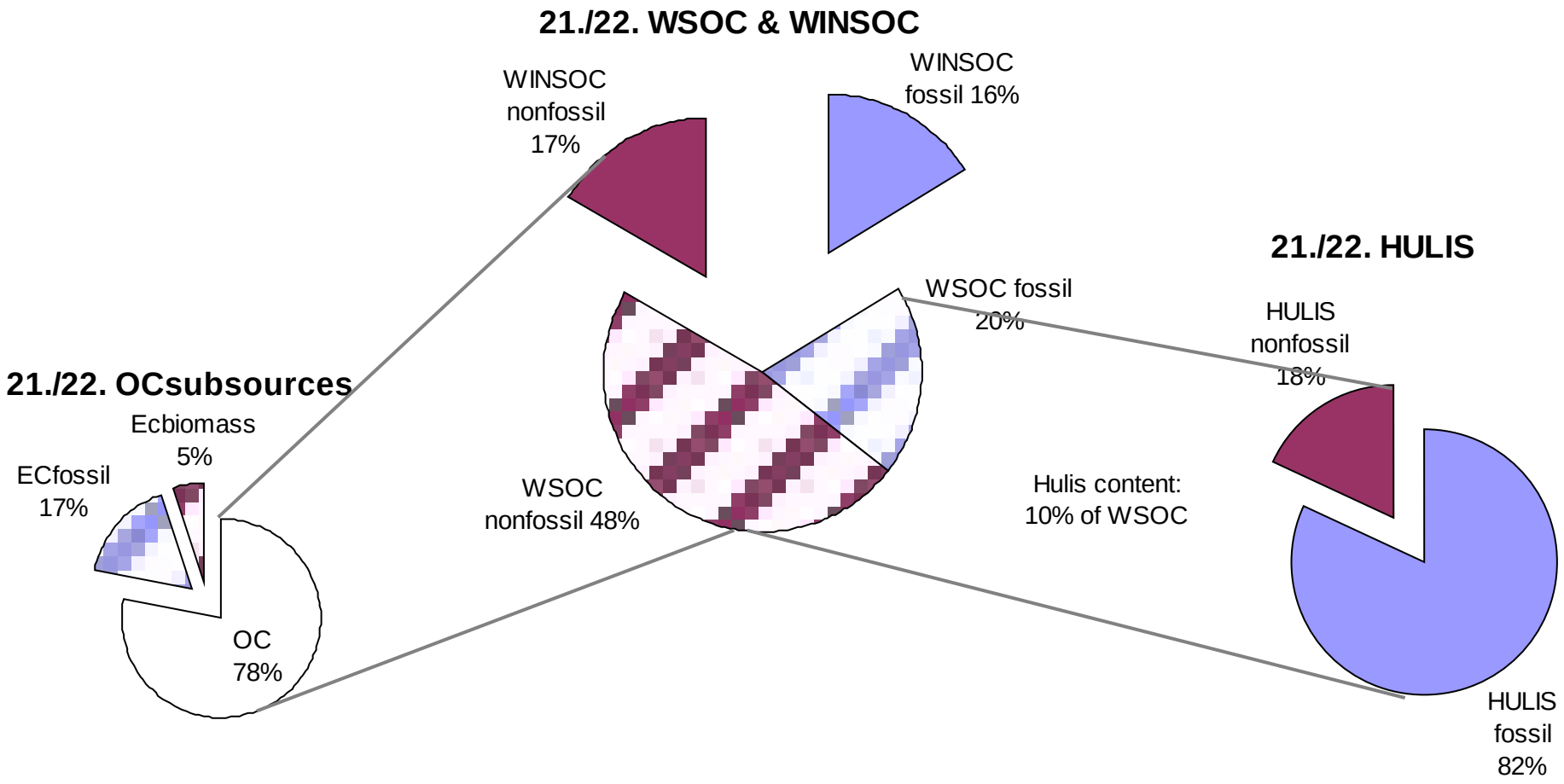


- Average TC: 4.0 µg/m^3 San Pietro
- 4.8 µg/m^3 Zurich
- 3.0 µg/m^3 Gothenburg
- 24 µg/m^3 Mexico-City
- Stations Zurich and Gothenburg represent urban background

Separation of HULIS based on Limbeck et al., 2005

1) **Water extraction of the filters** **(Removal of WINSOC)**
2) **C-18 solid phase extraction** **(Removal of hydrophilic OC)**
3) **Anion exchange** **(Removal of neutral/cationic OC)**
4) **Elution with ammonia** **(Isolation of HULIS)**
5) **Evaporation to dryness**
6) **Combustion in an Elemental Analyzer and on-line ^{14}C measurement**

Fossil and non-fossil sources of subfractions of OC



Comparison between San Pietro Capofiume, Italy (summer) and Graz, Austria (Easter)

Comparison with winter filters from the area of Graz, Austria

Area	Northern Italy	Austria
Characteristics	Rural, industrialized	Rural + urban
Season	Summer	Easter
Non-fossil fraction of TC	49%	
Non-fossil fraction of OC	57%	
Non-fossil fraction of WSOC	71%	
Non-fossil fraction of HULIS	18%	106%

Current limitations

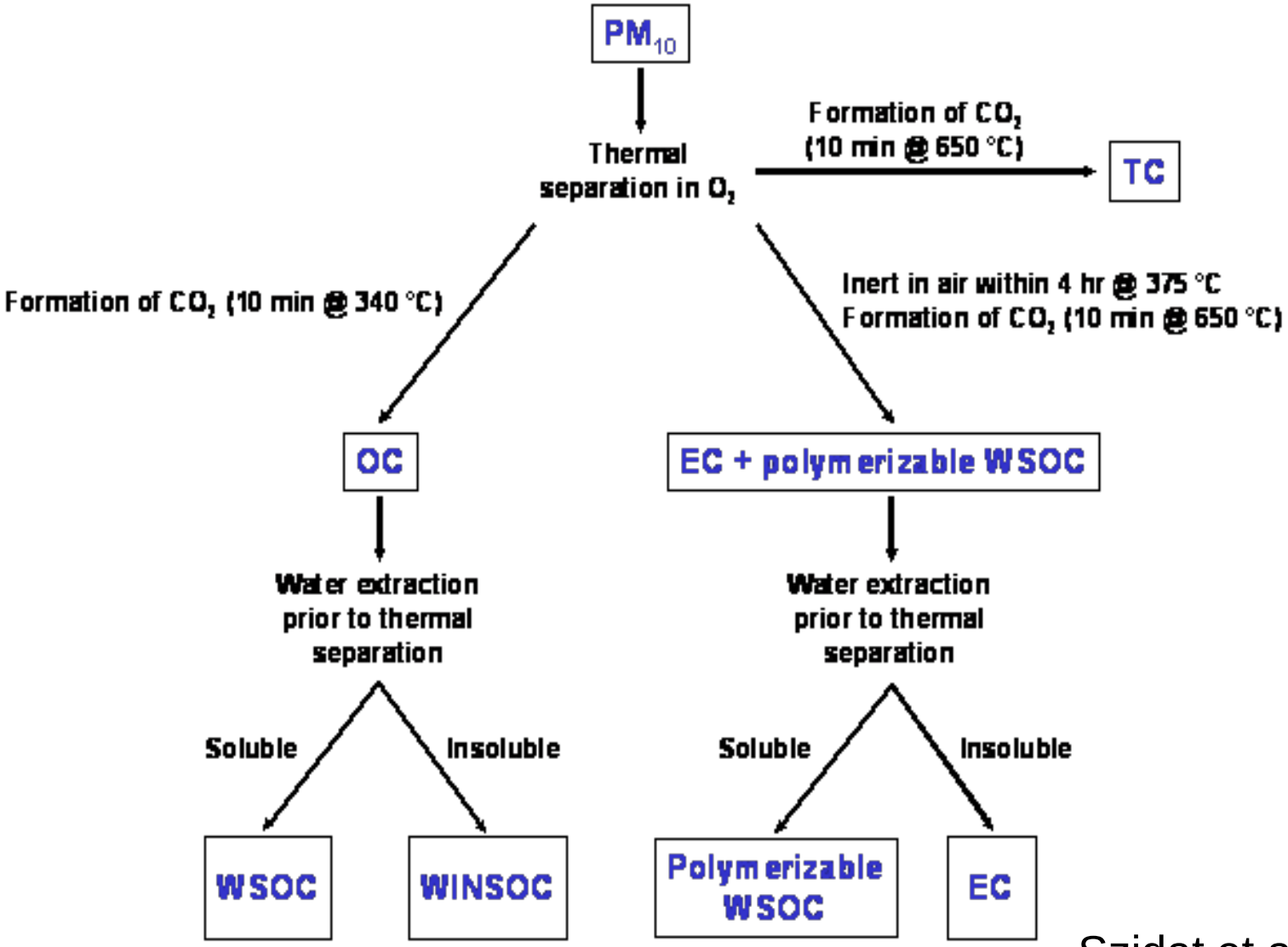
- **Overall yields for OC and EC of 70-80% induce uncertainties to the source apportionment of these fractions with ^{14}C**
  See poster of Nolwenn Perron
- **Procedure blank contributions of the ^{14}C measurement of the HULIS fraction has not been well established yet**
- **Yields of <10% for the evaporation step before ^{14}C measurement of the HULIS fraction is not yet satisfying**
  ^{14}C results of HULIS are preliminary

Conclusions and outlook

- **In general:**
  ^{14}C is a unique tracer of fossil and non-fossil sources for carbonaceous aerosols
- **Samples from San Pietro Capofiume, summer:**
  For increasing contributions of SOA (TC→OC→WSOC), non-fossil sources become more important;
  HULIS stem mainly from fossil precursors
- **Samples from Austrian stations, winter:**
  HULIS originate from non-fossil sources for samples with relevant contributions from wood burning
- **Outlook 1:**
  Improvements of the HULIS isolation are necessary
- **Outlook 2:**
  ^{14}C analysis of dicarboxylic acids are under investigation

Thank you for your attention

Discrimination into different fractions before ^{14}C analysis



Szidat et al., JGR 2006

Refined source apportionment of organic carbon

- ^{14}C in carbonaceous particles
- Methods of sampling and analysis
- First source apportionment results
- Conclusions and outlook

Refined source apportionment of organic carbon

- **^{14}C in carbonaceous particles**
- **Methods of sampling and analysis**
- **First source apportionment results**
- **Conclusions and outlook**

Refined source apportionment of organic carbon

- ^{14}C in carbonaceous particles
- **Methods of sampling and analysis**
- First source apportionment results
- Conclusions and outlook

Refined source apportionment of organic carbon

- ^{14}C in carbonaceous particles
- Methods of sampling and analysis
- **First source apportionment results**
- Conclusions and outlook

Refined source apportionment of organic carbon

- ^{14}C in carbonaceous particles
- Methods of sampling and analysis
- First source apportionment results
- **Conclusions and outlook**