Composition of Particulate matter and the sources of the carbonaceous fraction in urban and rural areas in Switzerland

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In recent years, there have been measurements in Switzerland including aerosol mass spectrometry (AMS), ^{14}C analyses, and aethalometer measurements, partially in conjunction with off-line analyses of ions or individual elements. Most of the AMS measurements were recently summarized by Lanz et al. (2009). It was found that nearly at all sites, the contribution of secondary organic aerosols was higher than the primary organics from traffic or wood burning (Figure 1).

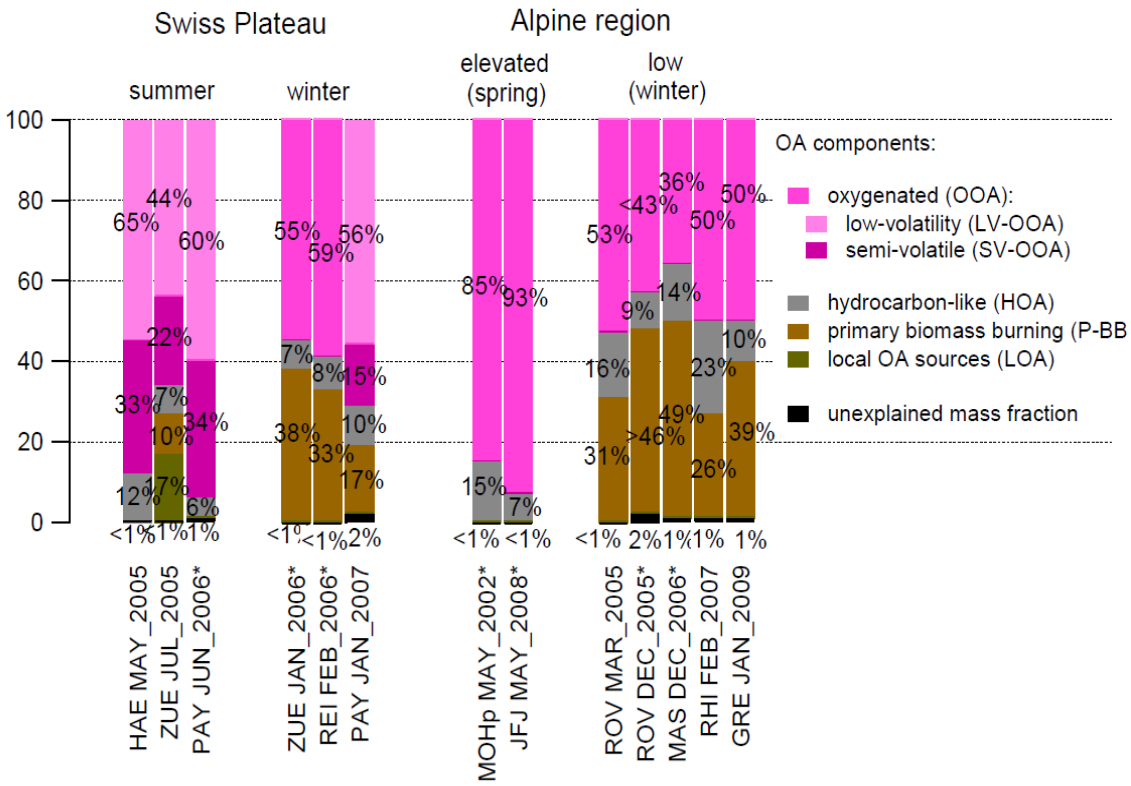

Figure 1: Relative contribution of organic components during different campaigns in Switzerland, Germany and France. The station names include HAE: Härkingen, ZUE: Zürich, PAY: Payerne, Rei: Reiden, MOHp: Hohenpeissenberg, JFJ: Jungfraujoch, ROV: Roveredo, MAS: Massongex, RHI: Rheintal, GRE: Grenoble. OOA is mostly due to secondary organic aerosols, HOA mostly due to traffic, primary biomass burning reflects the wood burning fraction in winter time, in summer it might be open fires and in general biomass burning.

During winter, wood burning is often the most important primary particulate organic source. A high impact has been shown not only for urban and rural areas in Switzerland but also for big cities like Paris and different sites between Portugal and Scandinavia. ^{14}C analyses showed that most of the organic carbon stem from non-fossil sources while the elemental carbon is dominated by fossil contributions at most sites. Comparisons between rural and urban locations reveal that the pollution is often governed by the regional background even in urban areas especially in winter during high PM10 episodes. Spatially resolved local contributions in the Zurich city center are found to be mostly from traffic exhaust (Figure 2) and re-suspended coarse mode particles (Bukowiecki et al., 2010). Both local contributions are strongly influenced by truck traffic. Mobile aerosol mass spectrometer and Multi Angle Absorption Photometer (MAAP) measurements were used on a specific route in the center of Zurich. First results are shown in Figure 2.

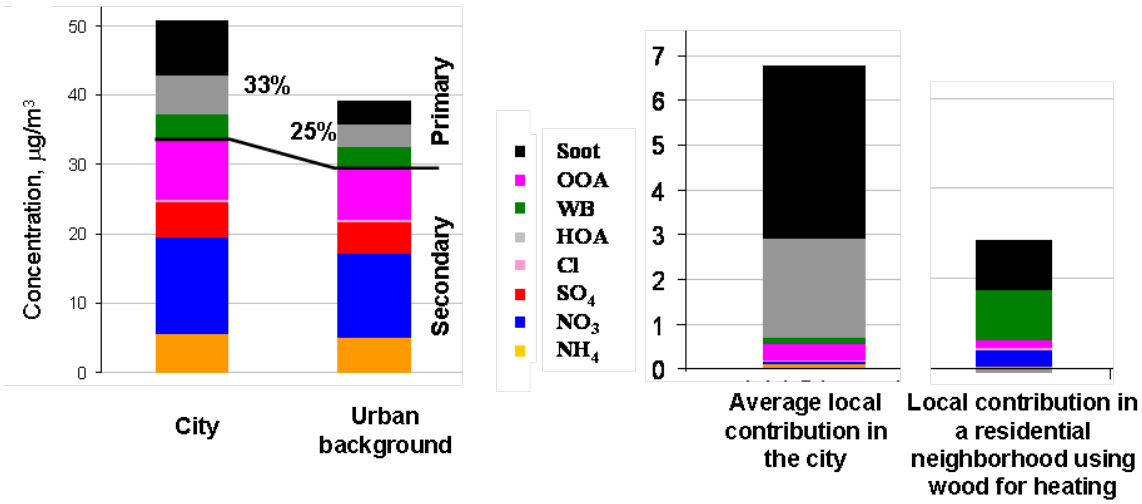

Figure 2: Average composition using mobile measurements. Urban background refers to measurements at Kaserne, Zürich while City refers to the average concentrations and chemical composition measured by the mobile PSI laboratory on-road in Zürich. The local contributions in the city versus the urban background can be assessed (Mohr et al., in preparation) (right Figure) which was done for the average and for a specific neighborhood (Erismannhof).

References

Bukowiecki, N., P. Lienemann, M. Hill, M. Furger, A. Richard, F. Amato, A. S. H. Prevot, U. Baltensperger, B. Buchmann, and R. Gehrig (2010) PM10 emission factors for non-exhaust particles generated by road traffic in an urban street canyon and along a freeway in Switzerland, Atmospheric Environment, 44(19), 2330-2340.

Lanz, V.A., A.S.H. Prevot, M.R. Alfarra, C. Mohr, P.F. DeCarlo, S. Weimer, M.F.D. Gianini, C. Hueglin, J. Schneider, O. Favez, B. D'Anna, C. George, U. Baltensperger (2009) Characterization of aerosol chemical composition by aerosol mass spectrometry in Central Europe: an overview, ACPD, 9, 24985-25021.

Zusammensetzung des Feinstaubs und Quellen der Kohlenstofffraktion im städtischen und ländlichen Raum

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Paul Scherrer Institut

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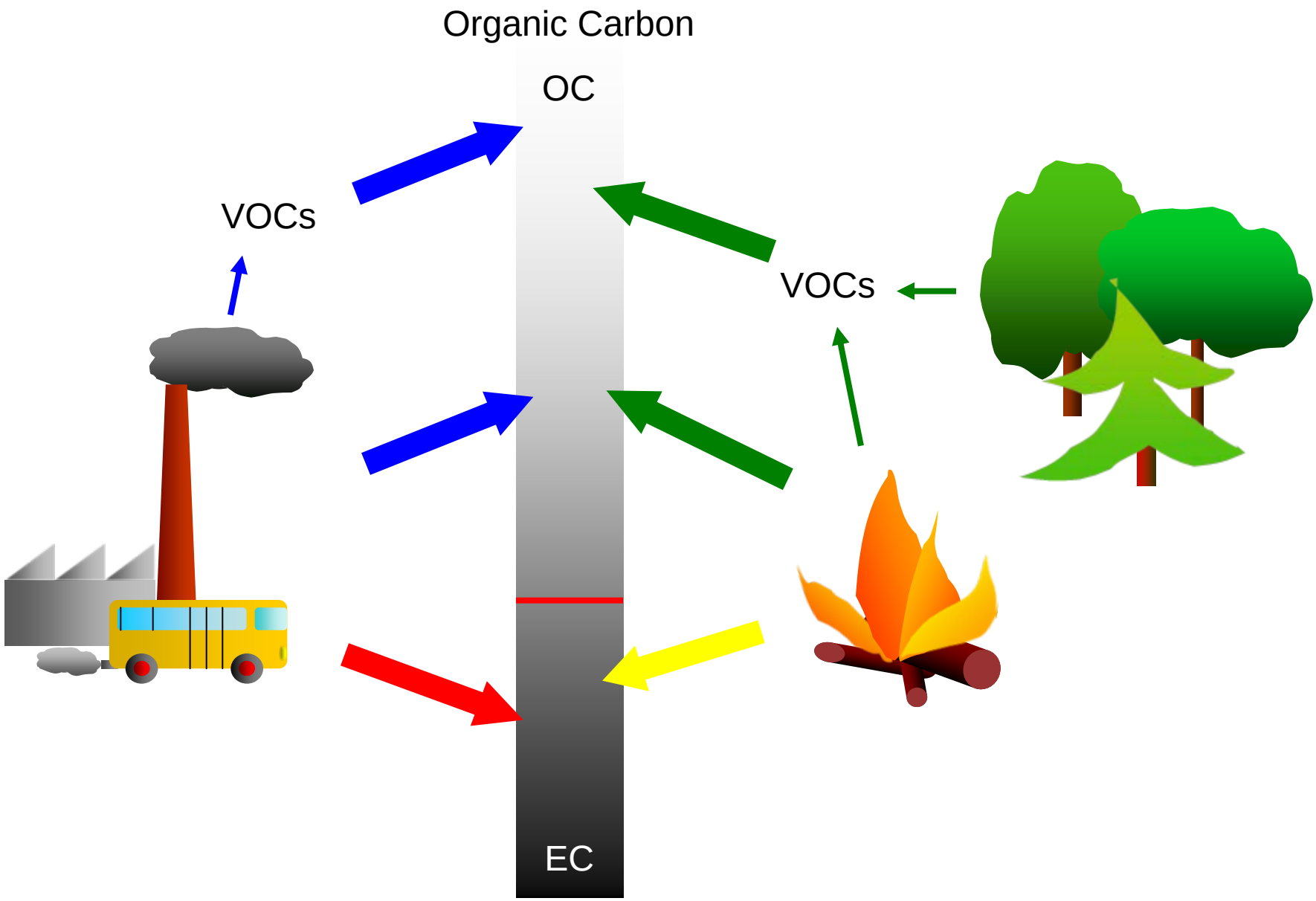
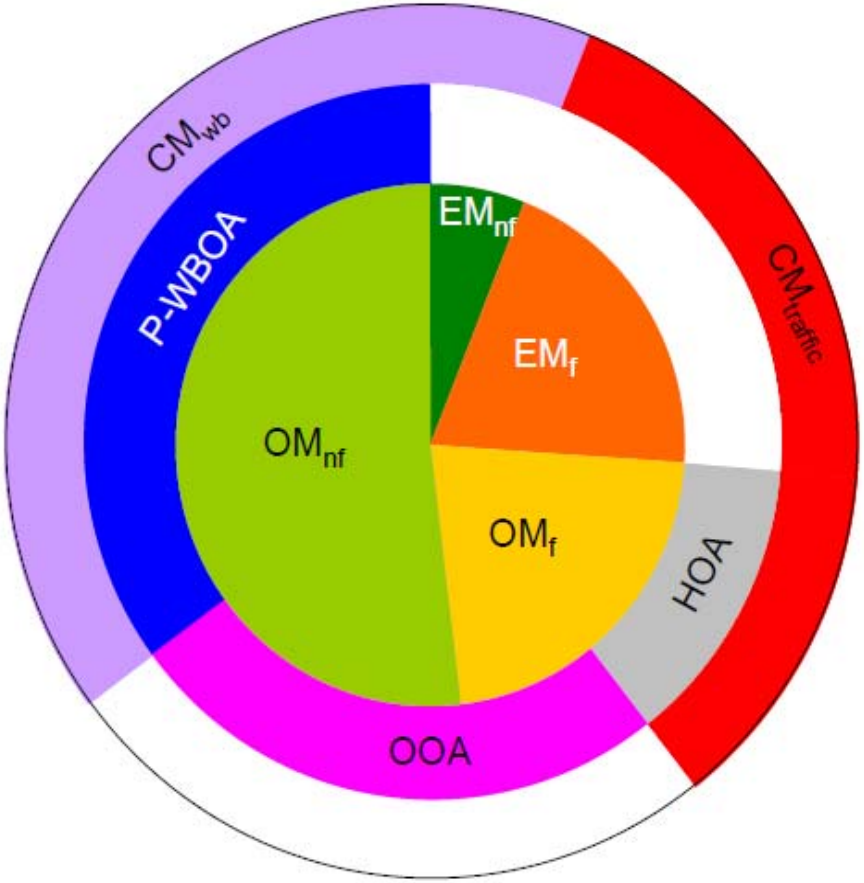
Carbonaceous aerosols and their sources



Fractions of the carbonaceous aerosol that is apportioned by the AMS, Aethalometer, ^{14}C method



Aethalometer model

- Wood-burning carbonaceous matter
- Traffic carbonaceous matter

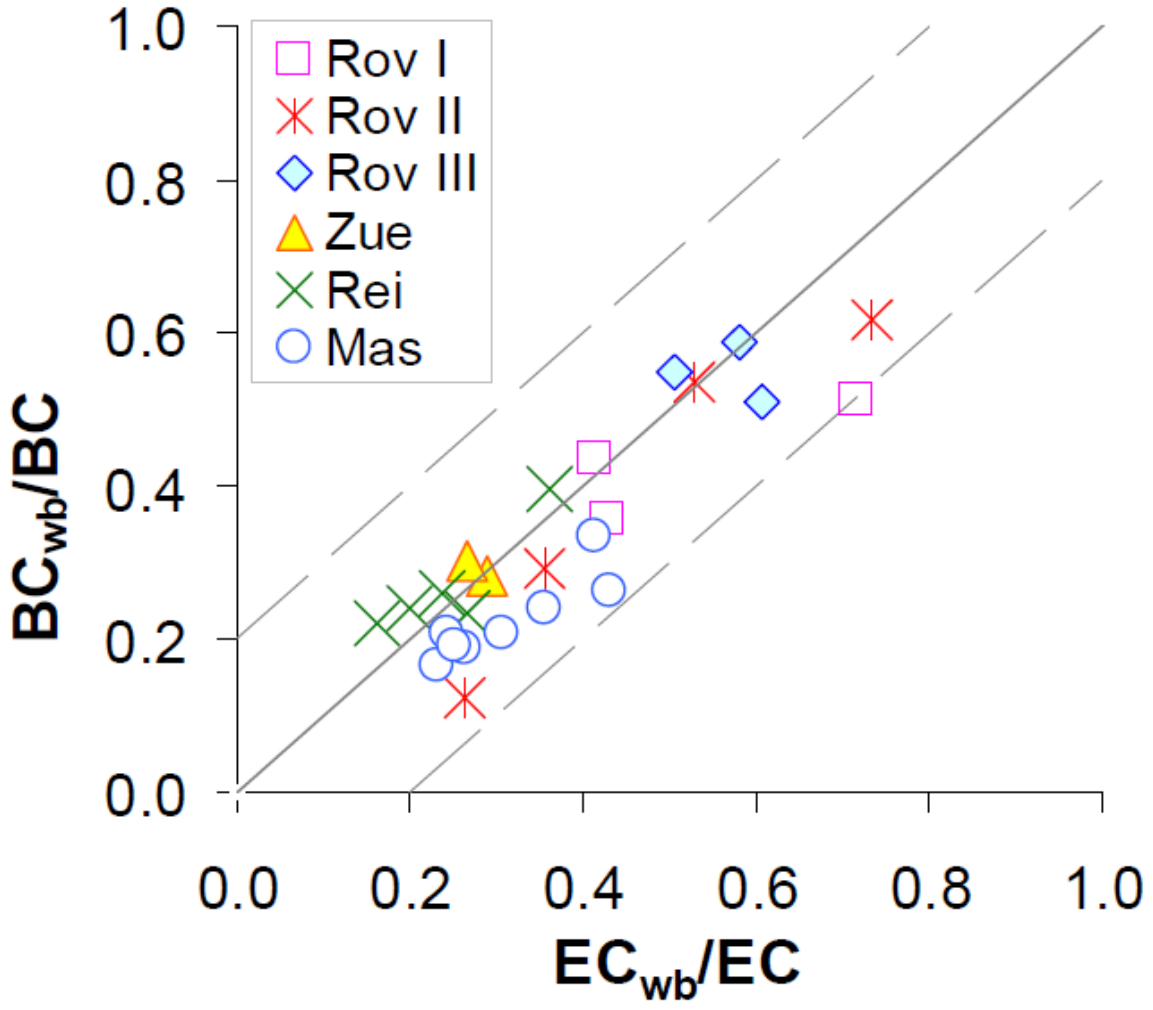
FA-AMS

- Primary wood-burning organic aerosol
- Oxygenated organic aerosol
- Hydrocarbon-like organic aerosol

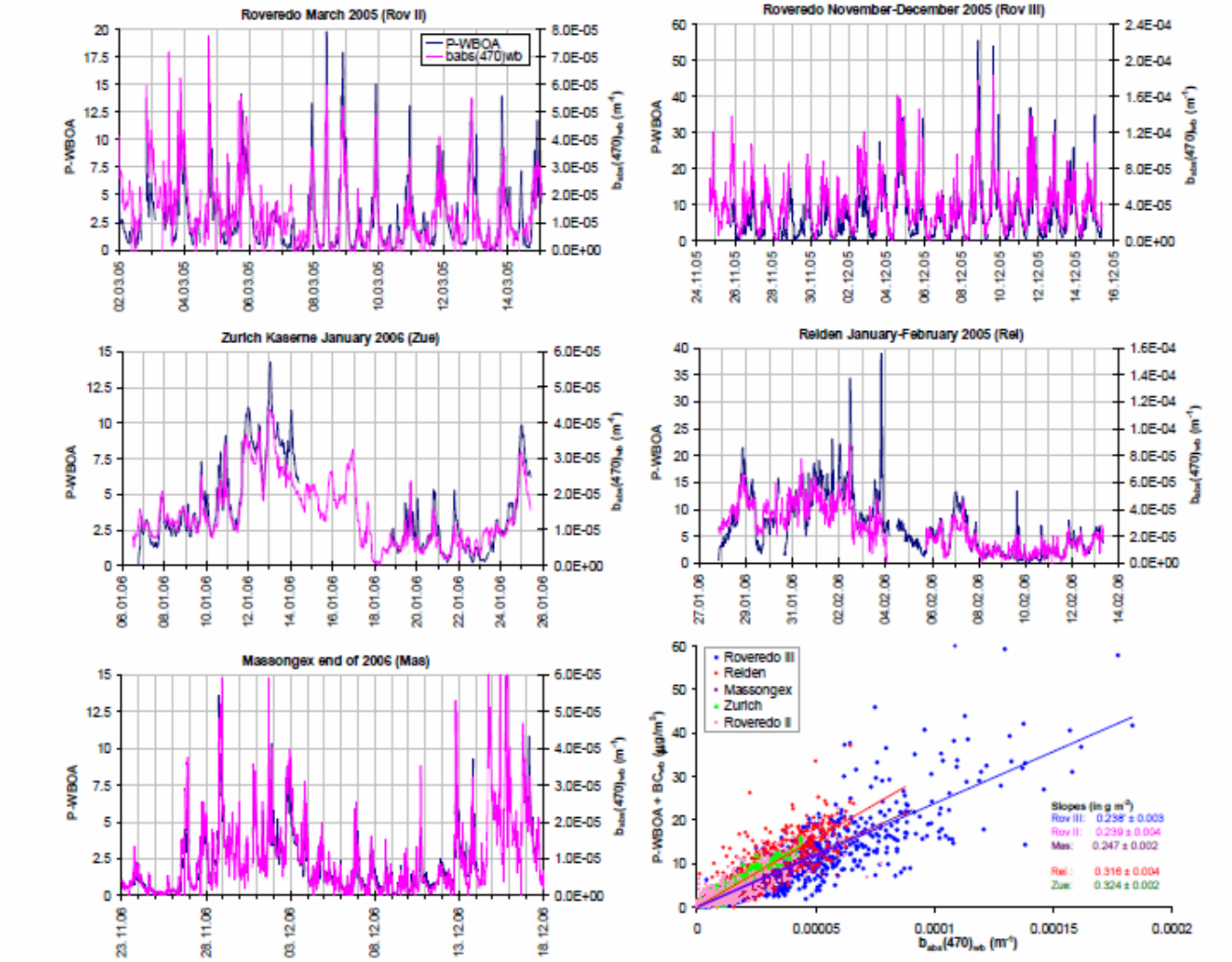
^{14}C method

- Non-fossil organic matter
- Fossil organic matter
- Fossil elemental matter
- Non-fossil elemental matter

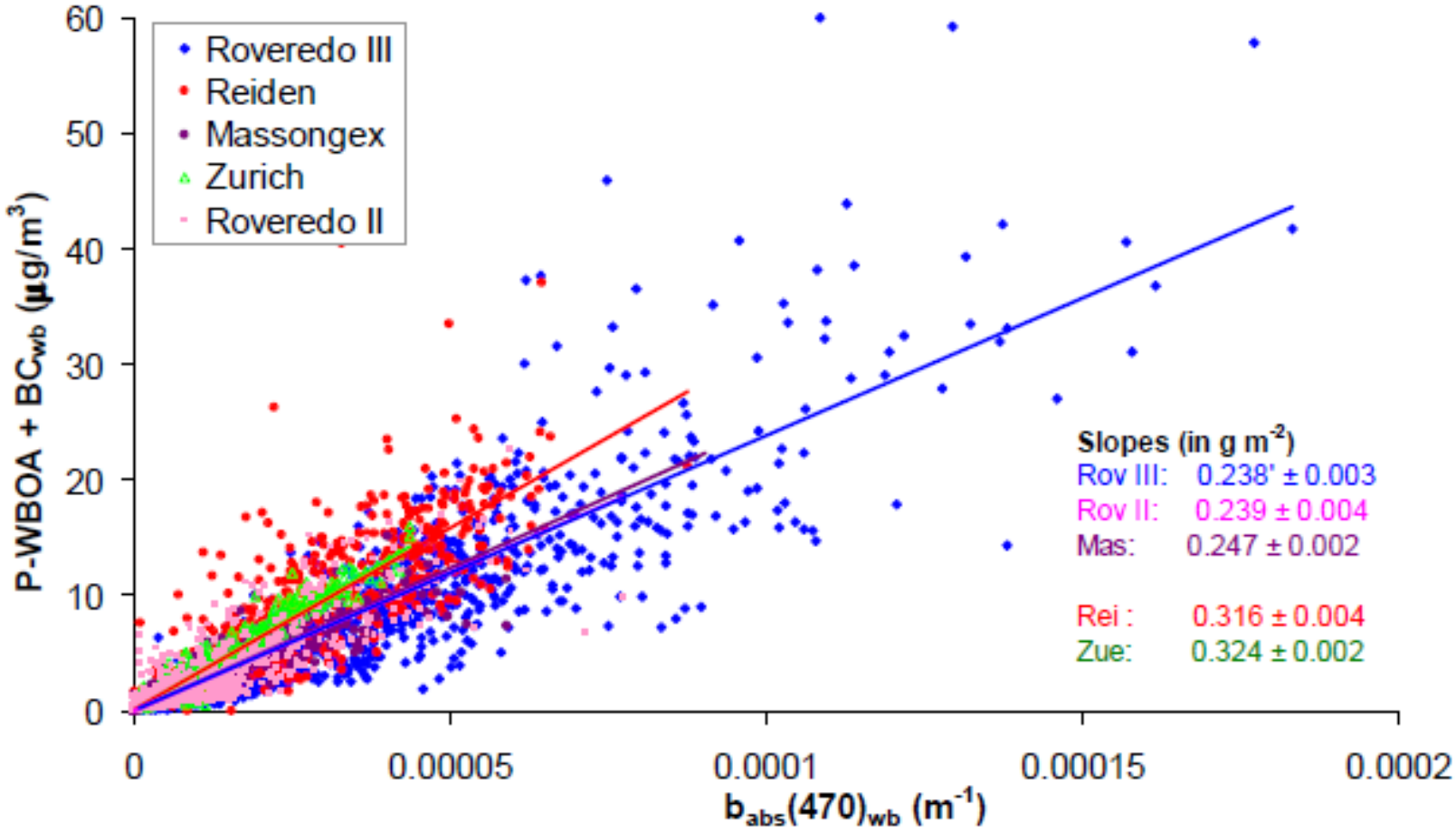
Aethalometer versus ^{14}C



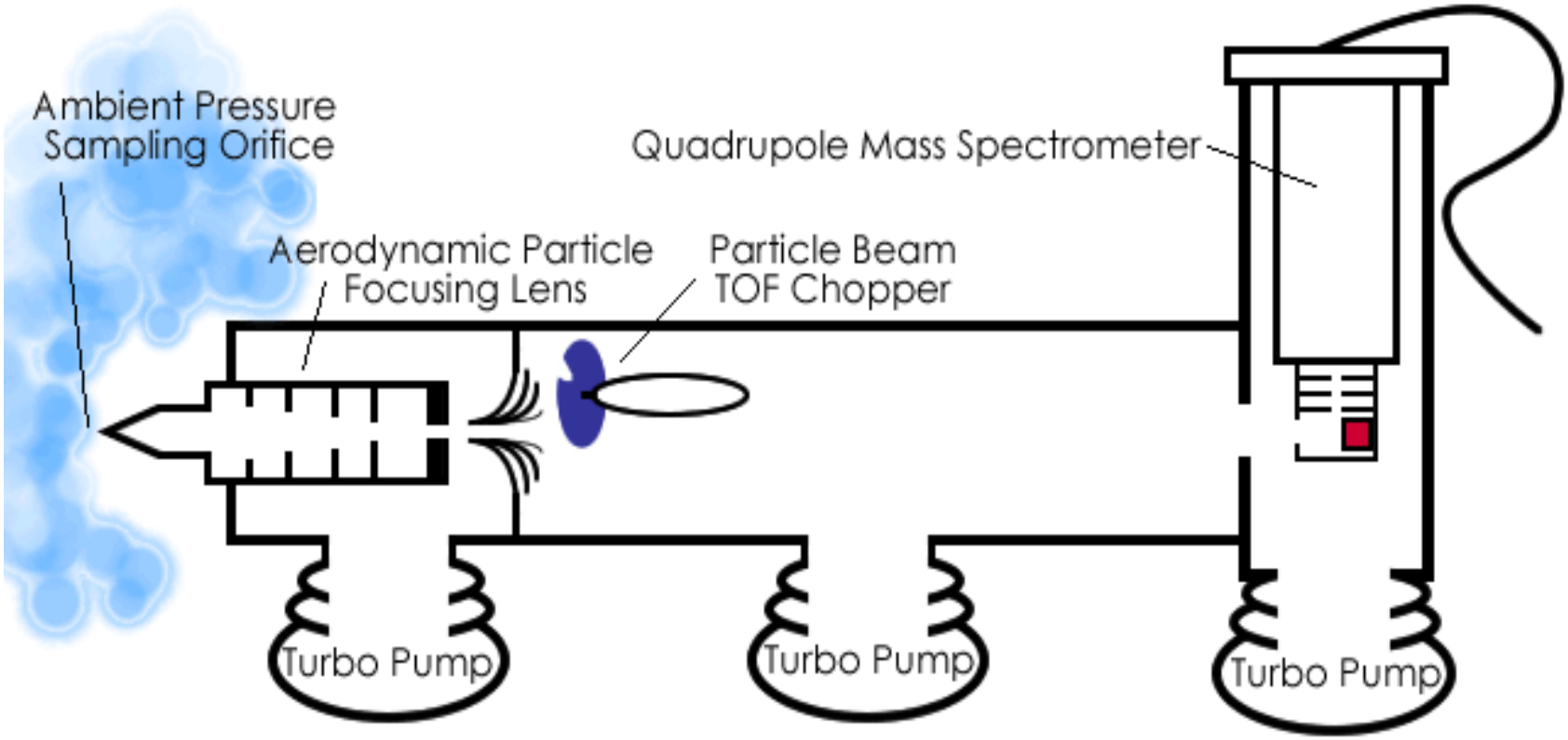
Aethalometer-Modell versus AMS analyses



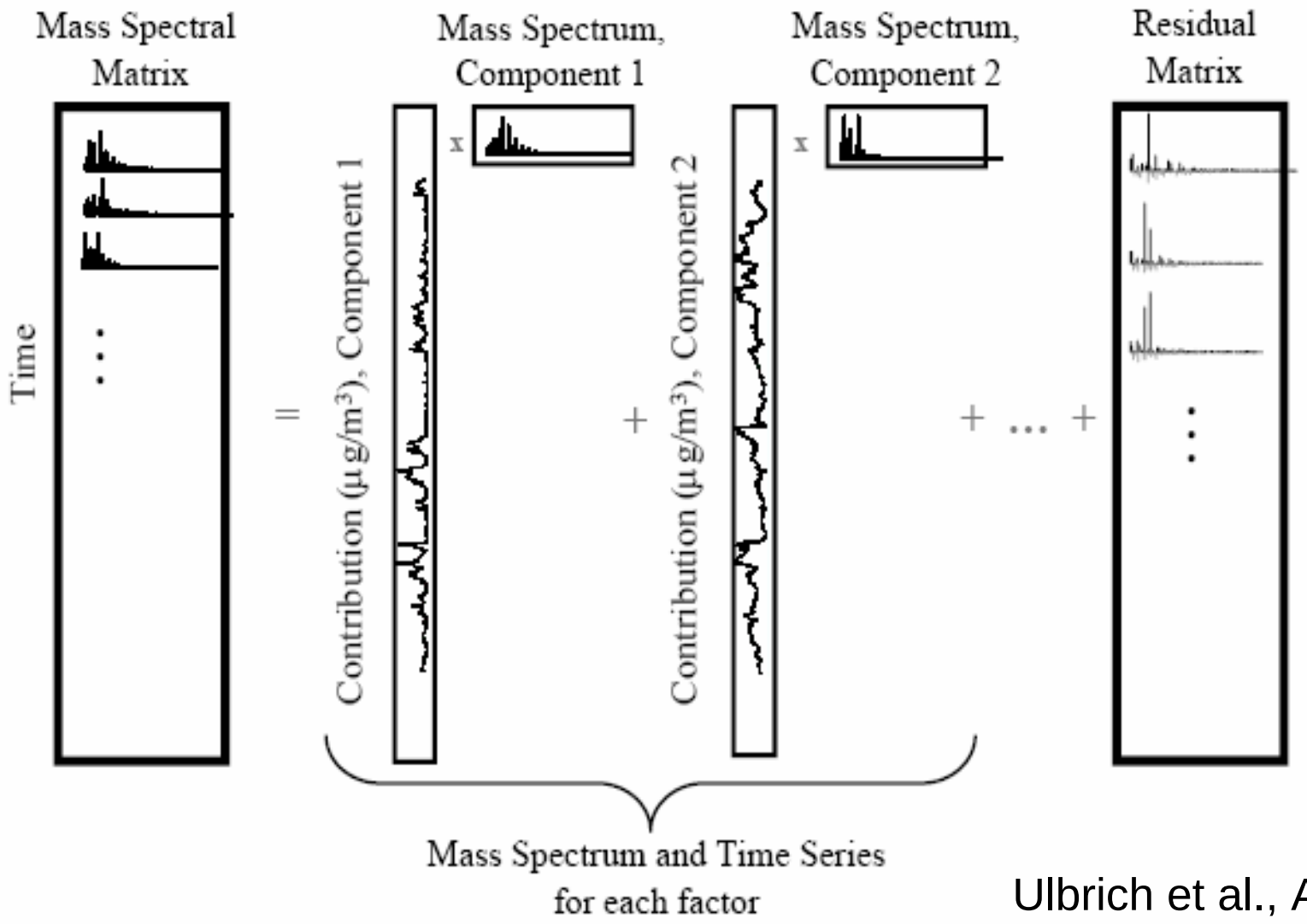
Aethalometer versus AMS measurements



Aerodyne Aerosol Mass Spectrometer (AMS)

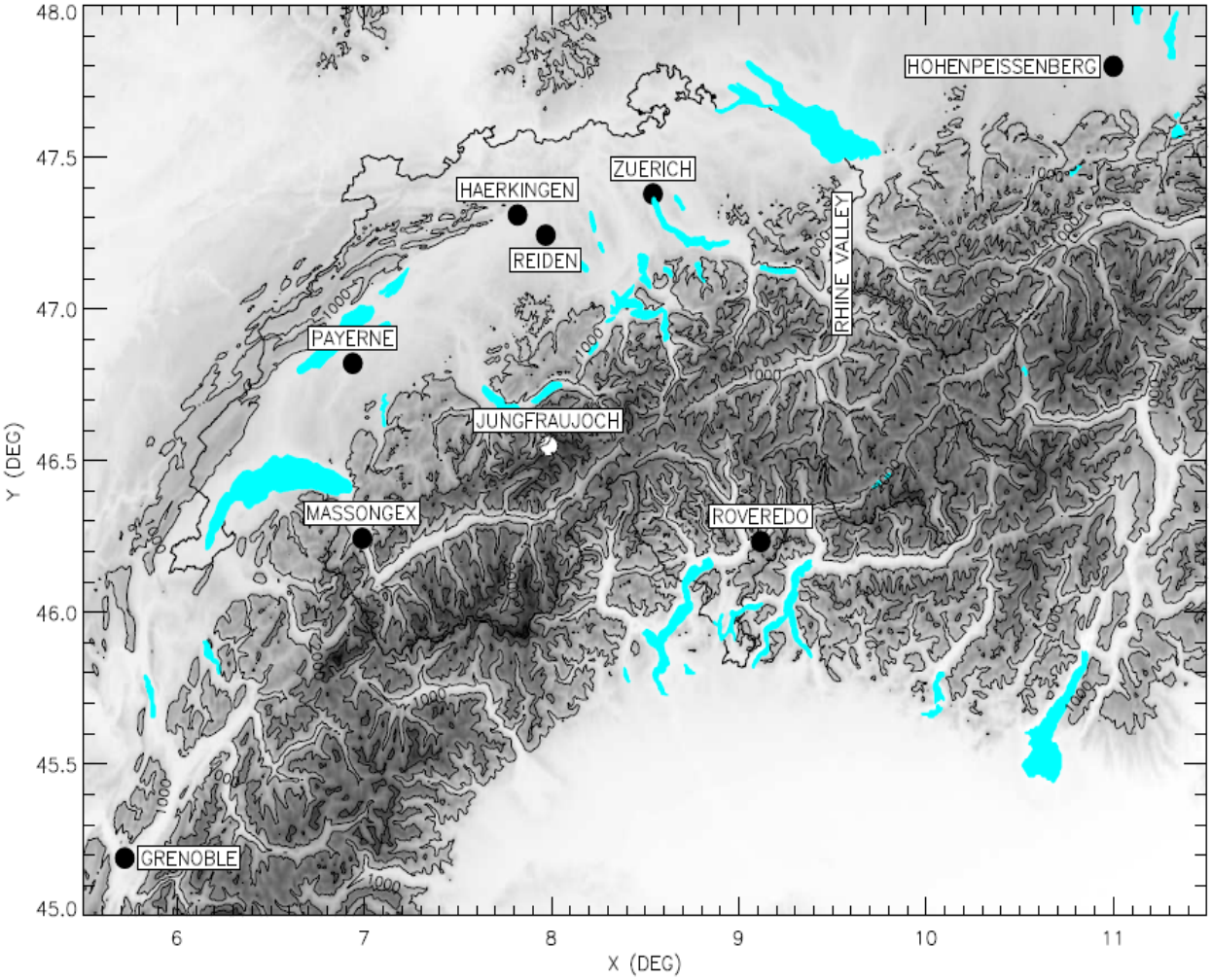


Positive Matrix Factorization (PMF) of full OM spectrum for source identification and attribution

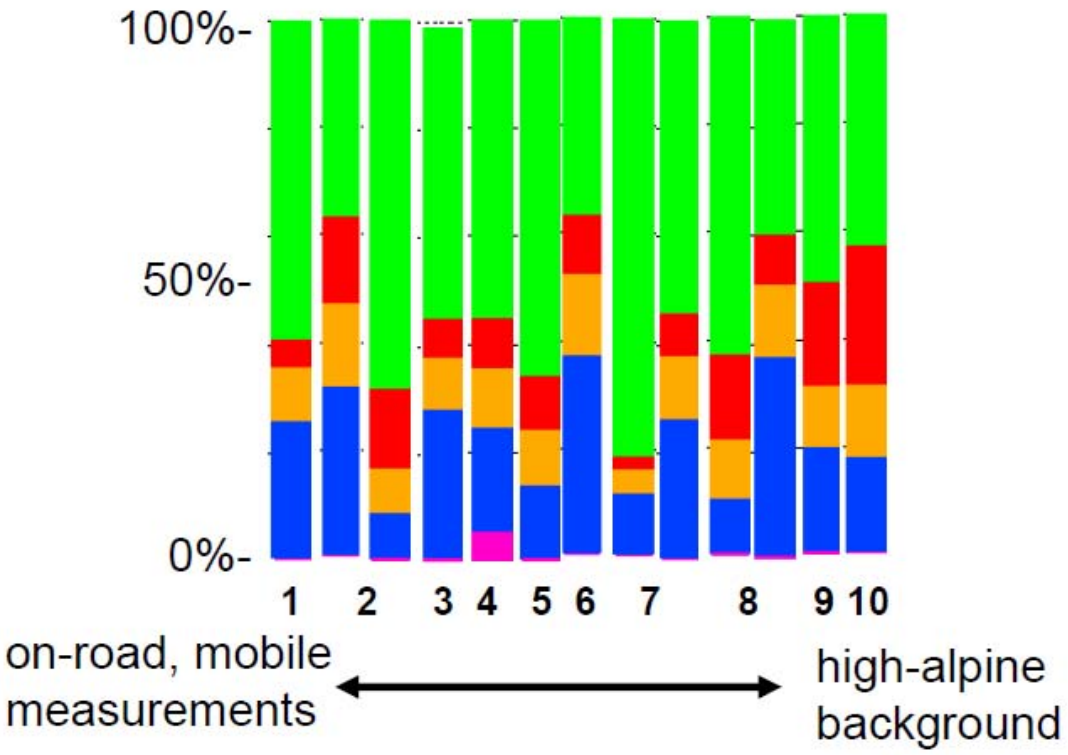


Ulbrich et al., ACP 2009

Field campaigns in Central Europe (previous and next graph)



Average relative contribution to non-refractory PM1



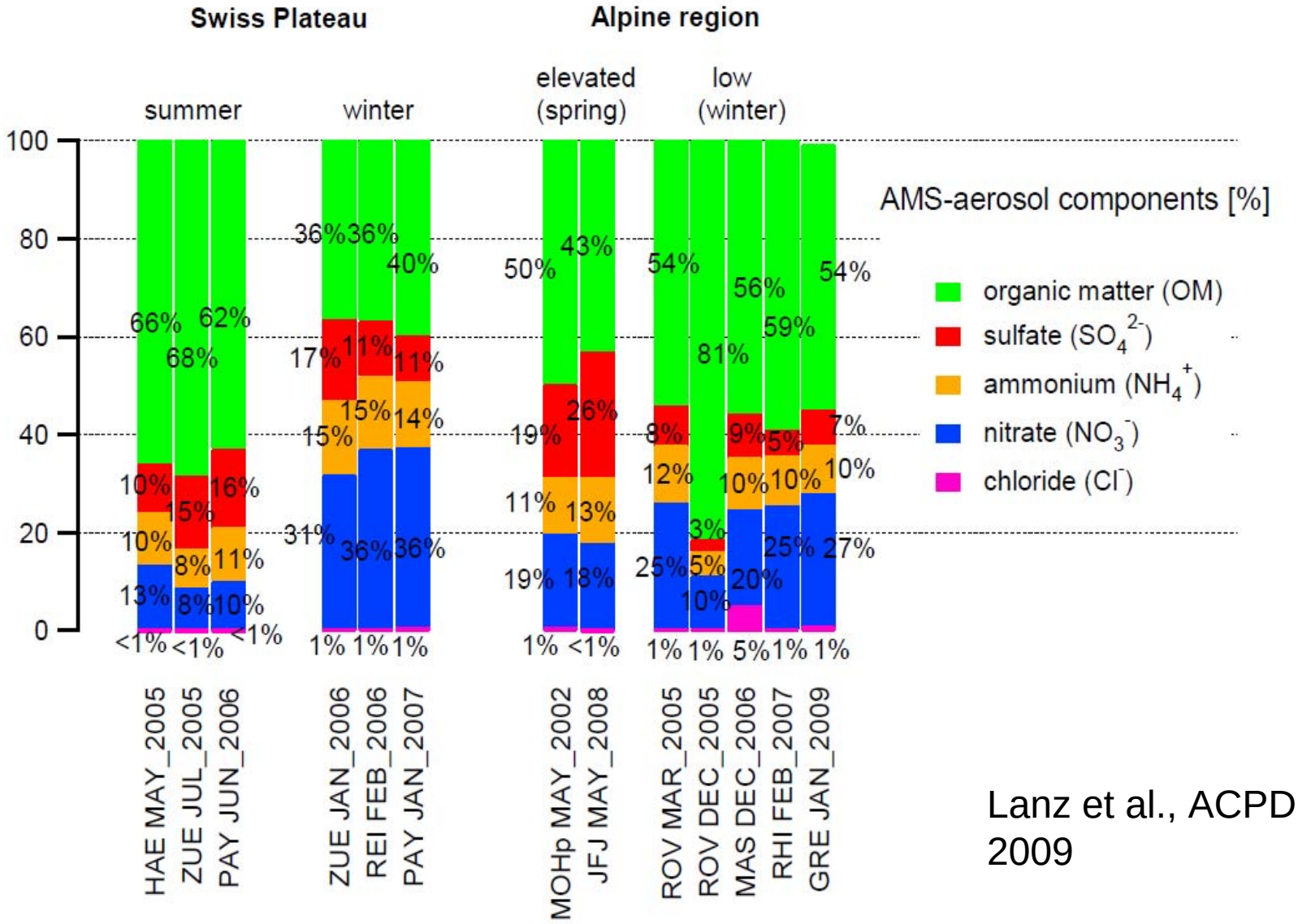
AMS-aerosol components [%]

36-81%	organic matter (OM)
3-26%	sulfate (SO_4^{2-})
5-15%	ammonium (NH_4^+)
8-36%	nitrate (NO_3^-)
0-5%	chloride (Cl^-)

large aerosol variability,

no obvious pattern

Composition of PM_1 (without BC) in Central Europe around the Alps

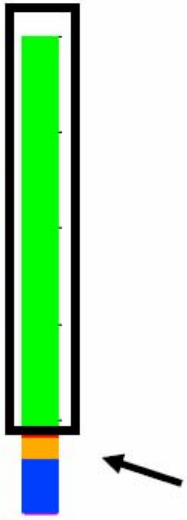


Lanz et al., ACPD, 2009

High contribution of organic mass in Roveredo

Local wood burning emissions:
77% of housholds use wood stoves
94% OM non-fossil *Szidat et al.*

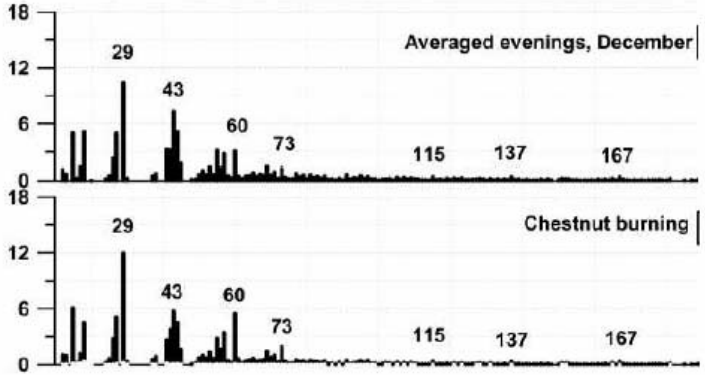
Stable thermal inversions
(„smog chamber“)



AMS-aerosol components [%]

- organic matter (OM)
- sulfate (SO_4^{2-})
- ammonium (NH_4^+)
- nitrate (NO_3^-)
- chloride (Cl^-)

Outlier: Roveredo (>80% OM)

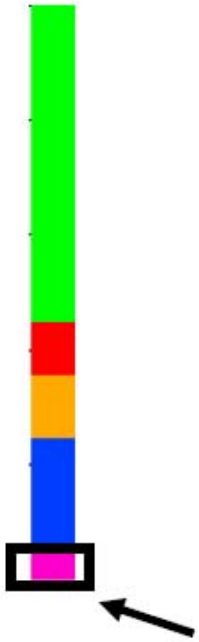


Alfarra et al., 2007, ES&T

High contribution submicron chloride in Massongex

Industrial HCl emissions:
$HCl,g + NH_3,g \leftrightarrow NH_4Cl,aer$

HCl from de-icing salts:
$NaCl,s + HNO_3,g \rightarrow NaNO_3,s + HCl,g$

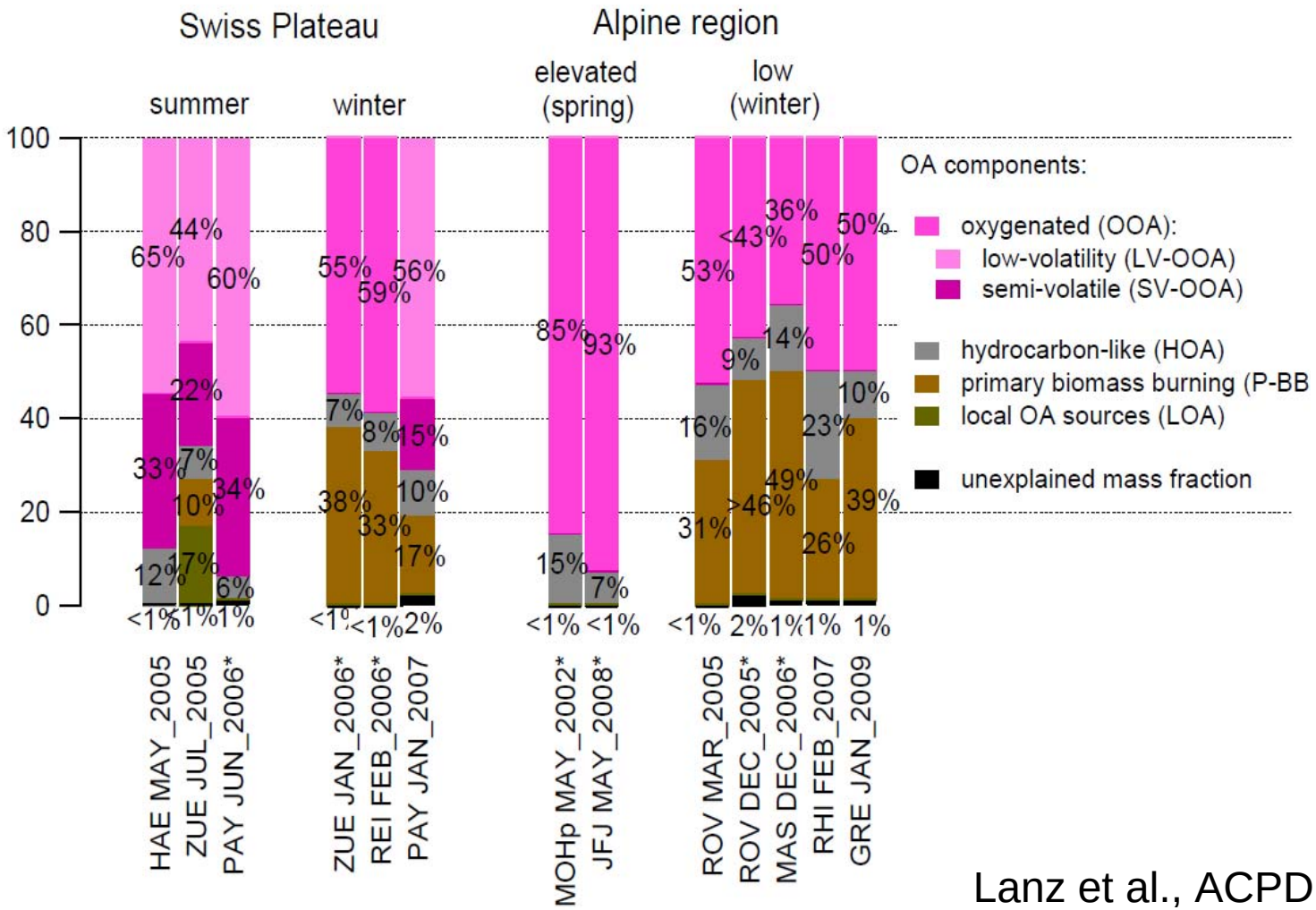


AMS-aerosol components [%]

- organic matter (OM)
- sulfate (SO_4^{2-})
- ammonium (NH_4^+)
- nitrate (NO_3^-)
- chloride (Cl^-)

Outlier: Massongex (~5% Cl^-)

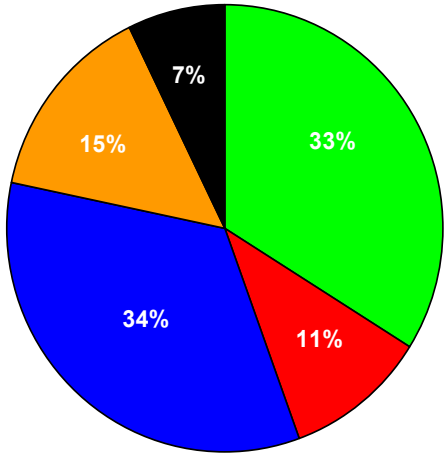
Organische Quellen von PM_1 in Zentraleuropa

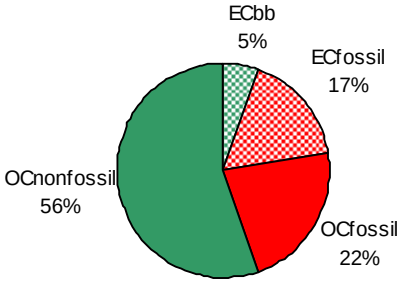


Lanz et al., ACPD, 2009

Fractions of BC, OM, NO_3, SO_4 and NH_4 and fossil and non-fossil contribution to EC and OC

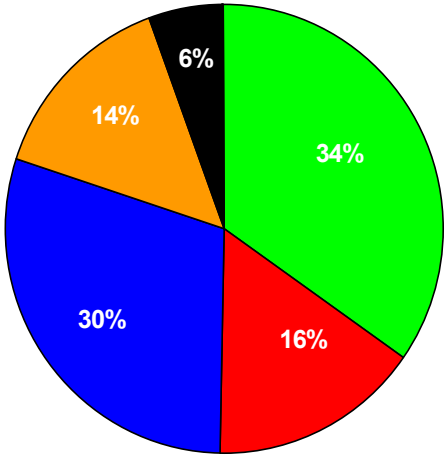
Reiden

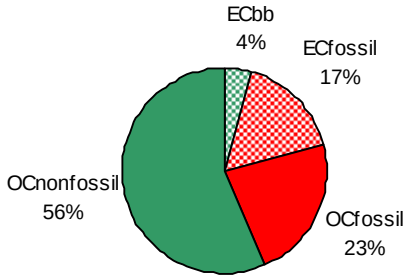




Black Carbon
Organic mass
Nitrate
Sulfate
Ammonium

Zürich

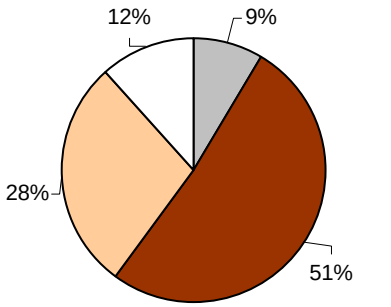


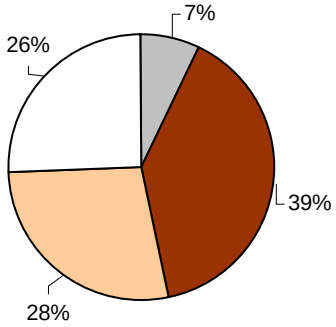


Nearly identical composition at urban background and rural highway station

Fossil and non-fossil primary and secondary organics on the Swiss Plateau

Zürich



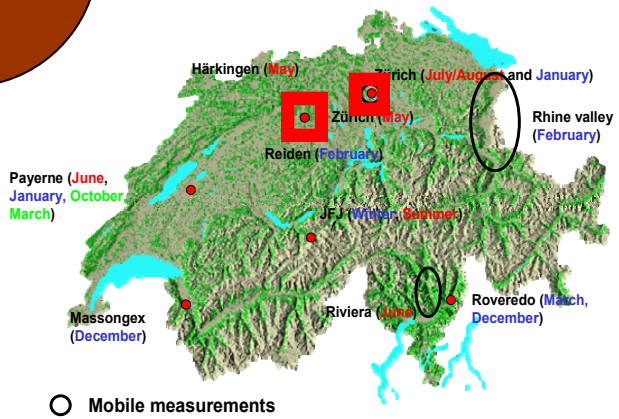


HOA (mostly traffic)

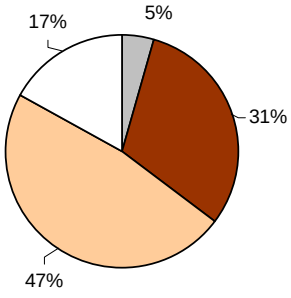
OA_wood

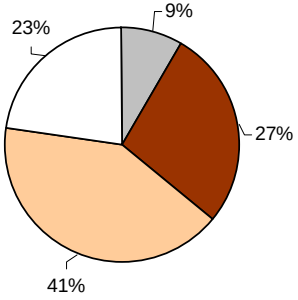
SOA non-fossil

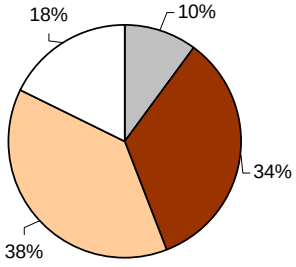
SOA-fossil

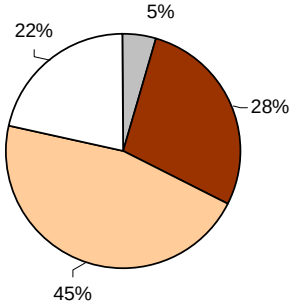


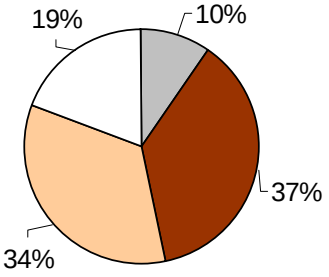
Reiden



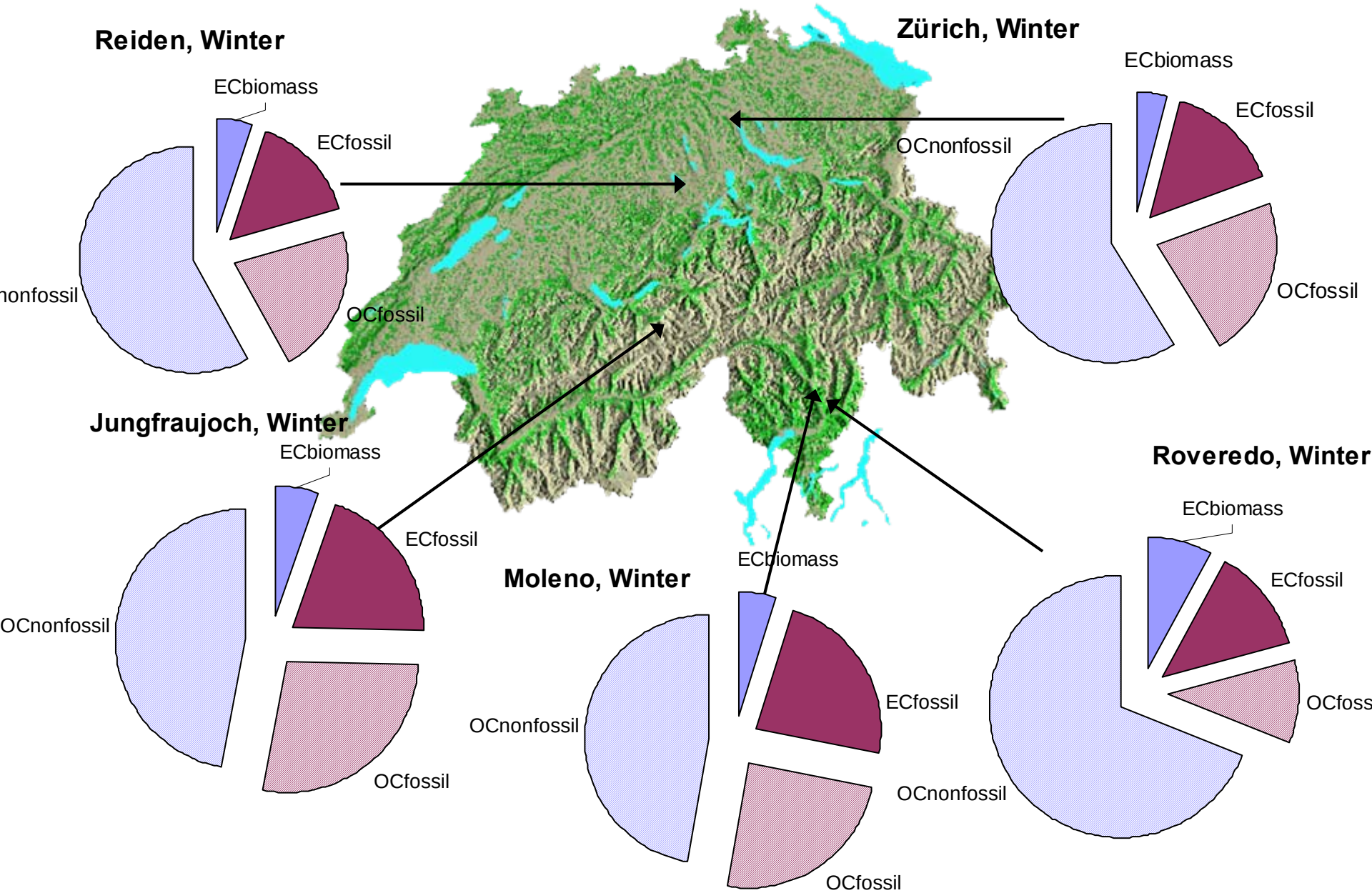




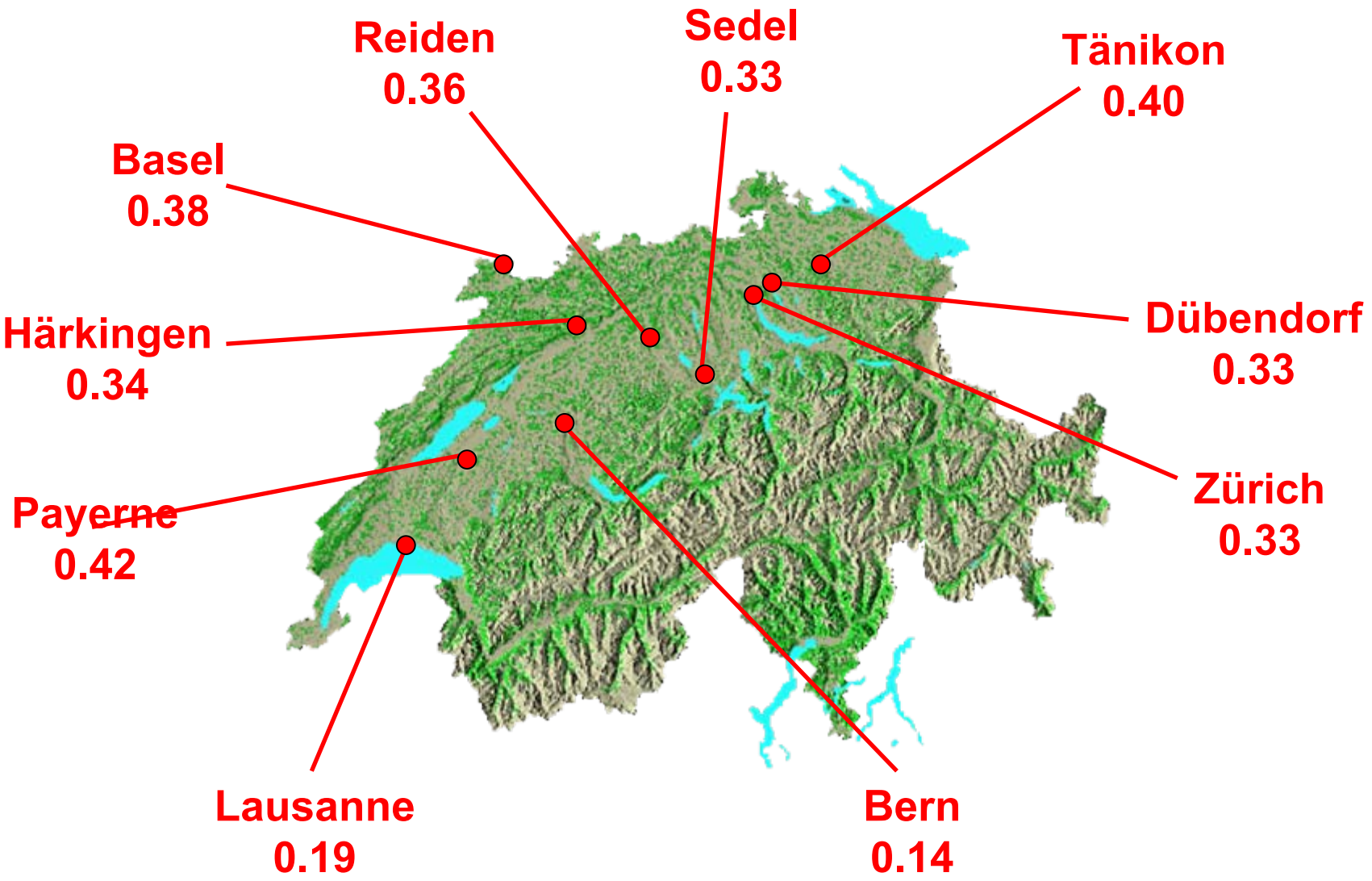




Overview over the winter ^{14}C analyses

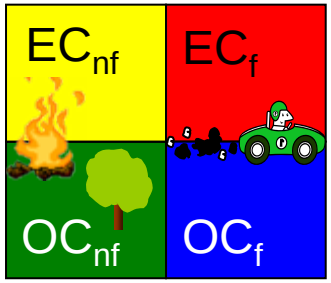


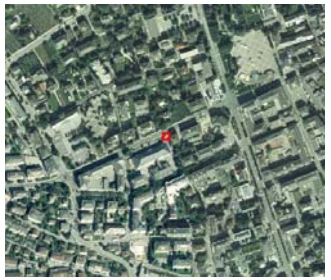
$EC_{biomass}/EC_{tot}$: Spatial distribution on 29.01.2006



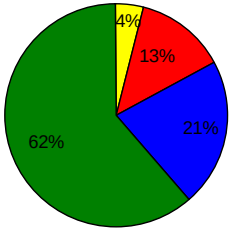
Working days and Sunday

	Massongex	Saxon	Sion	Brigerbad
	Industrial proximity	Rural zone	Town centre	Industrial proximity

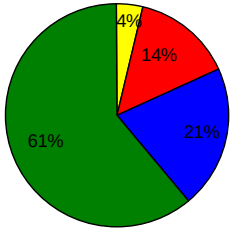




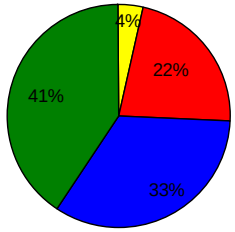
Working days



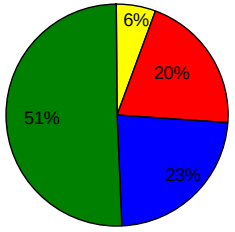
10.0 µg/m³



13.7 µg/m³

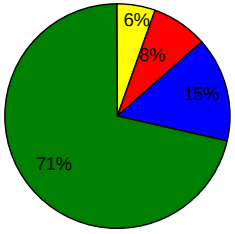


12.6 µg/m³

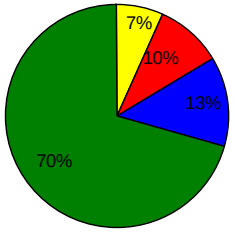


15.9 µg/m³

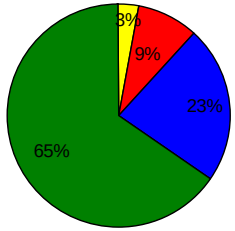
Sunday



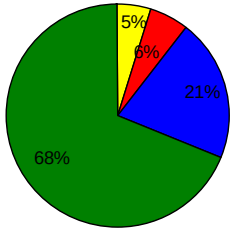
7.8 µg/m³



10.0 µg/m³



9.7 µg/m³

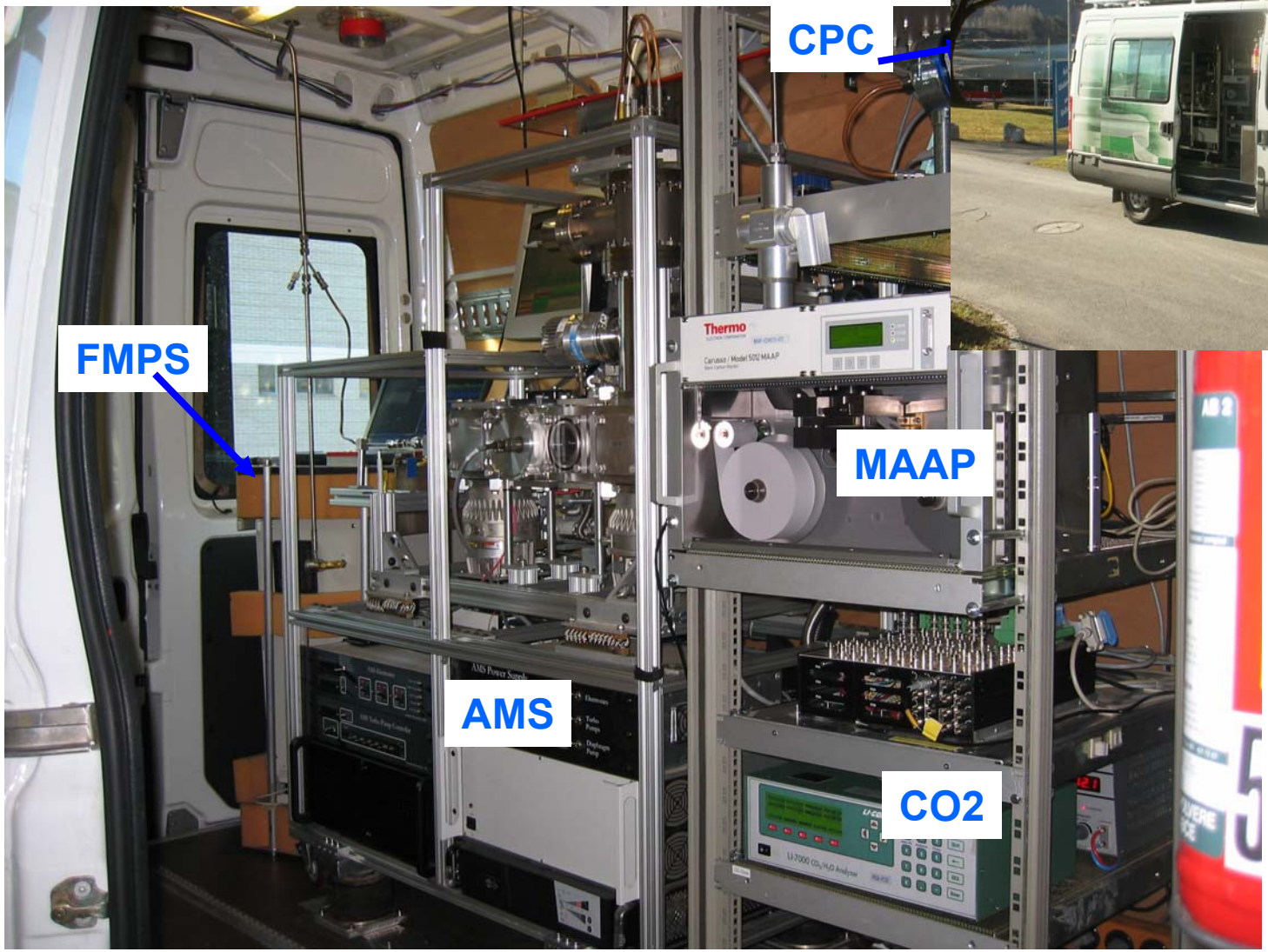


11.0 µg/m³

→ Influence of heavy-duty traffic on fossil emissions

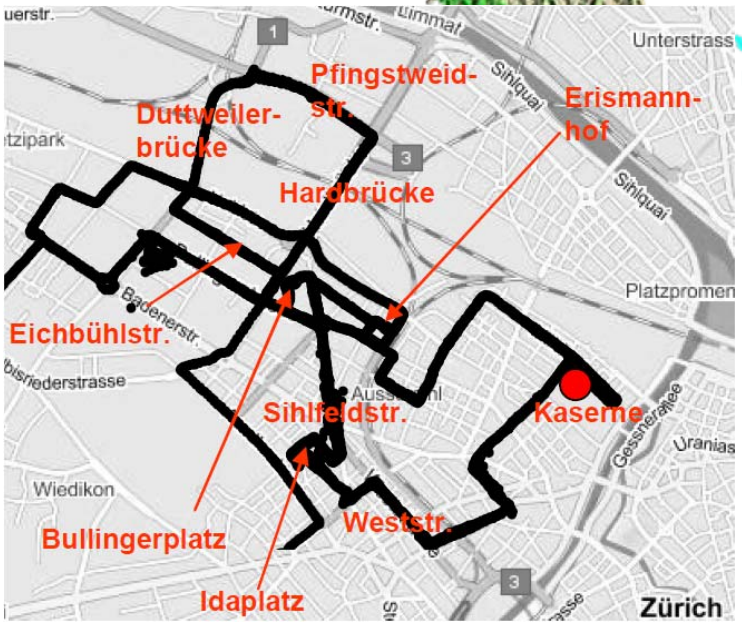
Perron et al., ACPD, 2010

The PSI mobile laboratory

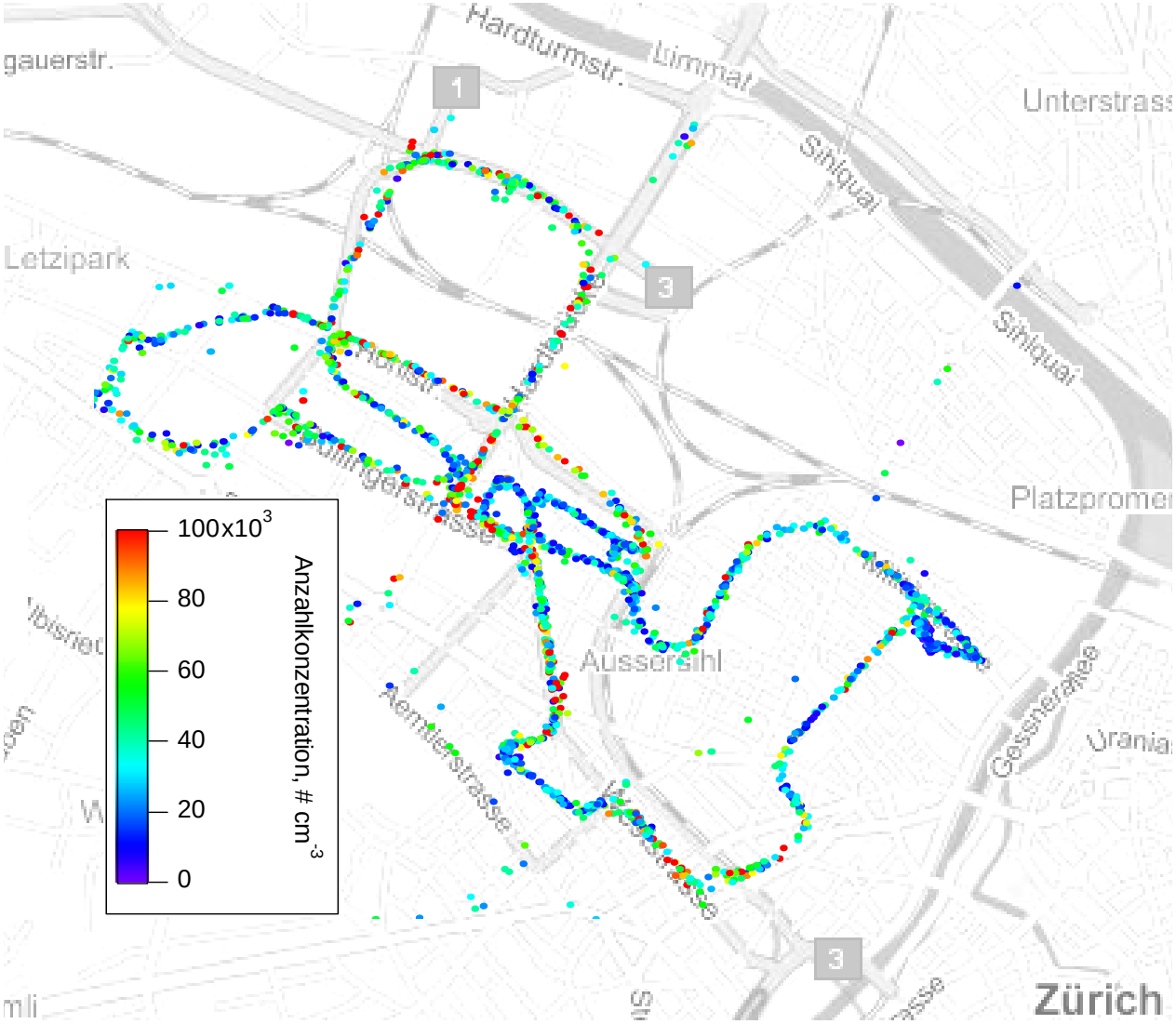


IVECO DAILY

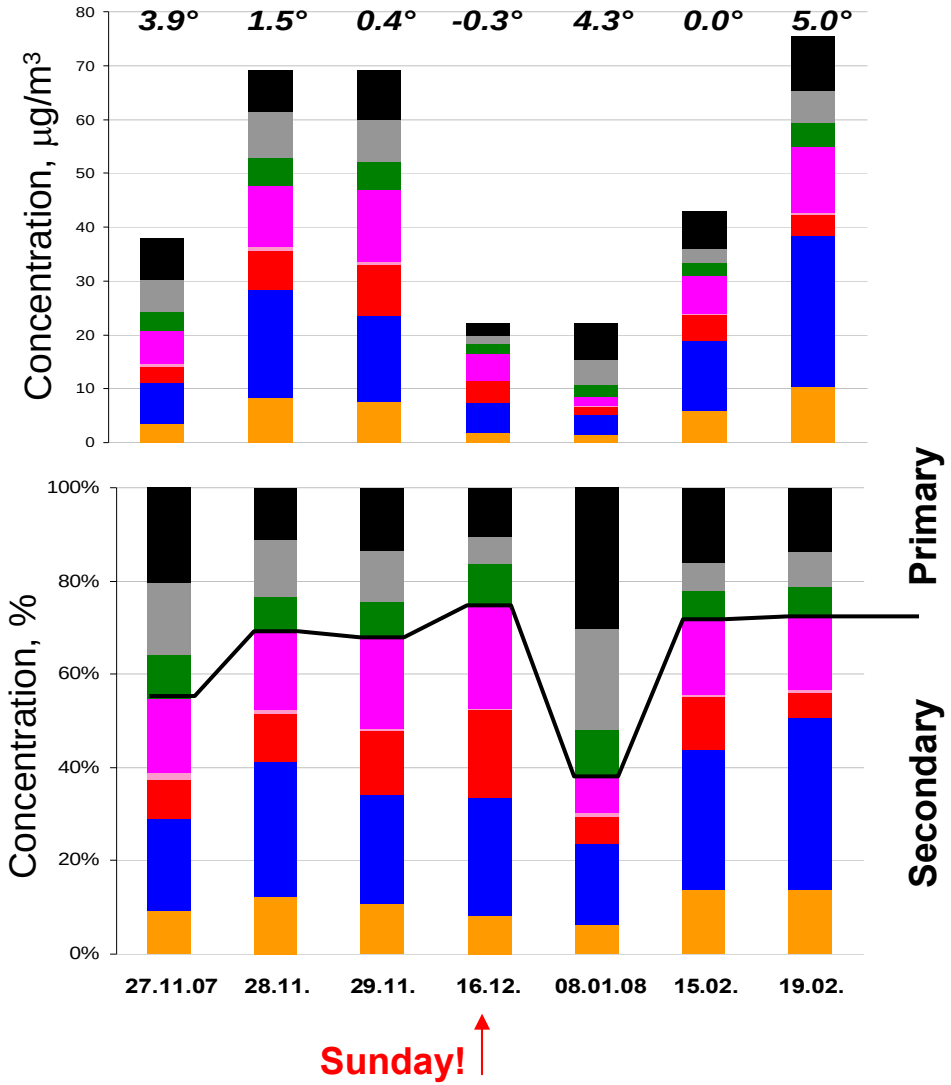
Mobile measurement route



Spatial distribution of aerosol number concentration



Composition and sources in Zürich on different days



Soot
OOA
WB
HOA
Cl
SO_4
NO_3
NH_4

OOA: mostly secondary organic aerosol

HOA: primary aerosol (mostly fossil)

WB: Wood burning

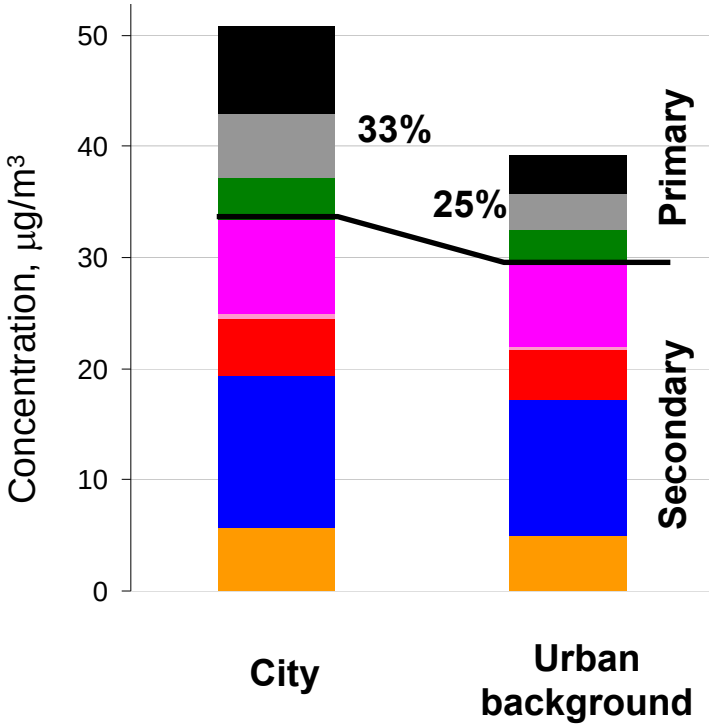
Composition in Zürich (city versus background)
Determination of local contribution

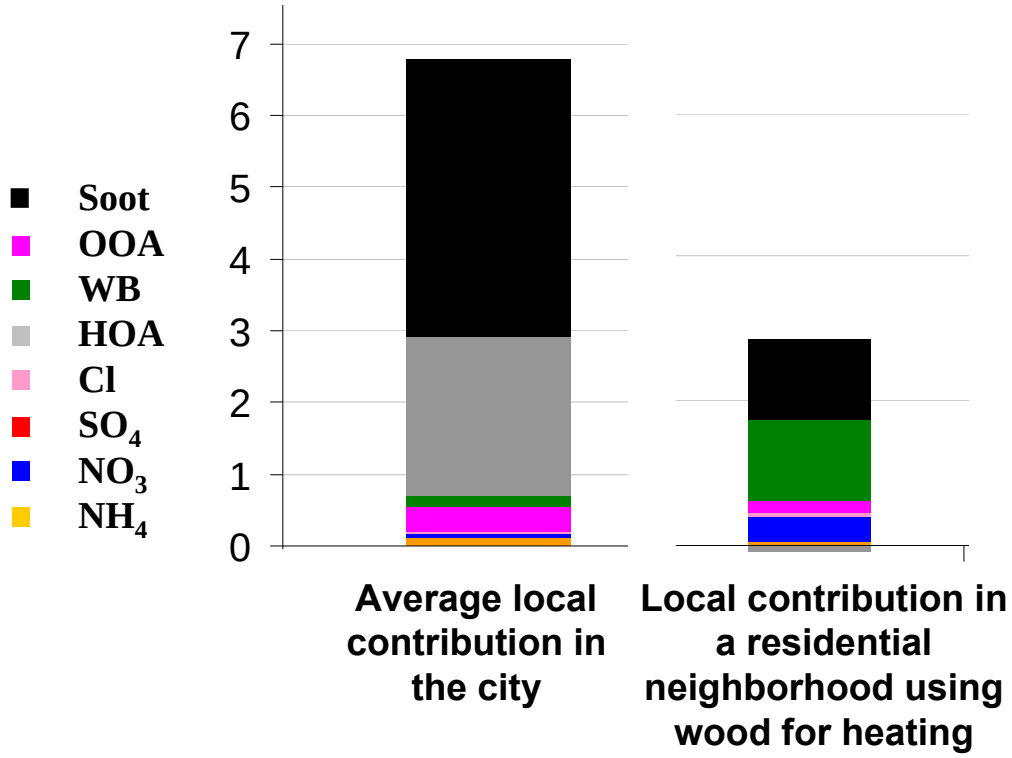
LOCAL contribution versus background

Correction of local meteorology

$$[x]_{local} = [x]_t - [SO_4]_t / [SO_4]_{background}) * [x]_{background}$$

Assumption: Production of SO_4 negligible in 1 hour. Relative composition of background stable.





Conclusions

- Most PM_1 mass is secondary, also in a city. The differences between the concentrations between rural and urban areas in Switzerland are not so high. This is different in Megacities.
- The local contribution on the streets of Zurich is mostly primary but it is mostly less than 20%, often less than 10% of the total mass in winter, except at days with regionally low concentrations
- You cannot expect miracles by traffic measures in cities. On the other hand, the reduction is not so strong but the fraction that is reduced is very toxic
- Except in some Alpine valleys, most of the EC is fossil
- Primary OC from wood burning is higher than OC from traffic in winter
- Most of the secondary OC is non-fossil, mostly wood burning related in winter
- Some weekday-weekend effects can be observed for the carbonaceous particles reflecting the frequency and type of traffic (no trucks on Sunday)

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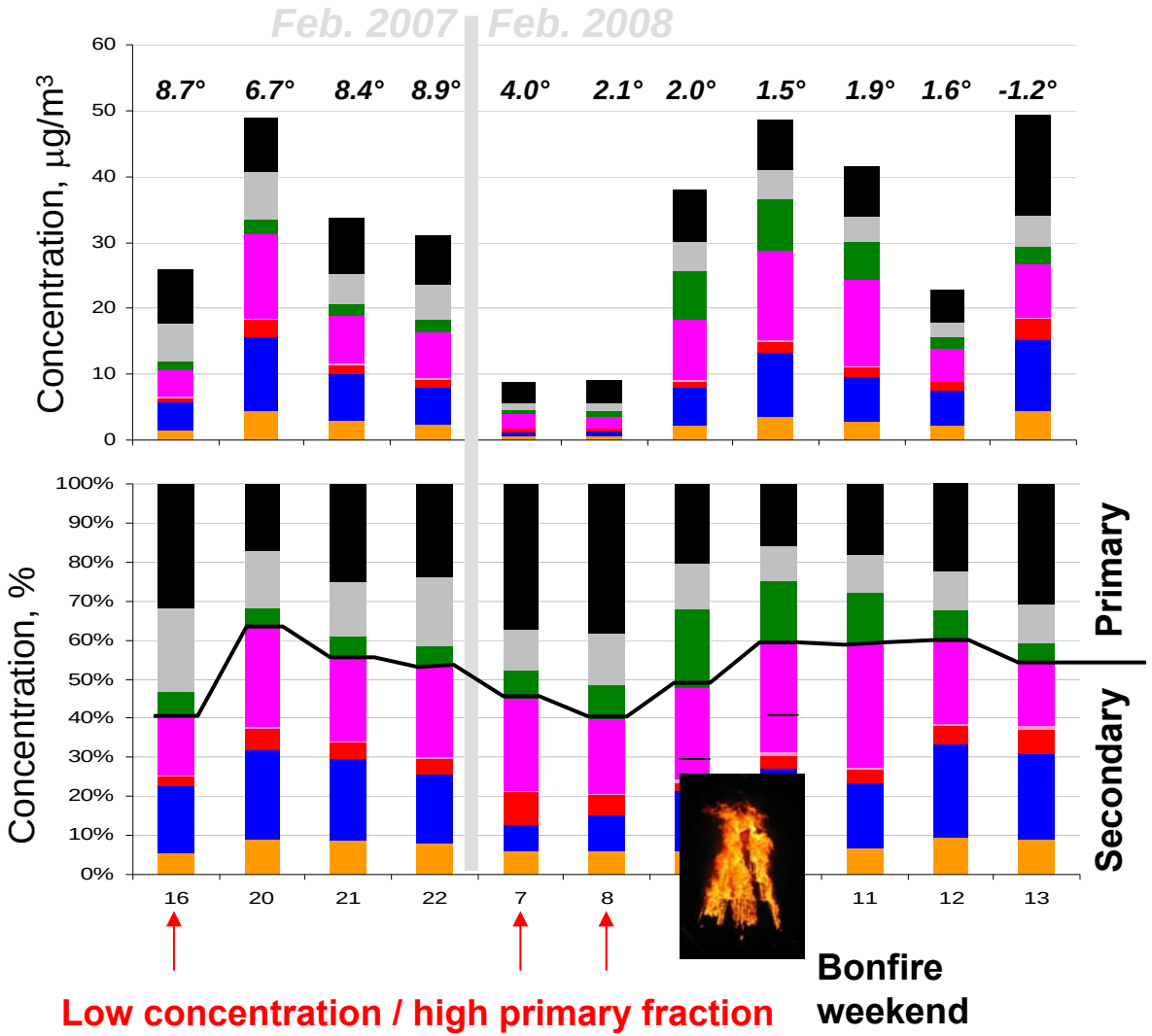
Composition and sources in the Rhinevalley on different days

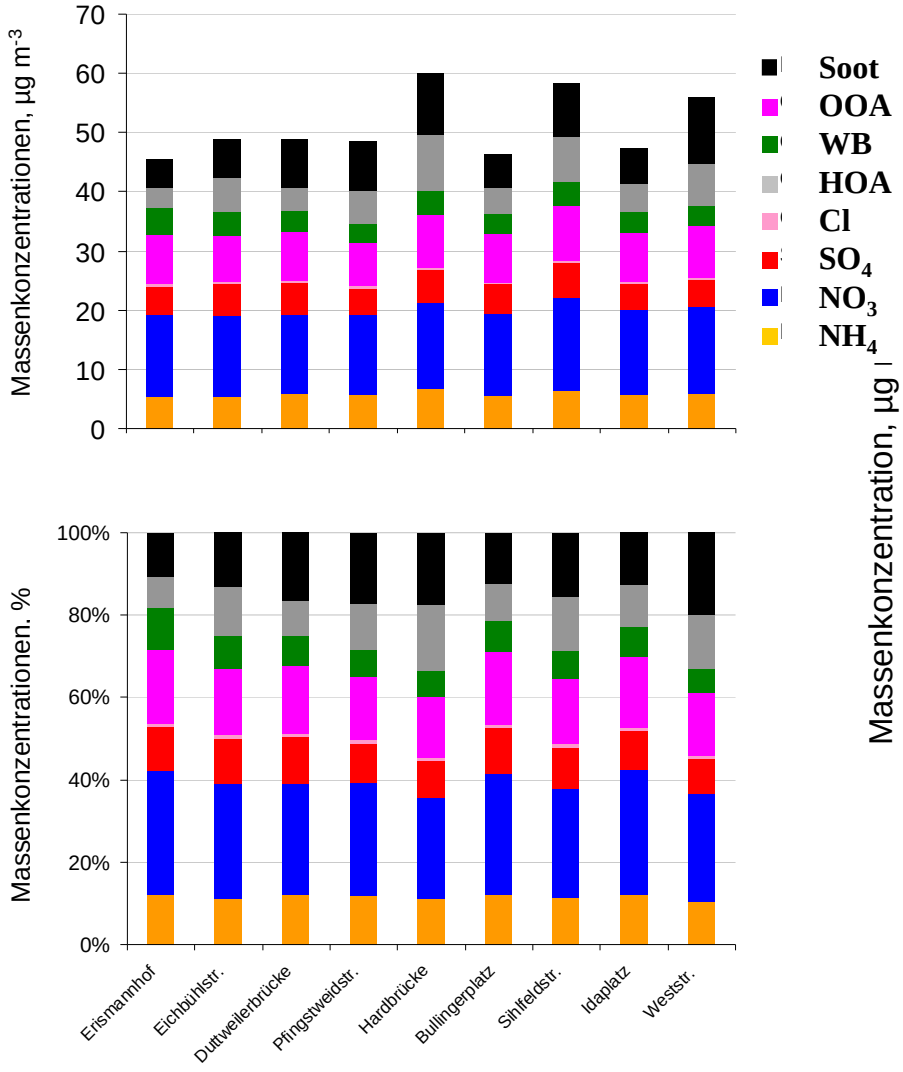


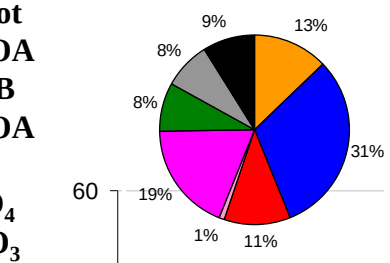
OOA: mostly secondary organic aerosol

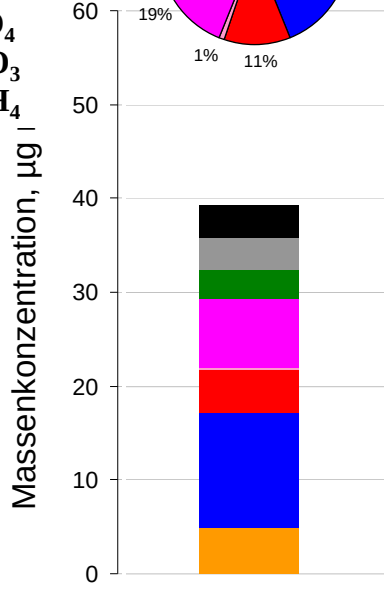
HOA: primary aerosol (mostly fossil)

WB: Wood burning

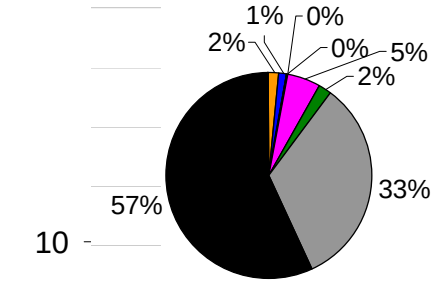
Variability of contribution at different locations and urban background versus local contribution

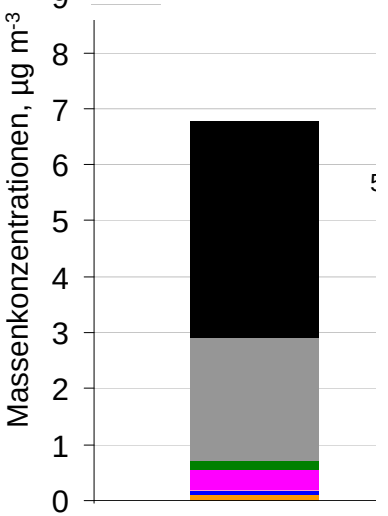






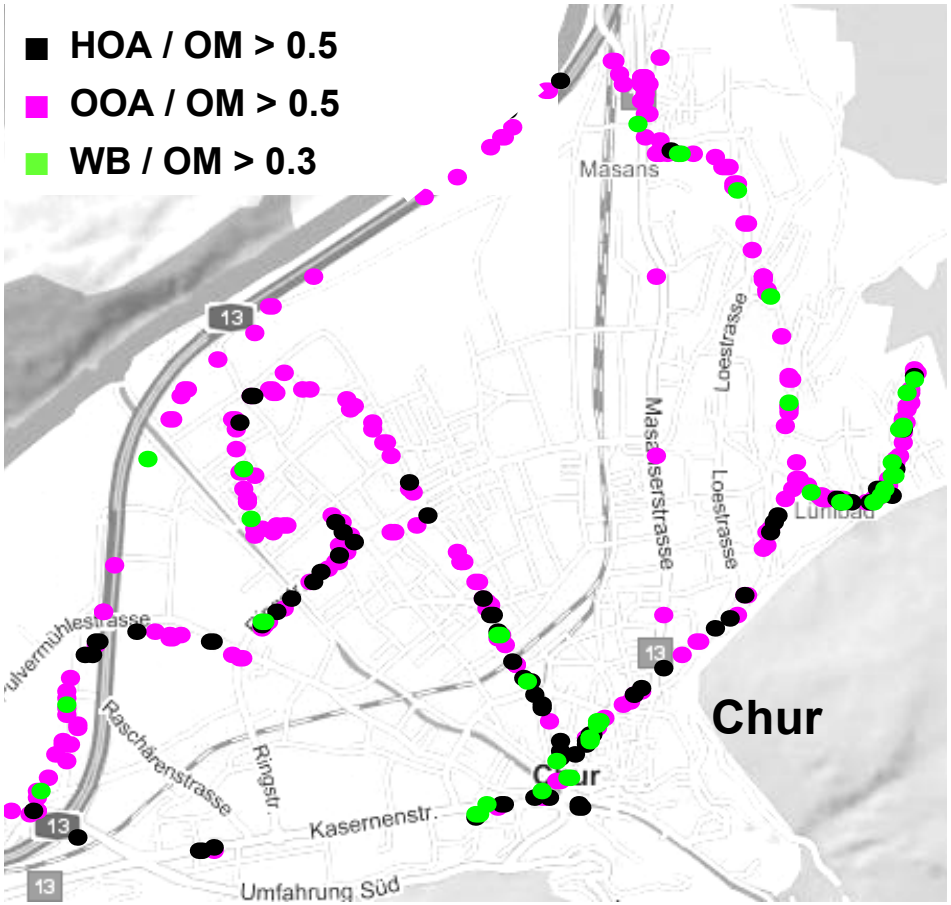
Urban background





Local contribution

Examplary analysis: Locations of high contributions of traffic, wood burning or OOA to organic mass in Chur

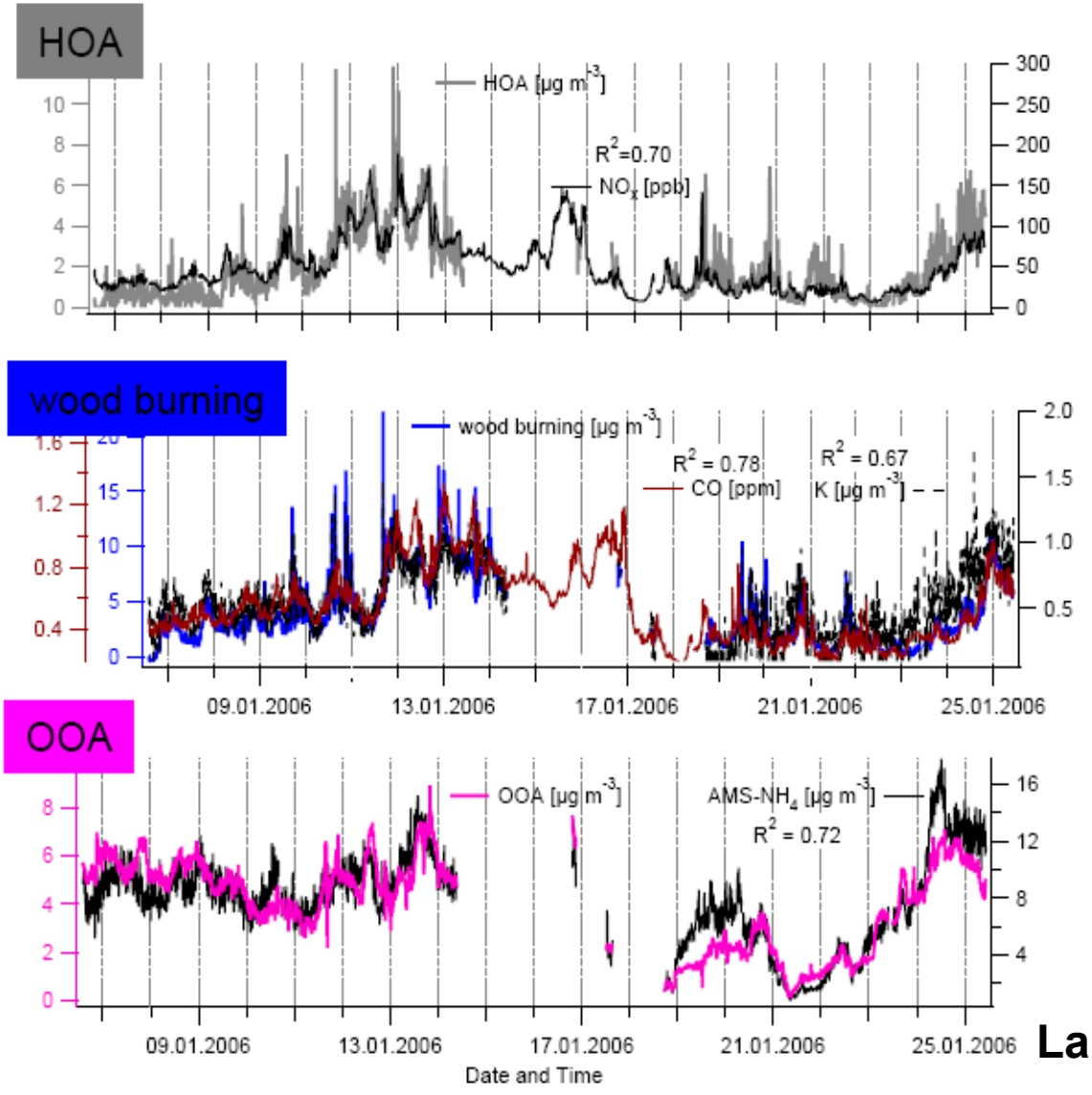


OOA: mostly secondary organic aerosol

HOA: primary aerosol (mostly fossil)

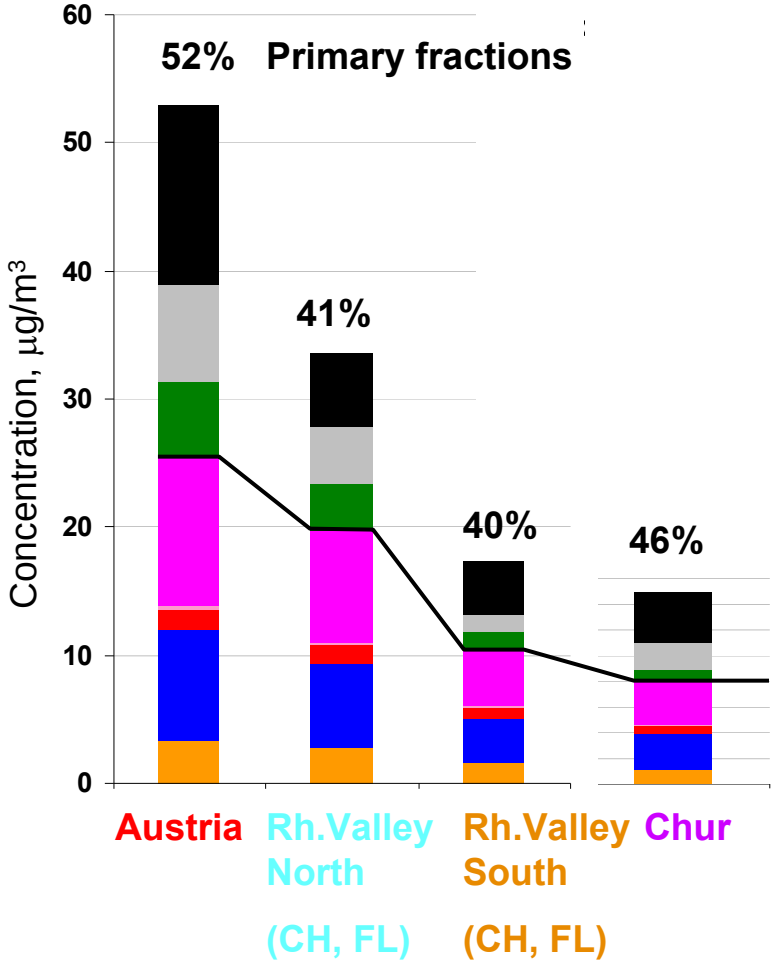
WB: Wood burning

Verification by comparing source strengths with tracers



Lanz et al., ES&T, 200

Composition in different regions in the Rhine valley

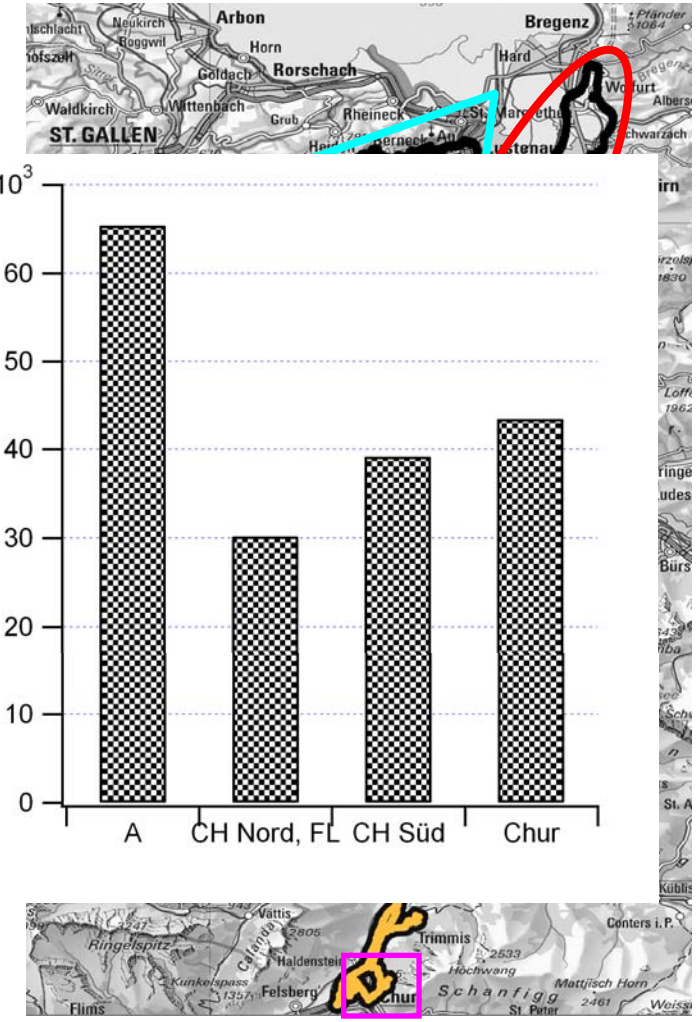


Soot
OOA
WB
HOA
Cl
SO_4
NO_3
NH_4

OOA: mostly secondary org aerosol

HOA: primary (mostly fossil)

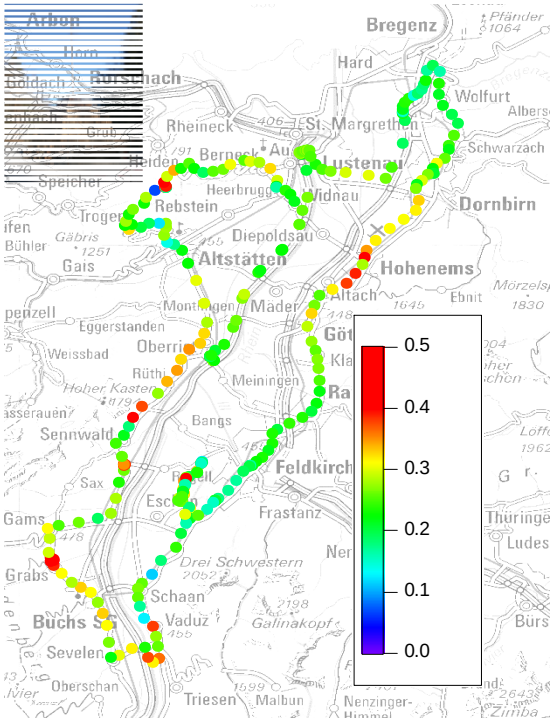
WB: Wood burning



Biomass burning contribution before and during the bonfires

Saturday morning

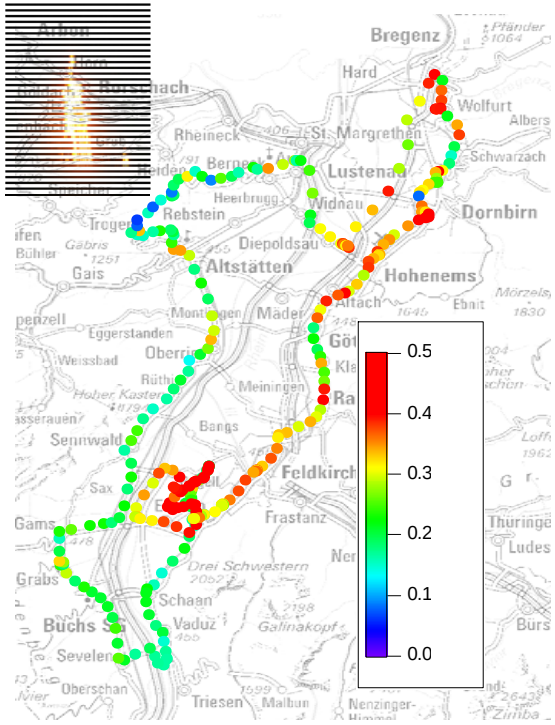
Before most bonfires



$OM_{wood\ burning}/OM$

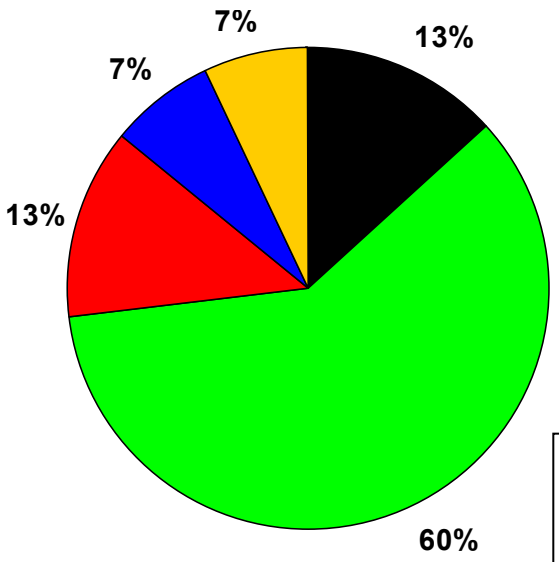
Sunday evening

During the bonfires



$OM_{wood\ burning}/OM$

Chemical composition in Zürich summer and Positive Matrix Factorization of the organic matter



Zürich (July)

Black Carbon
Organic mass
Nitrate
Sulfate
Ammonium

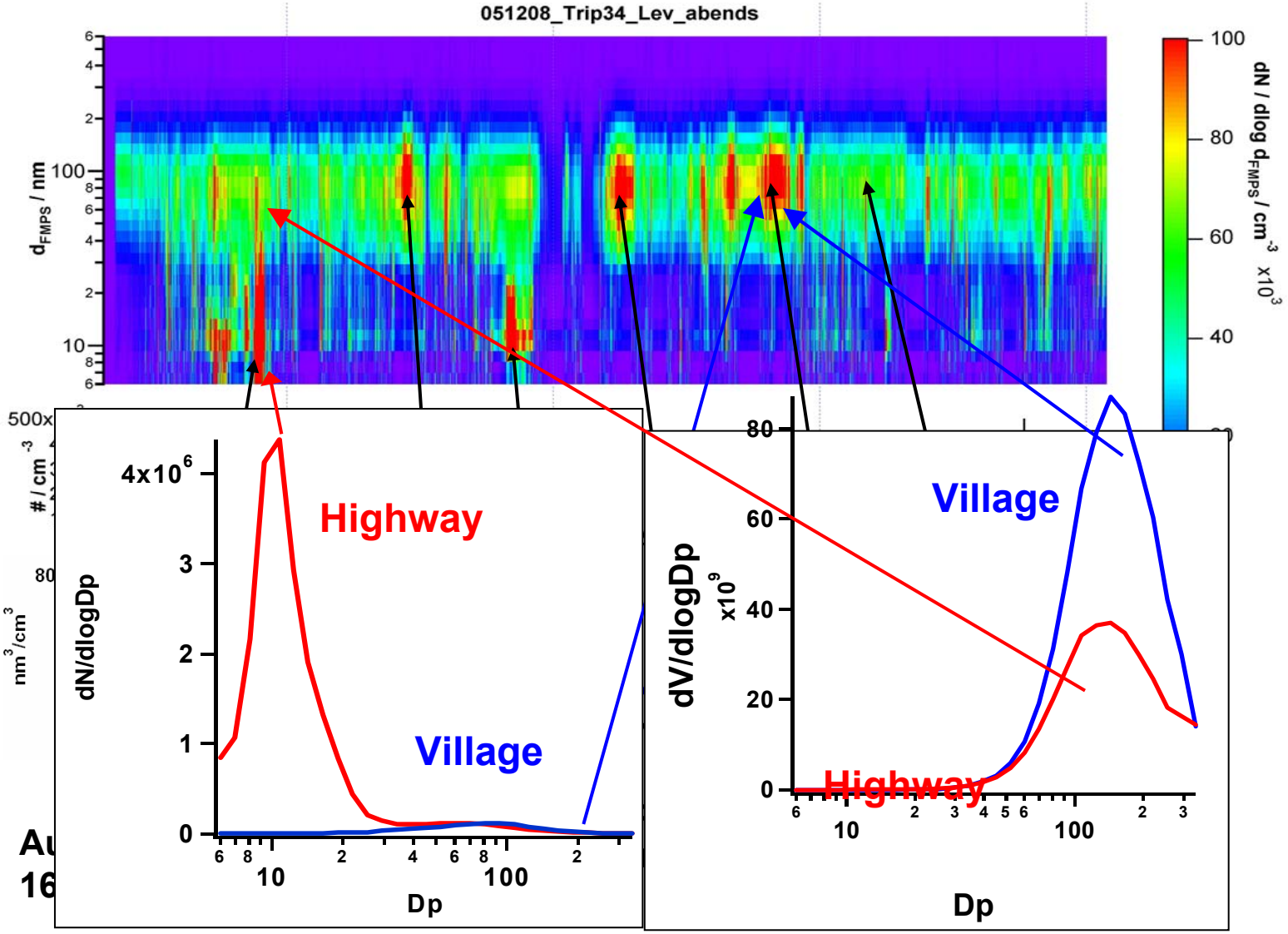
Organic mass

OOA: Secondary organic aerosol

HOA: mostly traffic

Lanz et al., ACP (2007)

Leventina 8. December 2005



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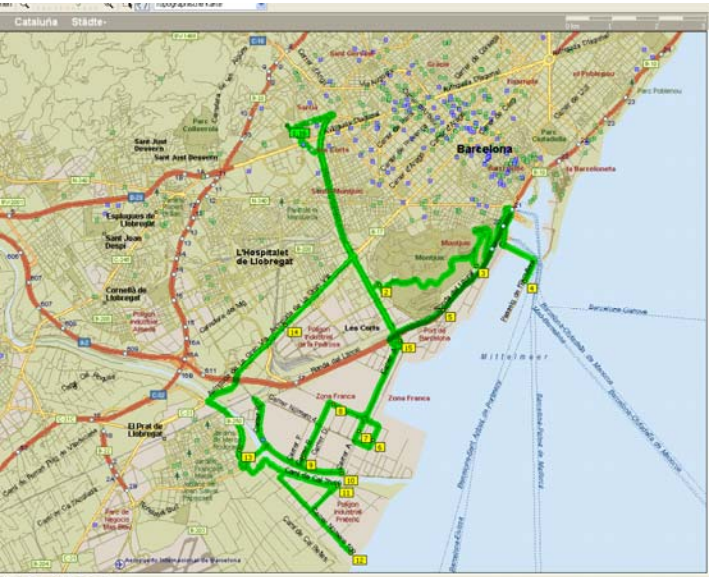
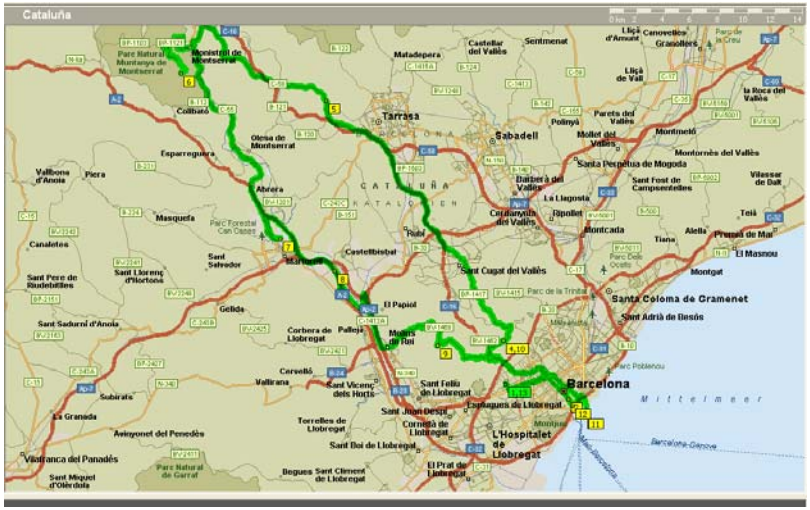
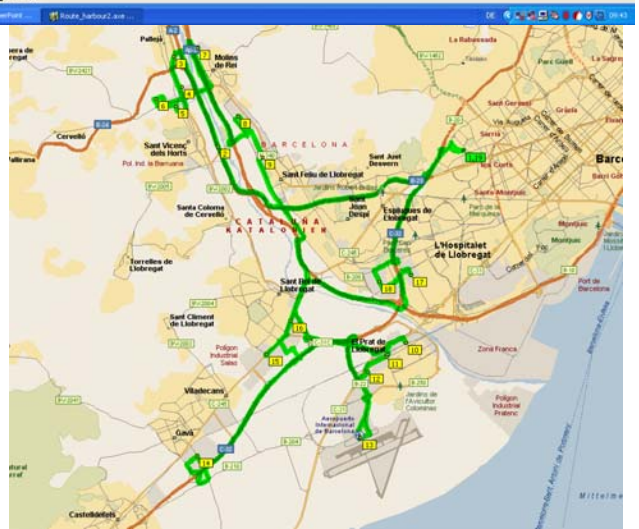
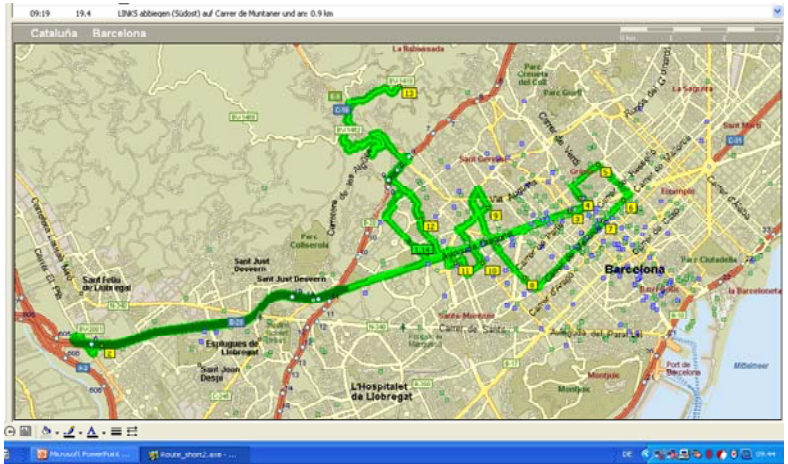
Examples of routes
Harbour, local Eixample, Diagonal; Transect Barcelona to Montserrat, industry chasing

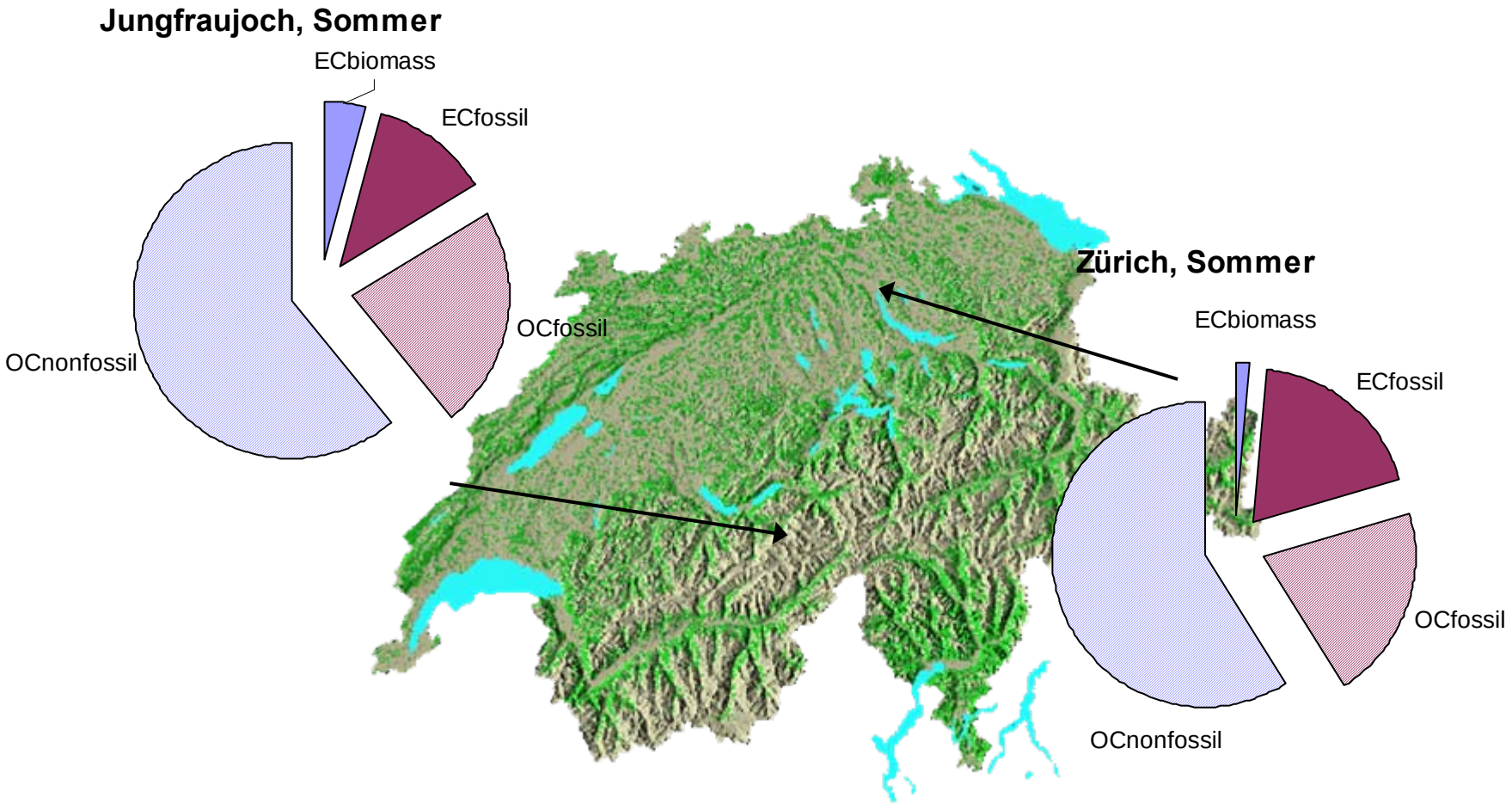




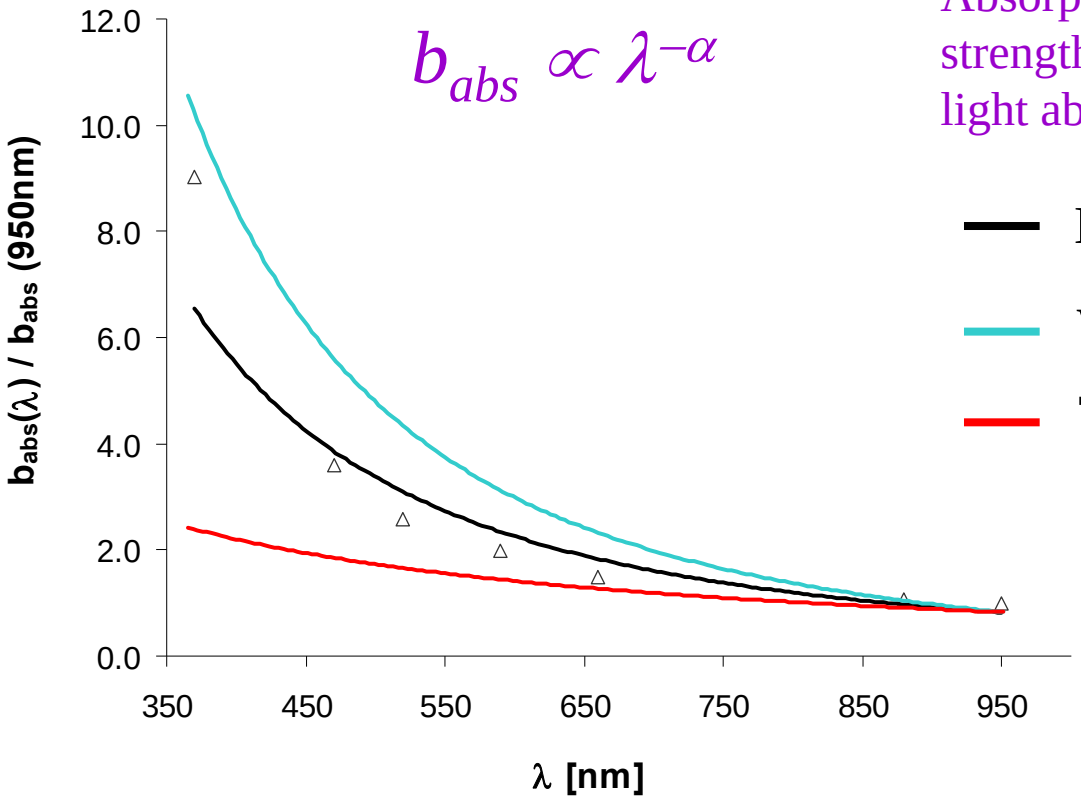




Overview over the summer ^{14}C analyses



Dependence of light absorption as a function of wavelength measured by an aethalometer



Absorption exponent α: a measure of the strength of the spectral variation in aerosol light absorption

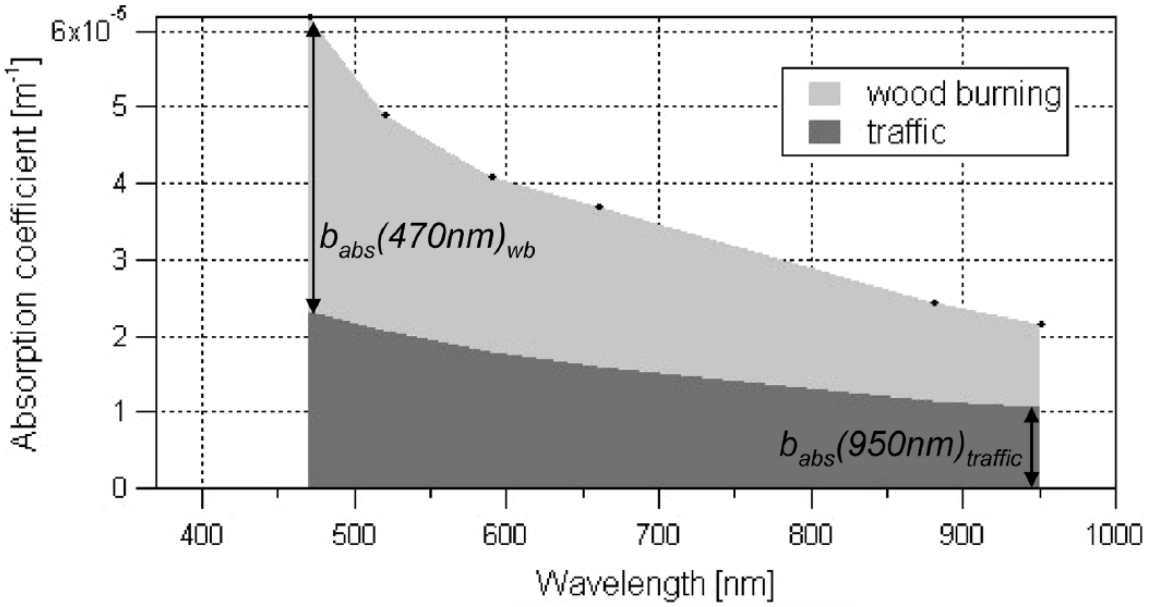
- Example power law fit ($\lambda^{-2.0}$)
- Wood burning[a] $\lambda^{-1.8 \text{ to } -2.2}$
- Traffic, diesel soot[a,b] $\lambda^{-1.0 \text{ to } -1.1}$

[a] Kirchstetter *et al.* 2004

[b] Schnaiter et al. 2003 & 2005

Sandradewi et al., Atmos. Environ., 2008

Aethalometer model to derive the contribution of woodburning and traffic to PM1

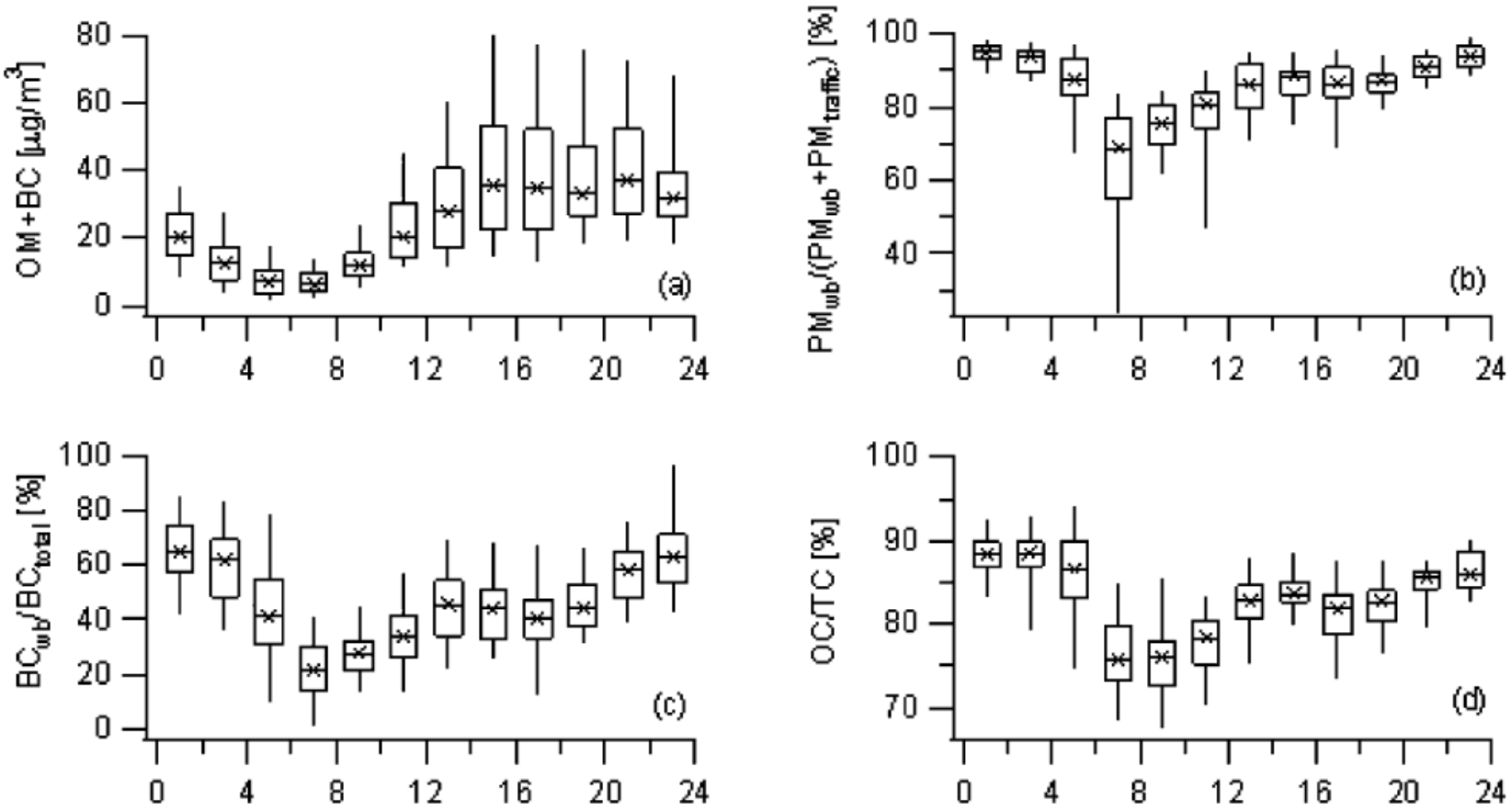


Carbonaceous material (CM)

$$\text{CM } (PM1) = \underbrace{c1 * b_{abs}(950nm)_{traffic}}_{PM_{traffic}} + \underbrace{c2 * b_{abs}(470nm)_{wb}}_{PM_{wb}}$$

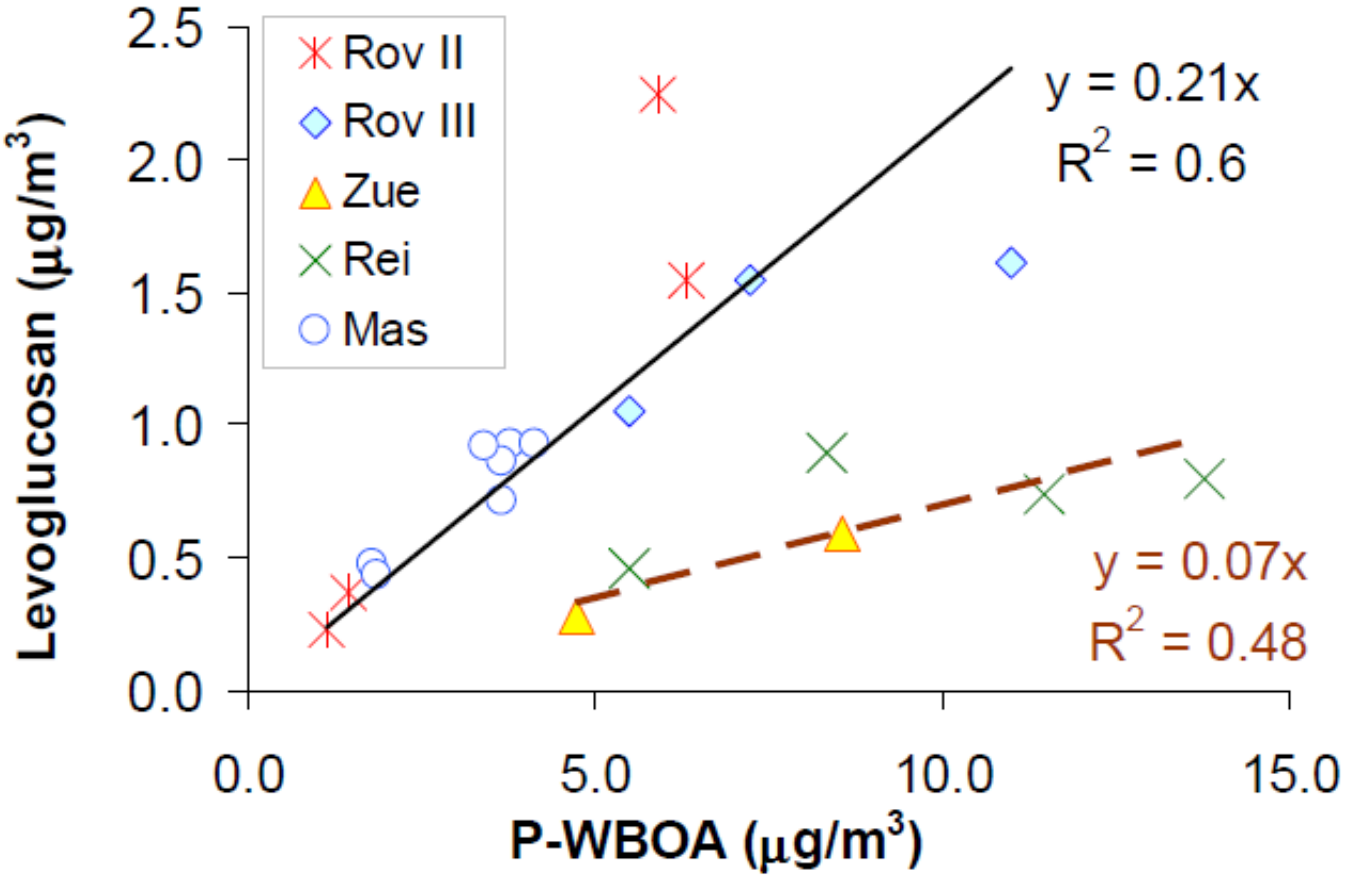
Sandradewi et al., ES&T, 2008

Average diurnal cycle of the carbonaceous material, OC/EC and wood burning versus traffic contributions



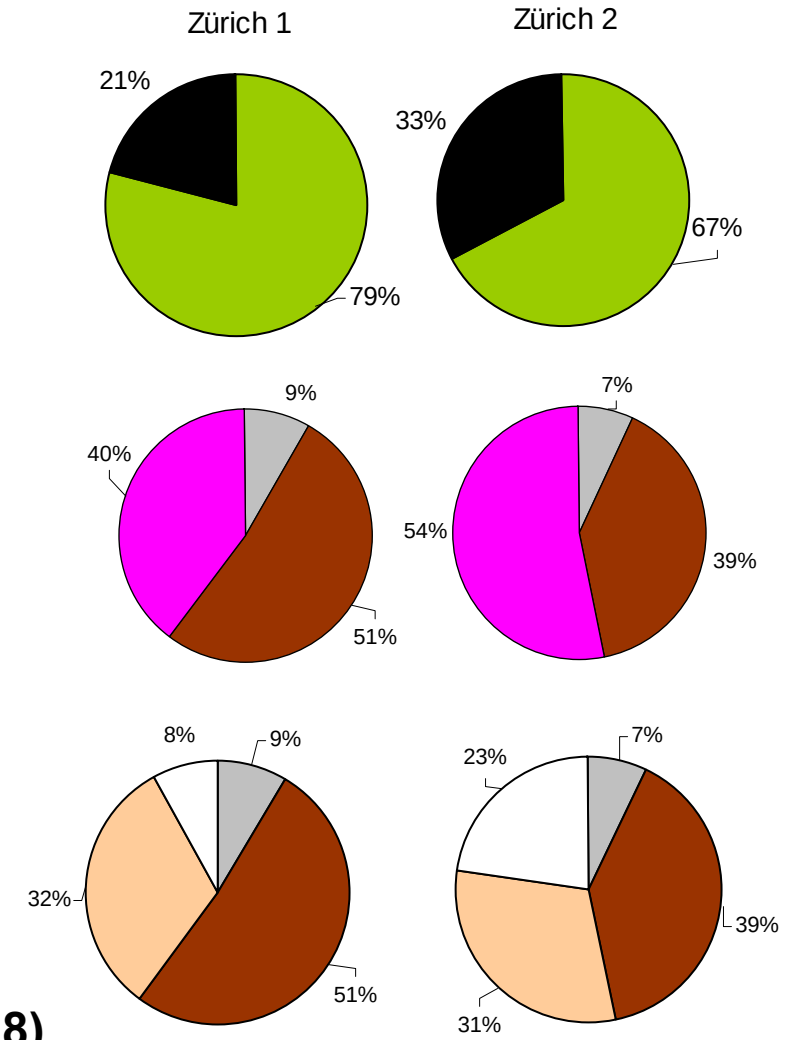
Sandradewi et al., ES&T, 2008

Levoglucosan is probably not very stable in some atmospheric conditions



Smogchamber studies in the US indicate that levoglucosan has a life time of less than a day up to days.. (Hennigan et al., GRL; 2010)

Combination of AMS and ^{14}C data for organics in Zürich for 2 time periods



Assumptions

OM/OC (wood burning) = 2

OM/OC (SOA) = 2

OM/OC (HOA) = 1.2

SOA non-fossil is 50% from wood burning and 50% from biogenic emissions

Lanz et al., ES&T (2008)