Universiteit Utrecht

Aerosol source apportionment from long term measurements at the CESAR tower Cabauw, NL

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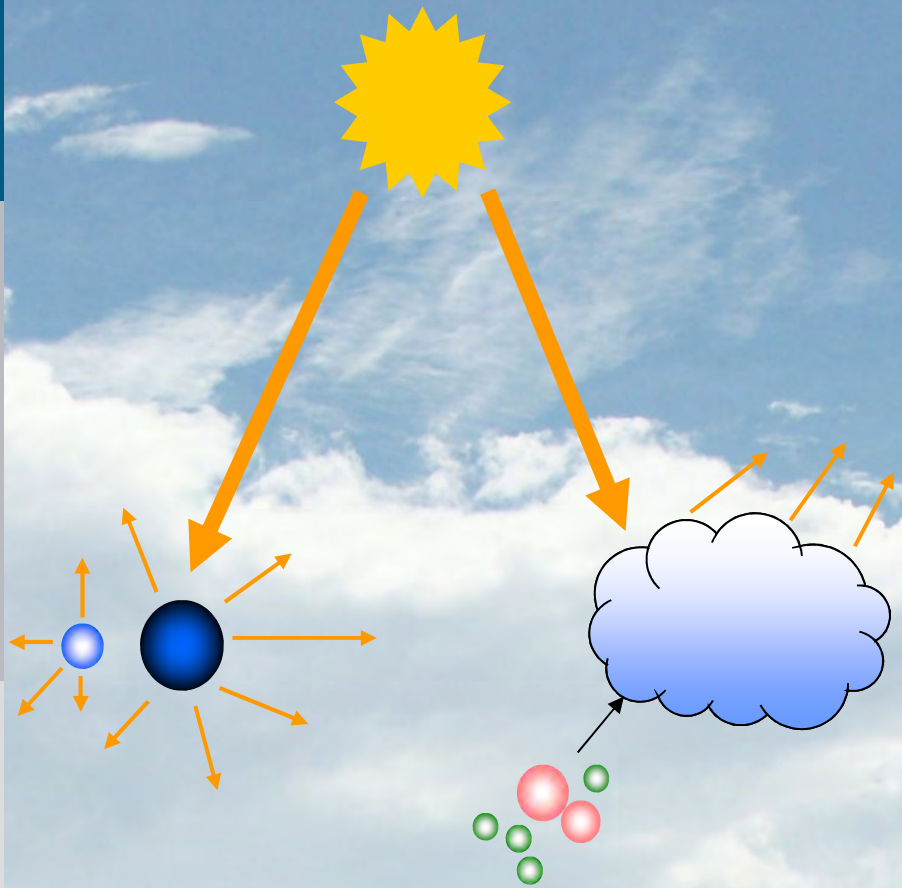
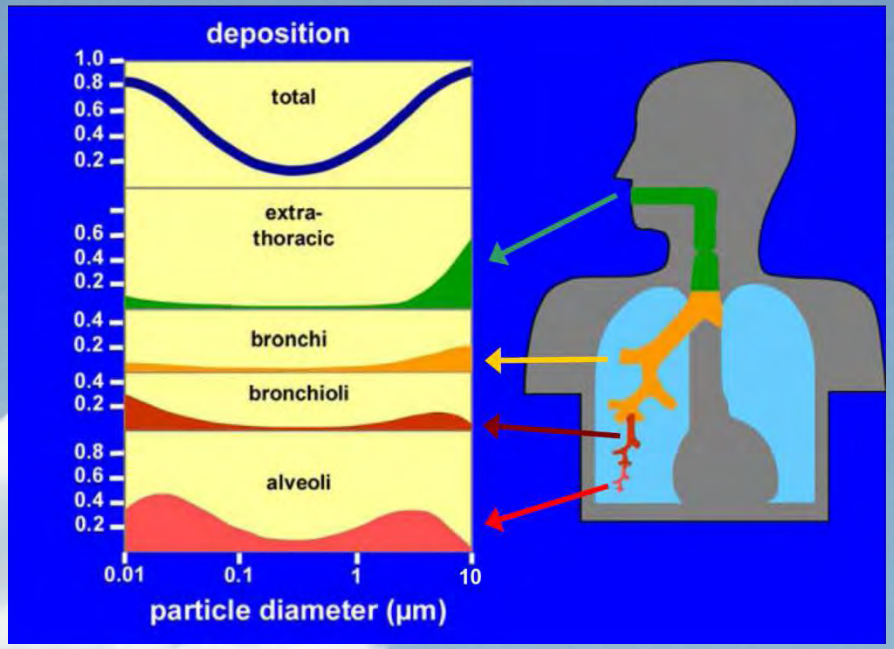
26. August 2015

Effects of Atmospheric Aerosols

JÜLICH
FORSCHUNGSZENTRUM



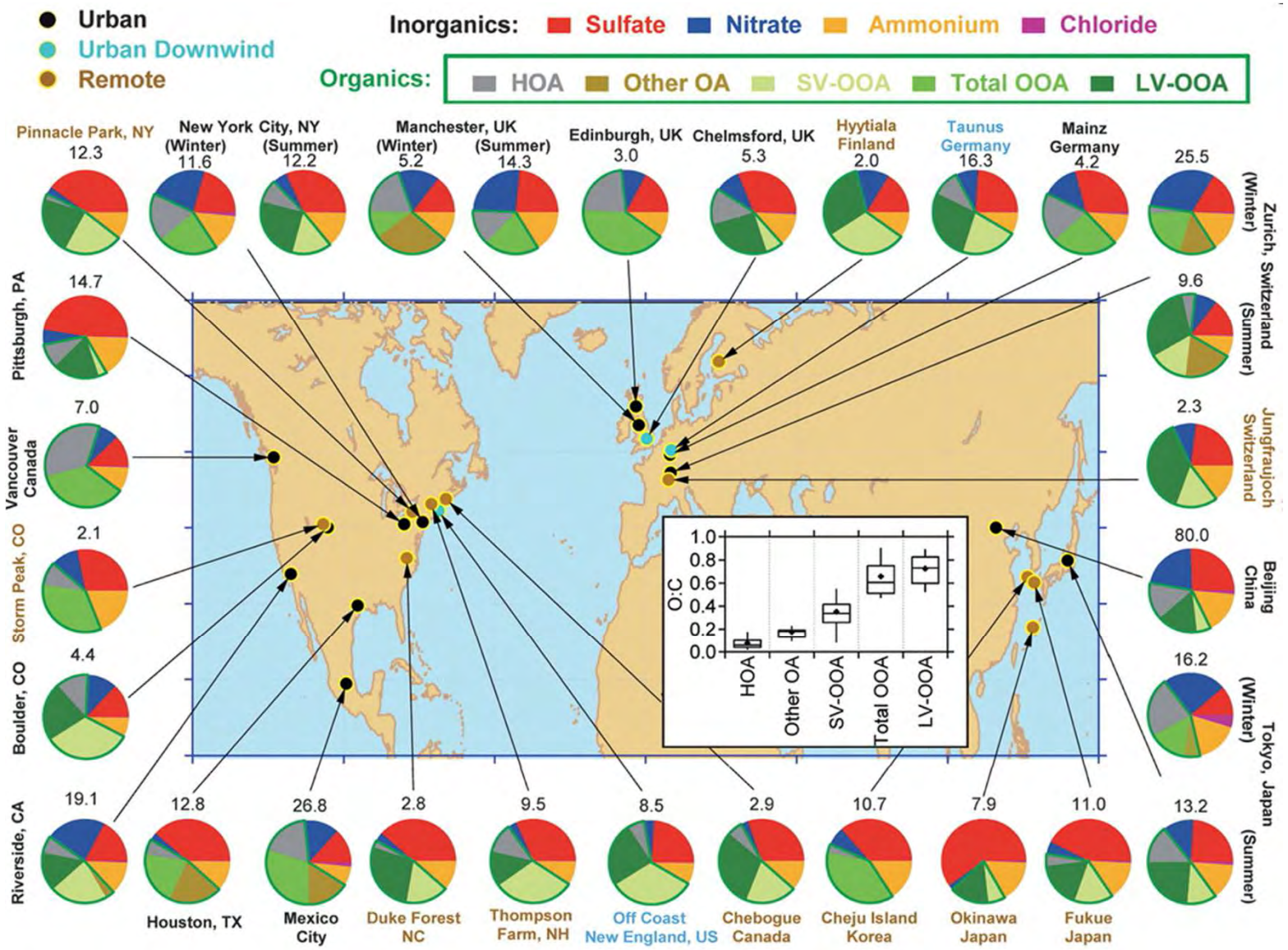


Direct effect on climate

Indirect effect on climate

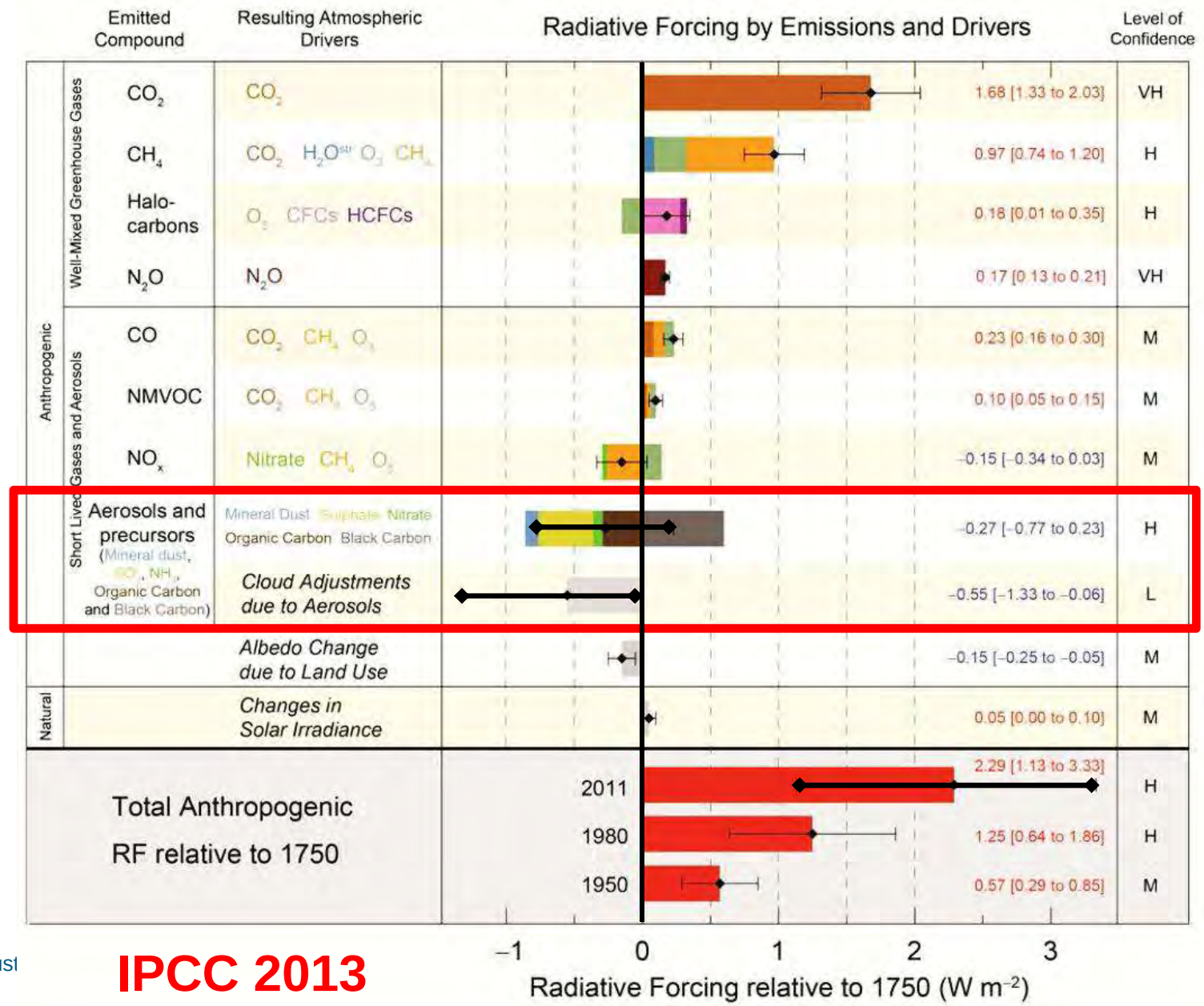
Adverse health effects

Atmospheric Aerosols: Composition



Jimenez et al., Science 2009

Effects of Atmospheric Aerosols



IPCC 2013

ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network)

- 17 ACSM (Aerosol Chemical Speciation Monitor) instruments measuring ambient aerosol at monitoring sites across Europe.
- Many since summer 2012.
- 3-week intercom-parison in Nov. 2013:
  - 12 Q-ACSM
  - 1 ToF-ACSM
  - 1 HR-ToF-AMS
- Crenn et al., 2015
- Fröhlich et al., 2015

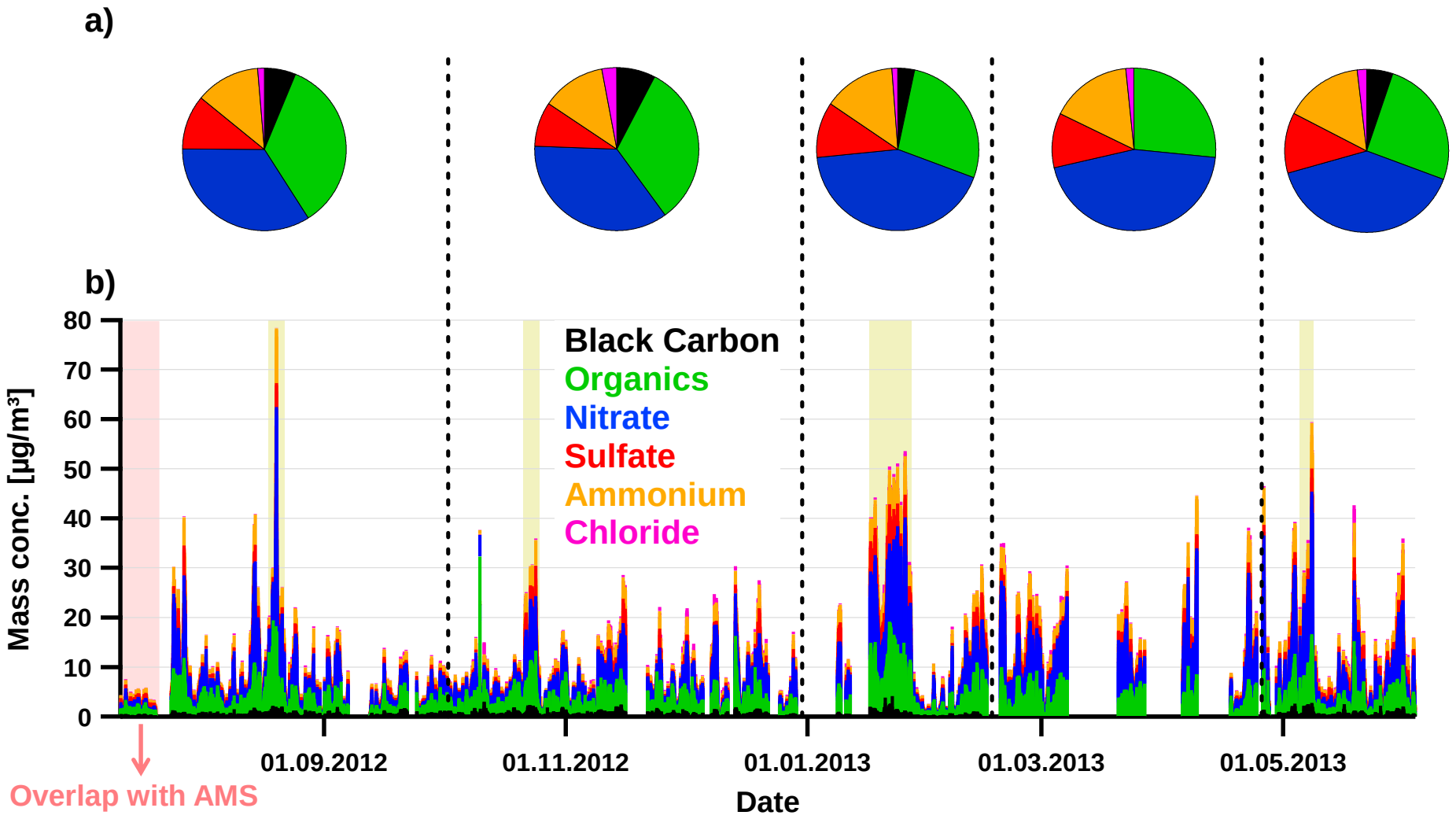
Cabauw Experimental Site for Atmospheric Research (CESAR)

Aerosol instruments used in this work

- Aerosol Chemical Speciation Monitor (ACSM)
  - PM_1 **Organics**, **Nitrate**, **Sulfate**, **Ammonium**, **Chloride**
- Multi-Angle Absorption Photometer (MAAP)
  - PM_1 **Black Carbon**

- High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS)
  - PM_1 **Organics**, **Nitrate**, **Sulfate**, **Ammonium**, **Chloride**
- Monitor for AeRosol and GAses (MARGA)
  - $PM_{1/2.5}$ water soluble like **Nitrate**, **Sulfate**, **Ammonium**, **Chloride**
- Scanning Mobility Particle Sizer (SMPS)
  - PM_1 total aerosol mass concentration

Overview ACSM/MAAP PM_1



Fractional species contribution to PM_1 mass (a) and time series of species mass concentration (b); Total average mass loading: 9.5 µg/m³

Instrument Comparison (2012 – 2013)

Correlation slopes (Pearson-R^2)			
	MARGA	**HR-ToF-AMS**	
ACSM vs.	---	1.00 (R^2=0.73)	**Organics**
	1.23 (R^2=0.96)	1.17 (R^2=0.89)	**Nitrate**
	0.88 (R^2=0.93)	0.82 (R^2=0.71)	**Ammonium**
	0.63 (R^2=0.86)	0.49 (R^2=0.76)	**Sulfate**
	1.05 (R^2=0.93)	0.90 (R^2=0.84)	**Total**
	(1943 data points)	**(289 data points)**	**(No. of points)**

ACSM + MAAP total mass vs. SMPS total mass (12275 data points):

Slope: 0.84 (R^2 = 0.82)

Good correlations were seen in general over the whole campaign as well as during periods with high mass loadings!

- **ACSM** slightly overestimated **nitrate** by 23% and 17% and underestimated **sulphate** by 37% and 51% comparing to MARGA and AMS, respectively. ACSM **chloride** was largely below the detection limit.

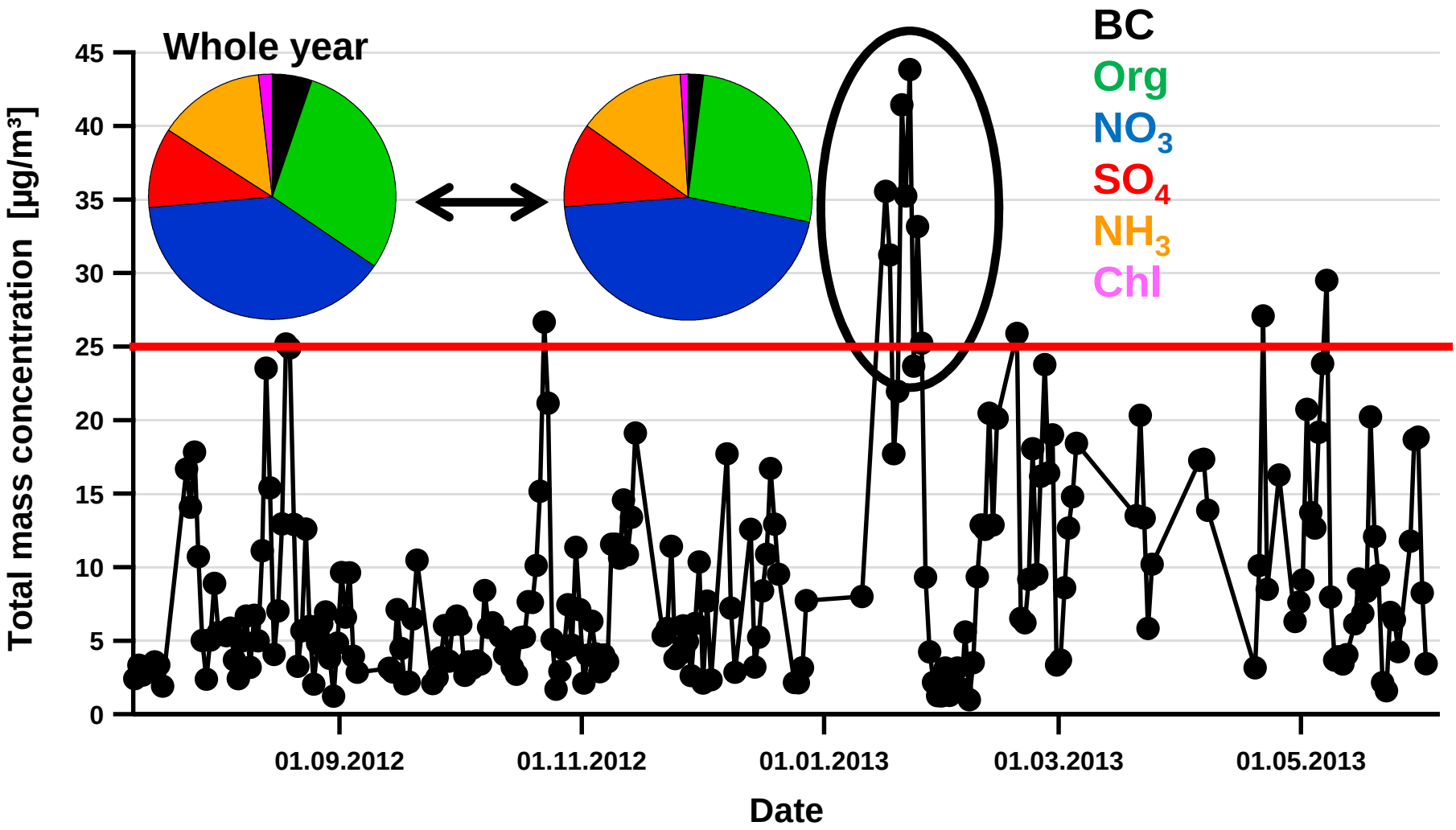
Air quality standards of the World health organization (WHO)

	24h-mean	Annual mean
$PM_{2.5}$	25 µg/m³	10 µg/m³

➢ **Keep in mind that we have PM_1 composition measurements**

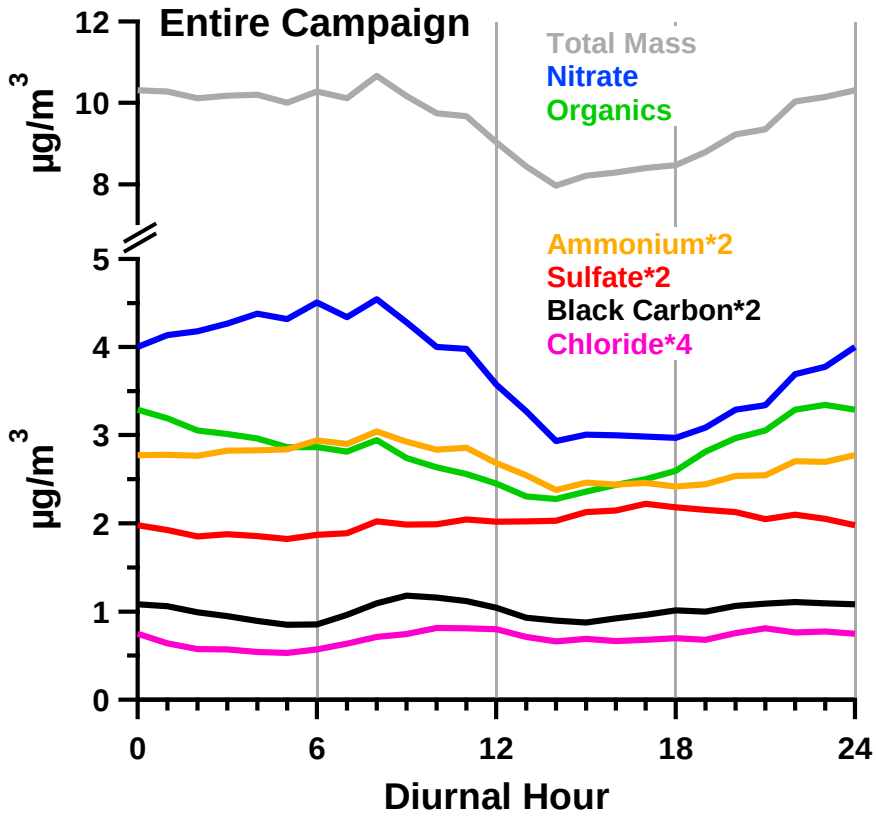
ACSM campaign: 9.5 µg/m³

Total ACSM+MAAP PM_1 mass: Daily means



➢ **12 exceedances of WHO $PM_{2.5}$ daily mean limit**

ACSM/MAAP: Diurnal Variations

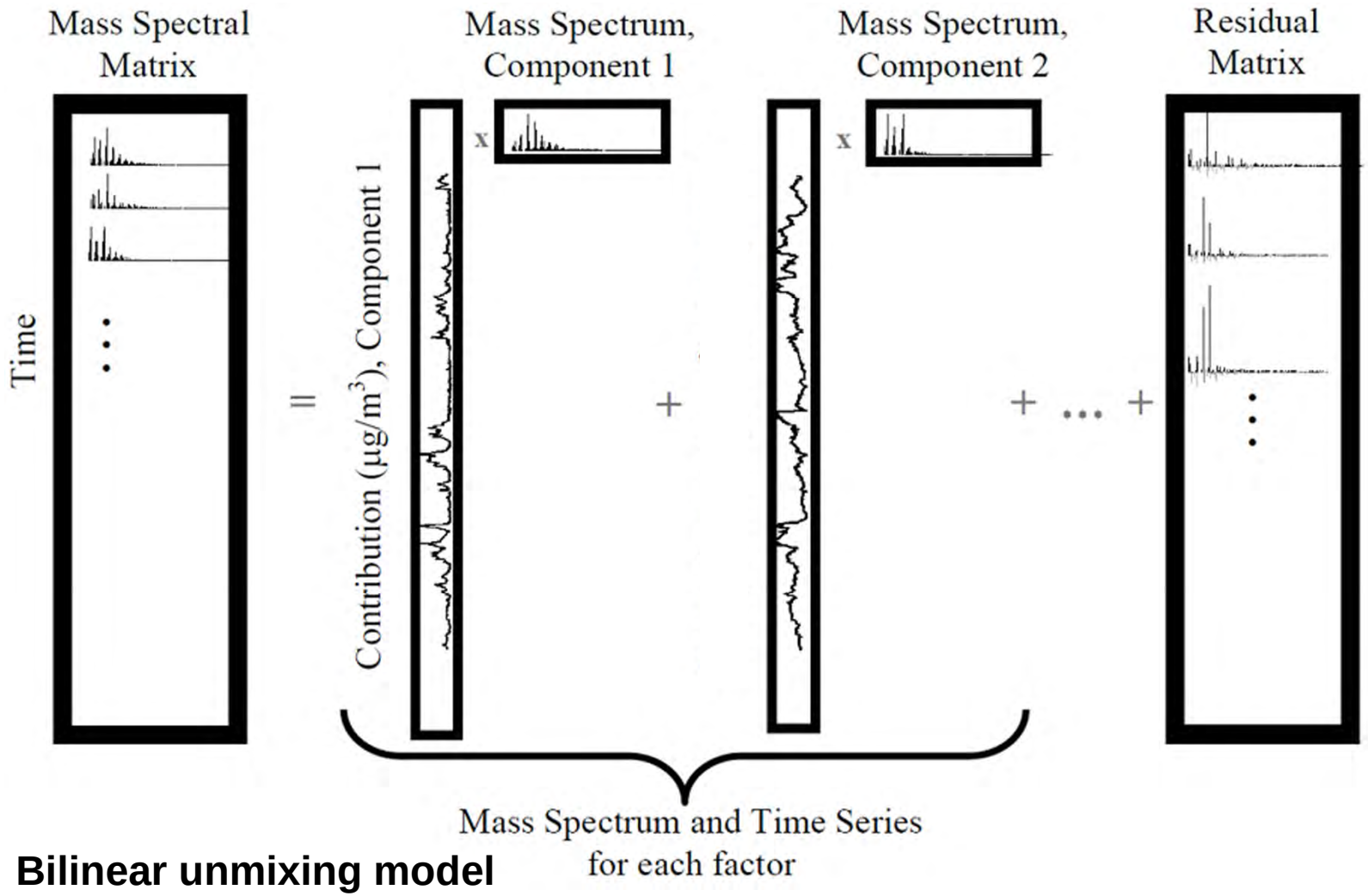


Primary aerosol

Secondary aerosols

- NO_3: heterogeneous conversion from NO_y in the night / volatilization of semi-volatile nitrates and photolysis of gaseous precursors during the day
- SO_4: Daytime maximum due to it's photochemical formation from SO_2
- NH_4: Neutralization of NO_3 and SO_4 and by NH_3 into NH_4NO_3 and $(NH_4)_2SO_4$
- BC: Direct emissions from traffic (morning and evening rush hours) and biomass burning events (domestic heating in the evenings/nights)

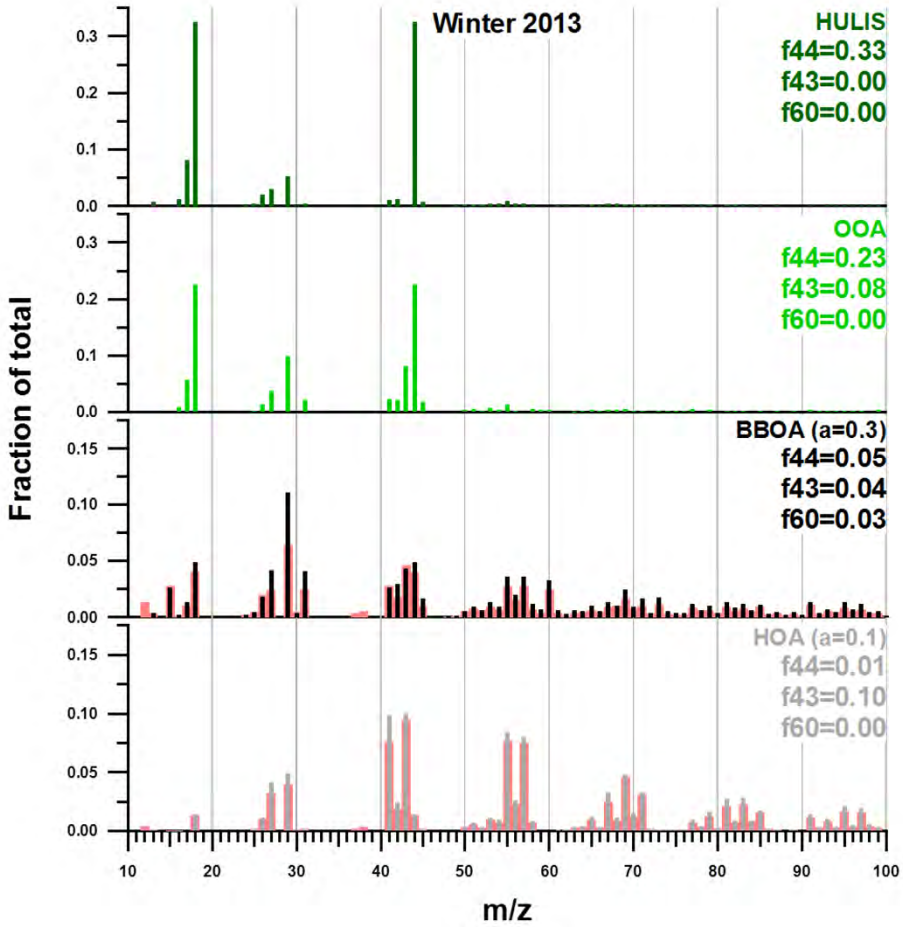
Positive Matrix Factorization (PMF)

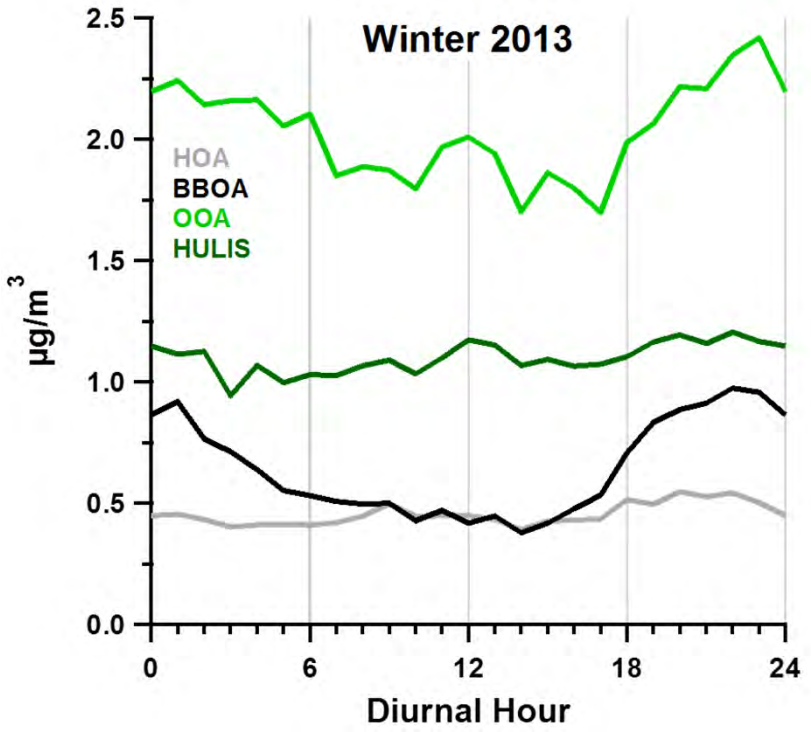


- **Bilinear unmixing model**
- **Least squares algorithm**
- **No a priori information, except: values are non-negative**
- **Factors represent sources (primary OA; POA) / aging (secondary OA; SOA)**

Ulbrich et al., ACP 2009

Multilinear Engine 2 results from ACSM OA

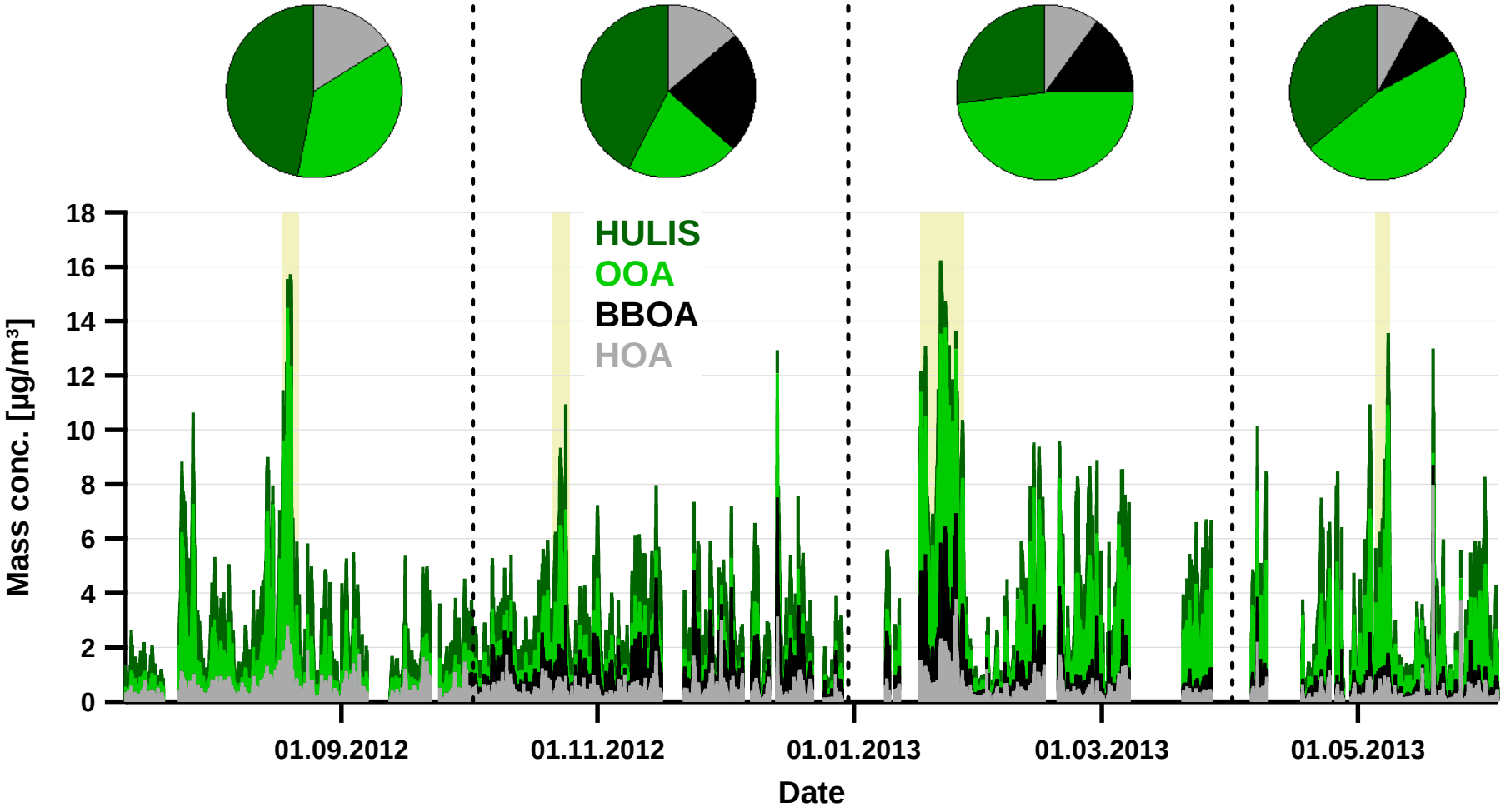




- **These factors were seen in most seasons investigated by AMS and ACSM**
- **Red bars: site specific reference mass spectra from Crippa et al., ACP 2014, used as constraints for ME-2 (a-value in brackets)**
- **HOA showed diurnal pattern from traffic rush hours, while BBOA was mainly emitted by domestic heating in the evening/night. Highly oxidized HULIS (attributed to humic like substances) showed no diurnal variation**

HULIS: Paglione et al., ACP 2014

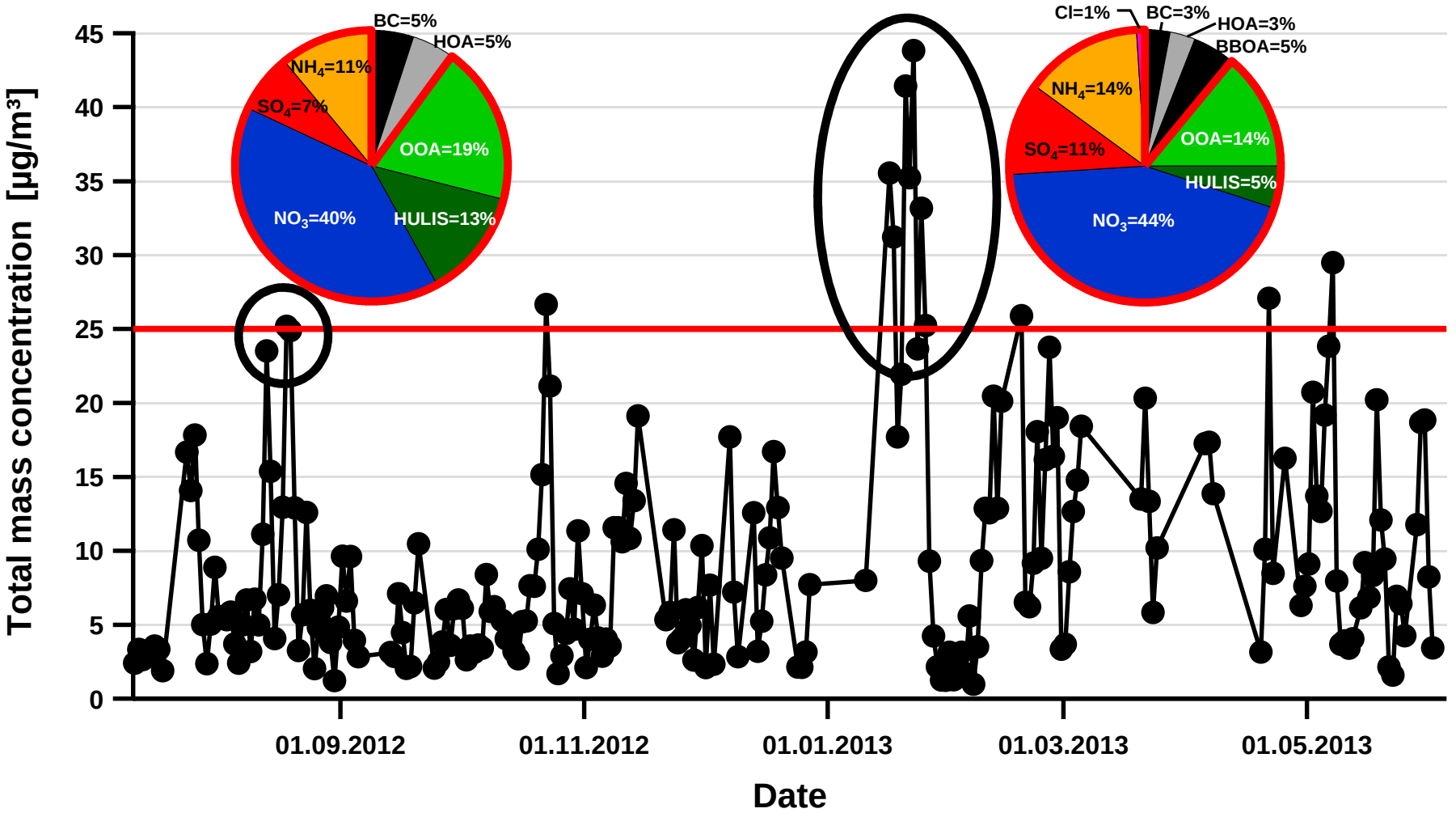
PMF/ME-2 results from ACSM PM_1 data



- **No biomass burning was found in summer. High contributions of chemically formed SOA (OOA and HULIS) were found where the HULIS concentration remained largely constant during the campaign, emphasizing its role as background aerosol at Cabauw**

HULIS: Paglione et al., ACP 2014

Sources of high particulate mass



- Pie charts show fractional abundances averaged over two selected high mass periods.
- Wedges highlighted in red represent secondary aerosol fractions.

Summary

- **Performed 1-year PM_1 aerosol chemical composition measurements in Cabauw:**

  12 exceedances of WHO $PM_{2.5}$ daily mean limit

- **Nitrate and organics are the most dominant species**
- **Nitrate (mainly as NH_4NO_3) was the main contributor in periods with high mass loadings**

  → Reducing NO_x and/or NH_3 emissions should have a large effect on reducing particulate matter

- **PMF/ME-2 analysis of the organic fraction show high contributions of chemically formed SOA**
  - Typical for rural sites
  - The local reduction of organic aerosol mass is more challenging, especially as the HULIS fraction showed no designated source.

Acknowledgement

- FZ Jülich
  Astrid Kiendler-Scharr (IEK-8)
- Utrecht University, NL:
  Rupert Holzinger, Josef Timkovsky
- KNMI, NL:
  Marcel Brinkenberg, Jacques Warmer, Cor van Oort, Fred Bosveld, Gert-Jan van Zadelhoff, Reinout Boers
- TNO, NL:
  Bas Henzing, Marcel Moerman
- ECN, NL:
  Alex Vermeulen, Mark Blom
- **Funded by ACTRIS TNA**