Proceedings of 12th ETH-Conference on Combustion Generated Nanoparticles

Trace elements in wood combustion performed with different domestic heating stoves

Agnes Richard, Maarten Heringa, Michael Sattler, Nickolas Karl Meyer, Markus Furger, André Prévôt, Christian Gaegauf

Introduction

Wood burning as a source of renewable energy is of increasing interest in Switzerland, where a large amount of wood heaters and boilers are in use. Therefore wood burning emissions are a significant source of air pollution especially in alpine valleys in periods with poor air mixing. For example, it contributed up to 85% of the organic aerosol mass measured in a winter campaign in Roveredo in 2005/2006. Besides high emissions in winter by domestic heating, wildfires and barbeques are common sources in summer. Consequently there is a high interest in chemical, elemental and quantitative analysis of wood smoke. A second question is, to what extent different burners, fuels and burning conditions influence the composition and temporal behavior of the emissions. To this purpose a collaboration of Ökozentrum Langenbruck, FH Windisch and PSI investigated four different wood stoves under varying burning conditions within a testing setup.

Experiment

Aerosol concentrations as well as chemical and elemental compositions were measured directly at the exhaust tube of different wood stoves for domestic heating.
The instruments (QAMS (quadrupole aerosol mass spectrometer), SMPS, FMPS, MAAP, TEOM, an APS, and a three-stage rotating drum impactor (RDI) were situated after two respectively three dilution stages, of which the first one was temperature controlled to avoid particle condensation. Results of the latter two instruments with time resolutions of the order of seconds to minutes were analyzed in this study and partly compared to the AMS measurements. RDIs collect aerosol samples in three different size ranges (PM10-2.5, PM2.5-1 and PM1-0.1) which are analyzed offline with synchrotron-based X-ray fluorescence spectrometry (XRF) at around 11 keV. The elements detected in the current setup at the new superXAS beamline at the Swiss Light Source (SLS, PSI) range from Al to Zn. This analysis method allows for detection of minuscule amounts (a few pg m^{-3}) of elements deposited on the sampling substrate (1.8-µm Mylar foil) without further treatment. However, the largest size fraction is not completely analyzed, as only the smaller particle size fractions occur typically in combustion processes.
Four different ovens were selected, one of which is fired automatically with pellets, while the others are traditional log ovens with beech log wood and fir wood to start the fire. Some additional burns were performed by adding bark to the pellets in the automatic burner. Therefore not only the size of the combustion chamber differed for the different types of ovens, but also the composition of the fuel. It was taken care of homogenous burning conditions, i.e. size, mass and positioning of the burned wood was kept constant as far as possible. Measurements included cold and warm starts, flaming and smoldering phases of the log wood burner, and also the cold start of the automatic burner. The concentrations of CO, CO_2 and O_2 were measured continuously to relate the particle emission to the oxygen concentration and to

control the combustion conditions. The dilution ratio was calculated through the ratio of CO_2 measurements in the stack and at the inlet of the instruments.

Results

Different ovens showed a slightly different elemental composition of the emitted wood smoke. For example, the burns performed with an automatic pellet stove revealed highest amounts in K and Ca, whereas other ovens also showed significant amounts of Si, Mn and Zn. Figure 1 shows the temporal behavior of different elements in the pellet burner measured with the RDI. One data point corresponds to a measurement time of 5 minutes. The last panel shows the concentrations measured with the APS. Interestingly, the concentrations of S, Cl, K, and Zn increase during the burns.

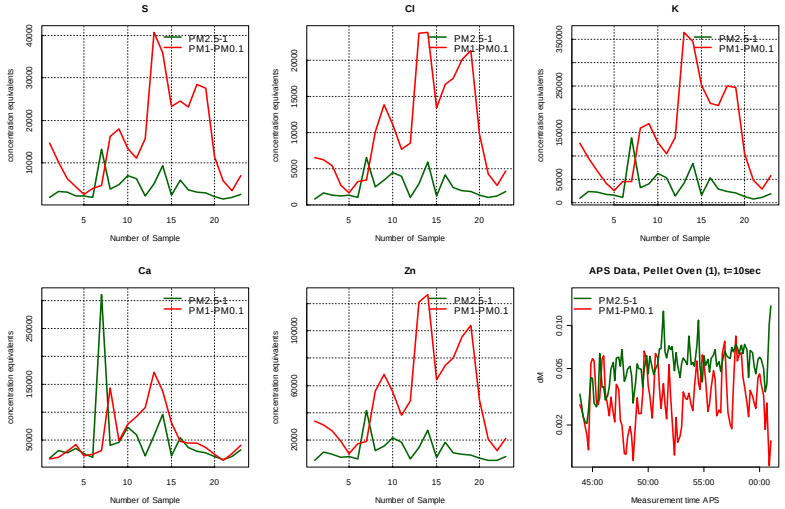

Figure 1: RDI (panels 1-5) and APS (panel 6) data from the automatic pellet burner, time resolution of 5 min (RDI) and 6 sec (APS), respectively.

As was seen in earlier wood burning studies, the main components of the submicron particles are potassium, sulfur and chlorine, while their ratios depend on the type of burner. In Figure 2 the correlations between the most abundant elements are shown, tagged elements are found frequently in the literature.

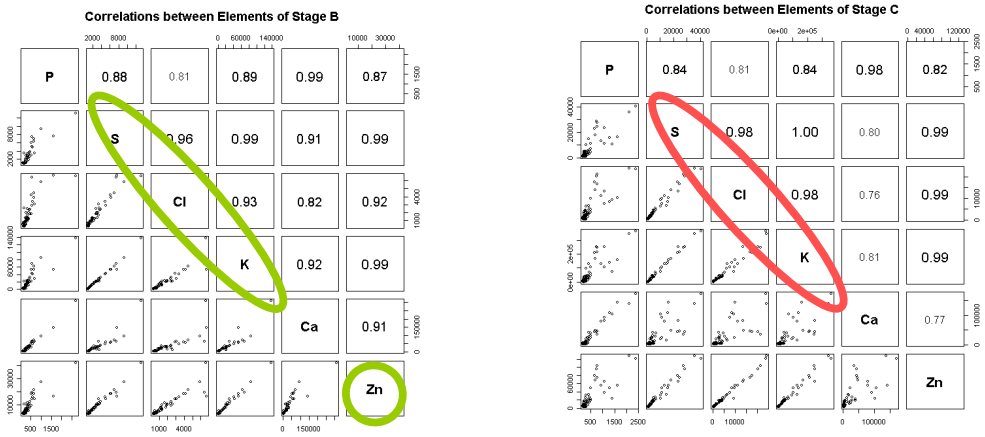

Figure 2: Correlations of most abundant elements, PM2.5-PM1 (left) and PM1-PM0.1 (right).

The AMS results displayed in Figure 3 show the different chemical composition in different burning phases, i.e. high concentrations of organics in the startup phase, and more salts during stable burning conditions. The measured m/z 60 and 73 are fragments of levoglucosan and similar organic compounds.

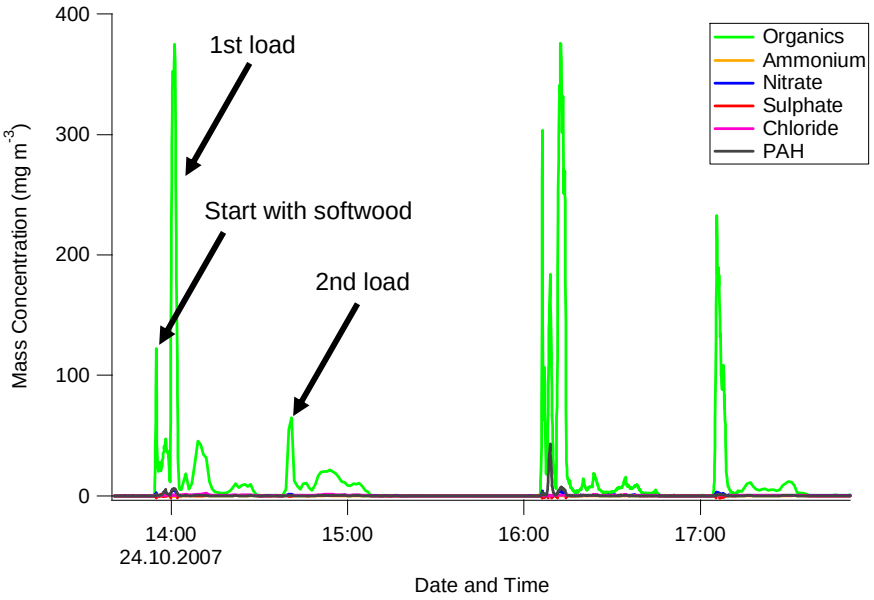

Figure 3: AMS data from log wood stove, two burns with two loadings each

Conclusions and Outlook

The time-resolved elemental analysis with RDIs gives information about the temporal variability of elements.

The AMS measurements show that the wood burning markers, m/z 60, 73 and 137, are mainly formed during the start of the fire. The high-resolution TOF-AMS data made it possible to discriminate peaks with the same nominal mass and to assign them to the correct species.

To consolidate these findings a follow-up campaign is planned for October 2008 in co-operation with the University of Applied Sciences of Lucerne and FHNW Windisch.

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Laboratory of Atmospheric Chemistry

Paul Scherrer Institut

Outline

- wood burning campaign 2007
- measurement techniques
  - aerodynamic particle sizer (APS)
  - rotating drum impactor (RDI)
  - aerodyne mass spectrometer (AMS)
- results
  - APS data
  - RDI data
  - AMS data
- conclusions

motivation

- wood burning is as a source of renewable energy of increasing interest
- wood burning contributed up to 85% of the organic aerosol mass in Roveredo[1]
- wildfires are common in summer

=> chemical, elemental and quantitative analysis of different burners + burning conditions

ökozentrum langenbruck

M. Sattler
C. Gaegauf

n|w Fachhochschule Nordwestschweiz
Hochschule für Technik

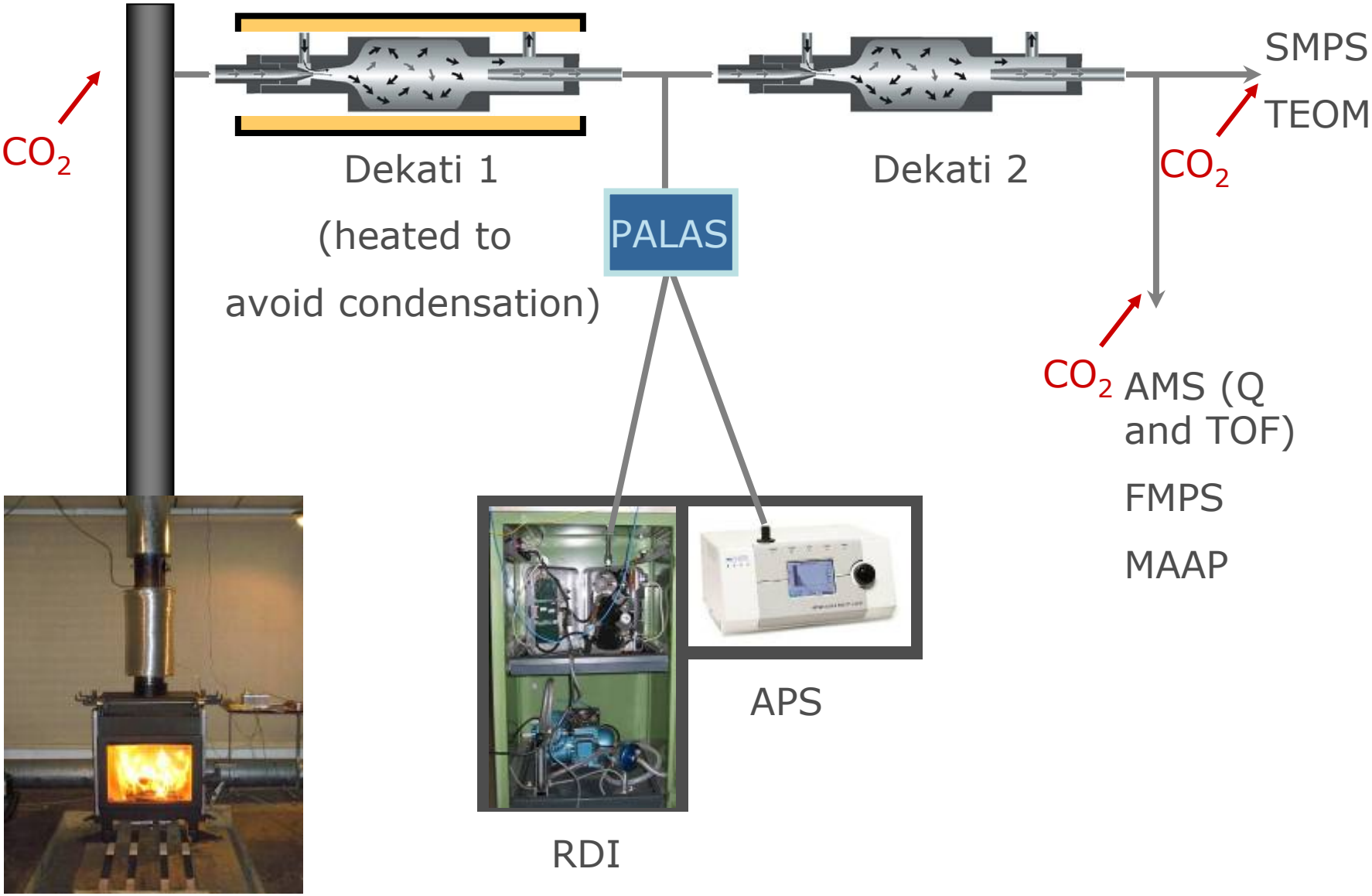
N.K. Meyer
H. Burtscher

PAUL SCHERRER INSTITUT PSI

M. Heringa
R. Chirico
A. Richard

[1](Alfarra et al., 2007)

woodburning study 2007

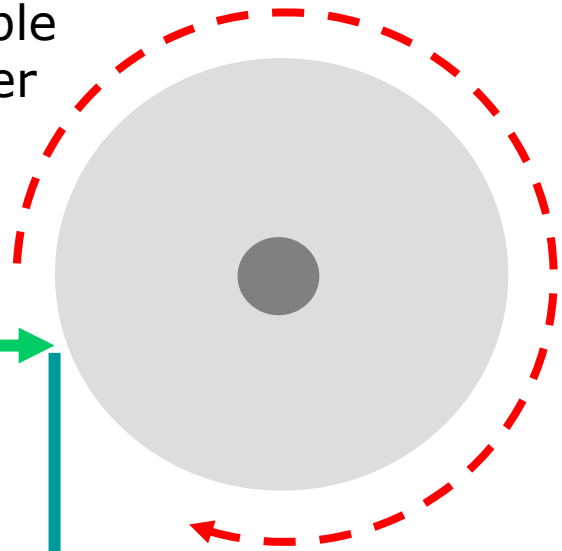


rotating drum impactor (rdi)

elemental analysis (Al-Zn)

sample holder

incoming X-ray beam



fluorescence light

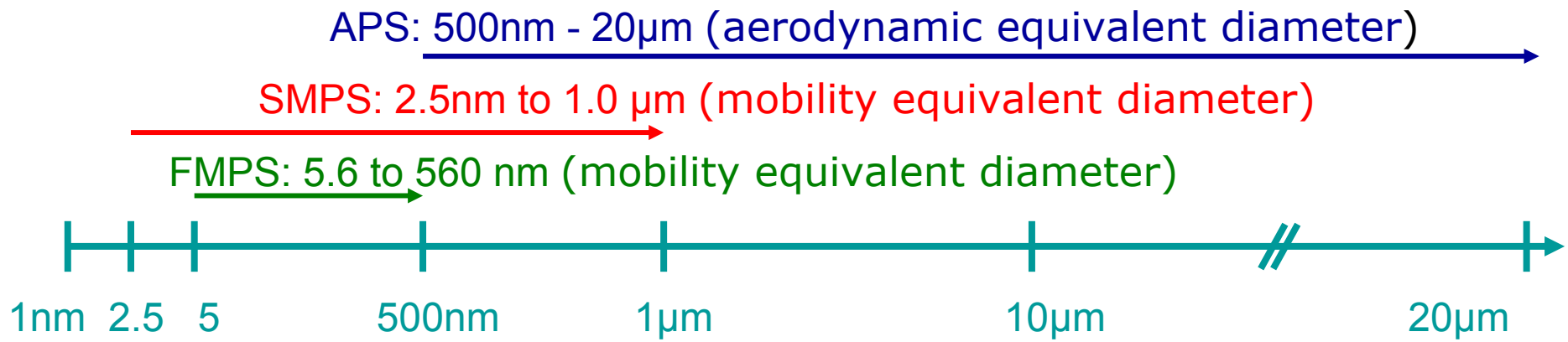
Swiss light source

Silicon detector

particle sizer (APS, SMPS, FMPS)

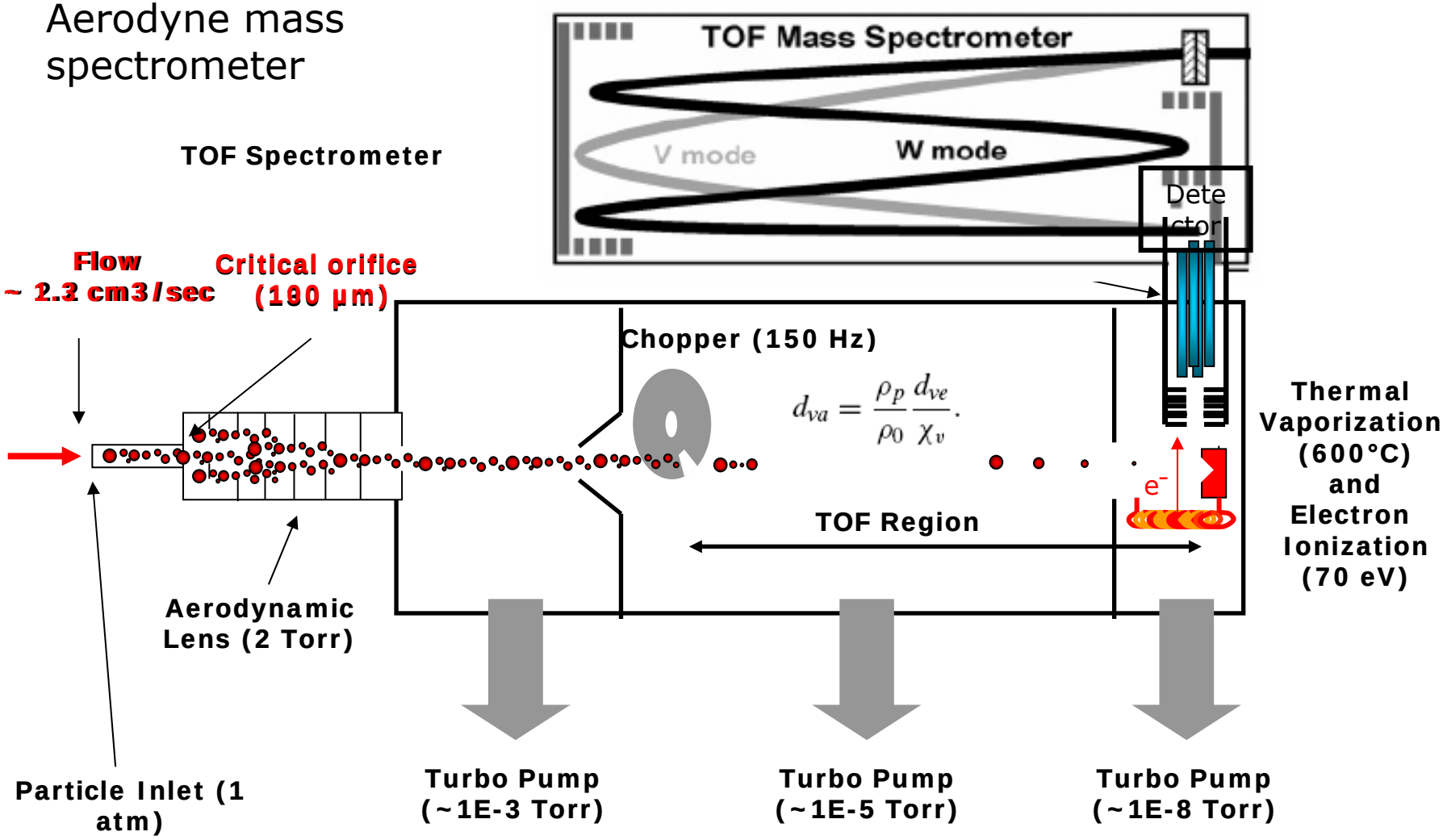
- time of flight measurement of individual particles in an accelerating flow field
- particle size binning based on an internally stored calibration curve
- size range: 0.5 to 20 µm
- time resolution: down to 1 sec
- concentration Accuracy: ±10%

- ***aerodynamic diameter*** is defined as the physical diameter of a unit density sphere that settles through the air with a velocity equal to that of the particle in question

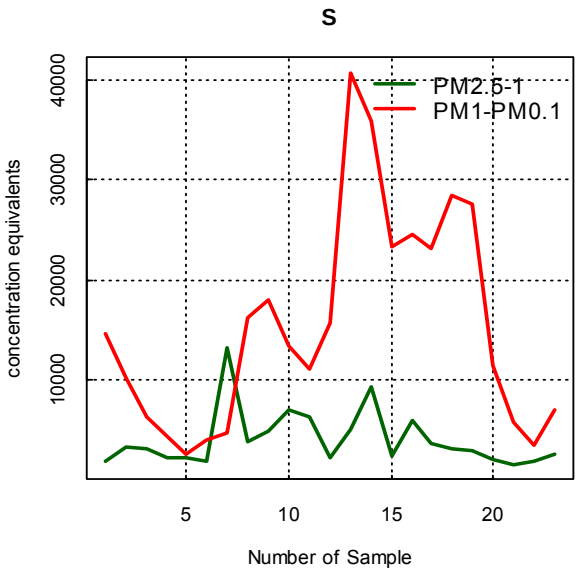


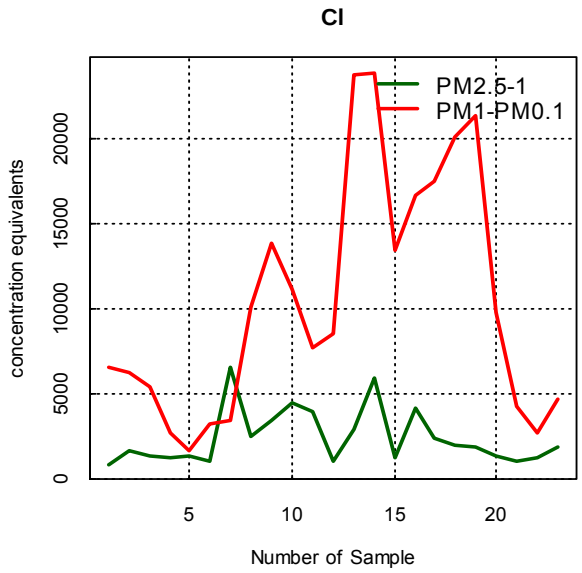
Q-AMS and HR-ToF-AMS

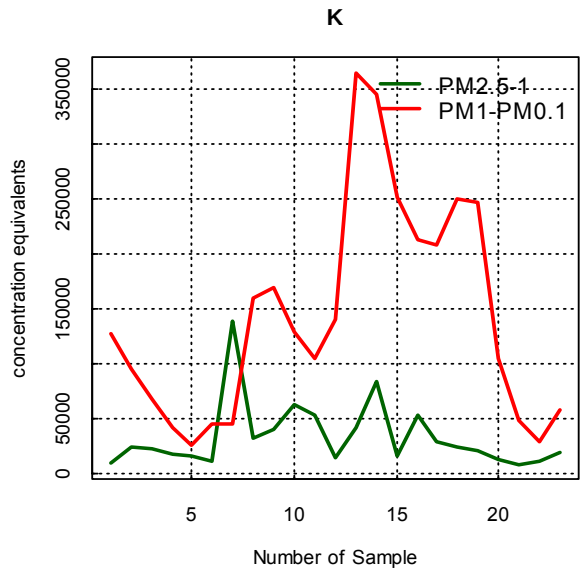
Aerodyne mass spectrometer

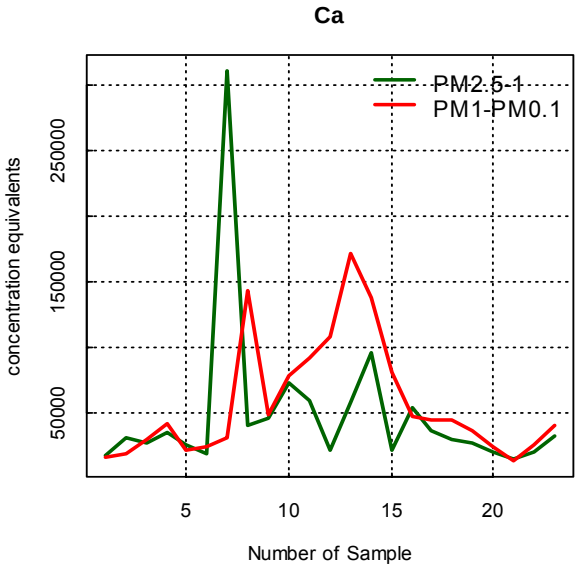


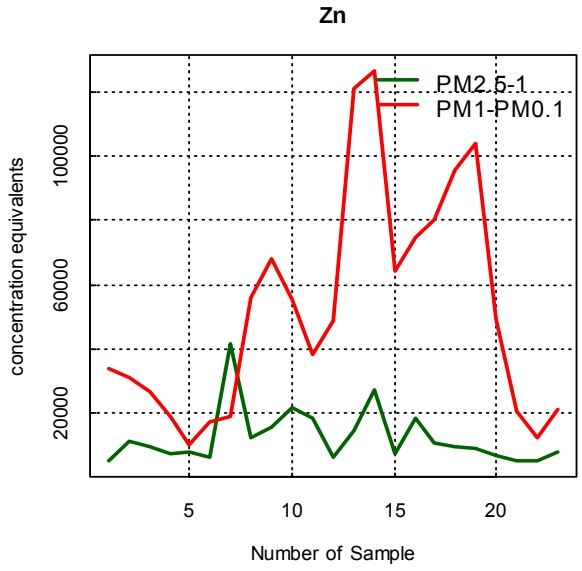
pellet oven, cold start: preliminary

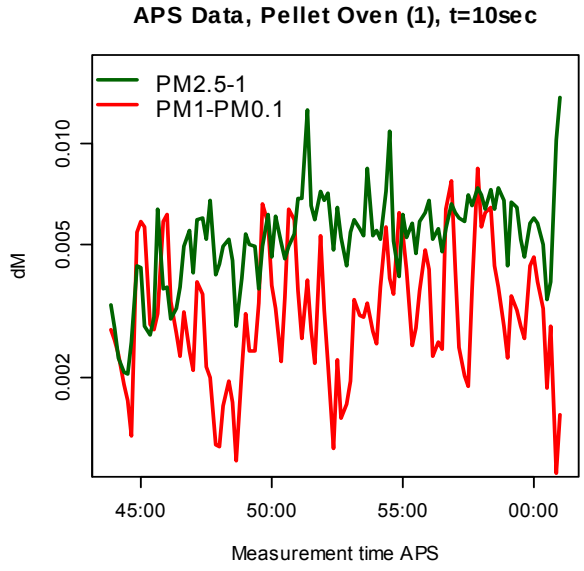






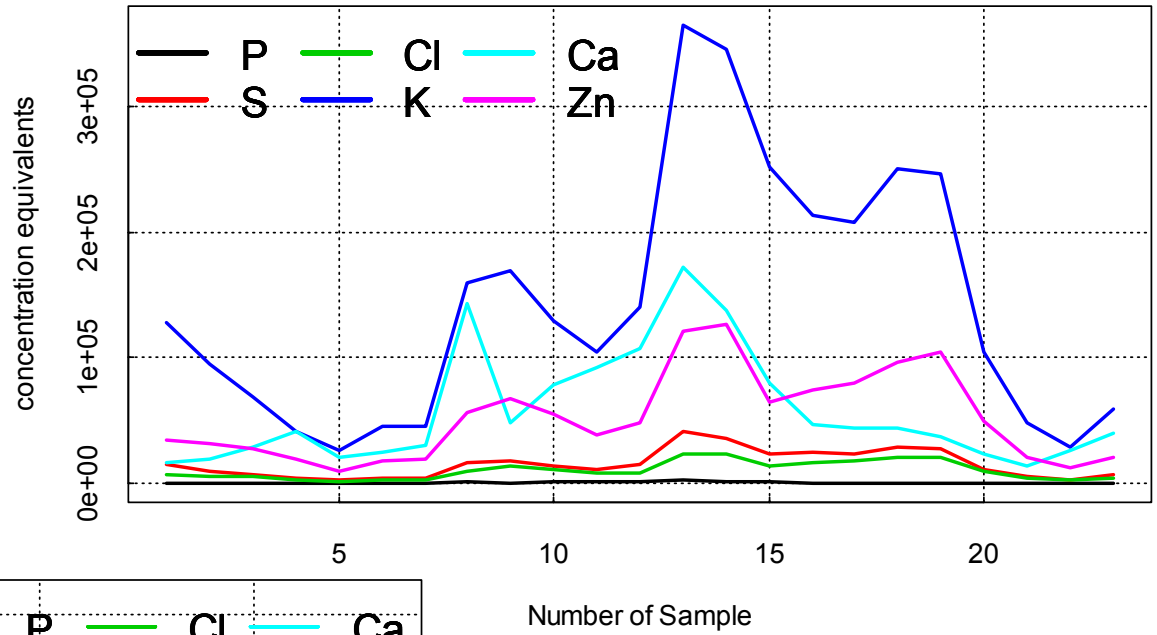


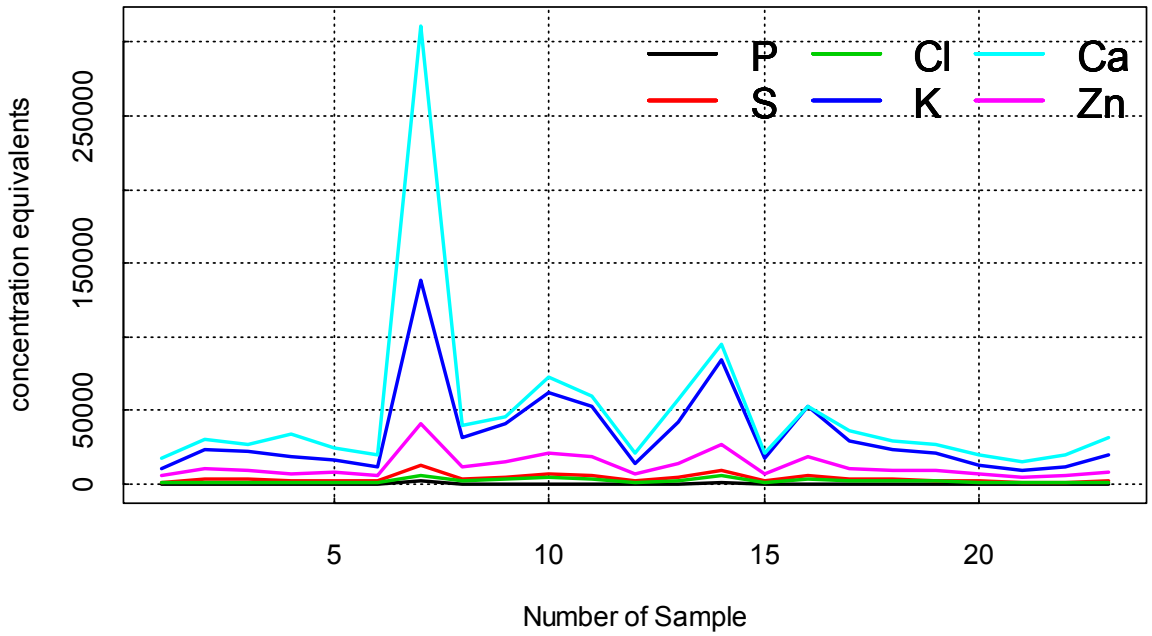




pellet oven, rdi

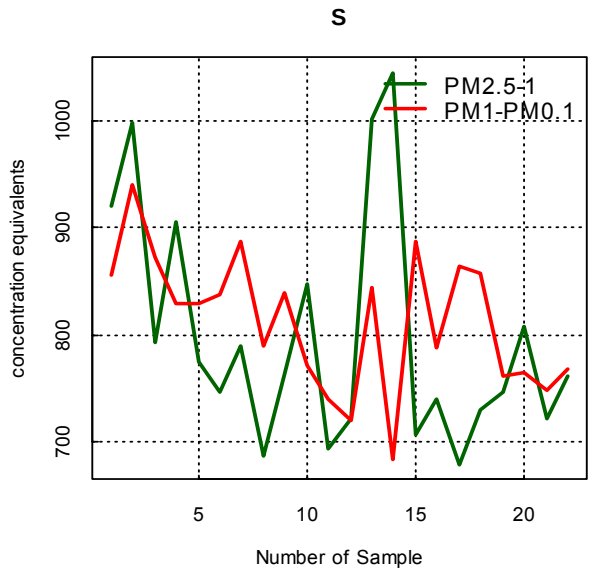
PM1-PM0.1

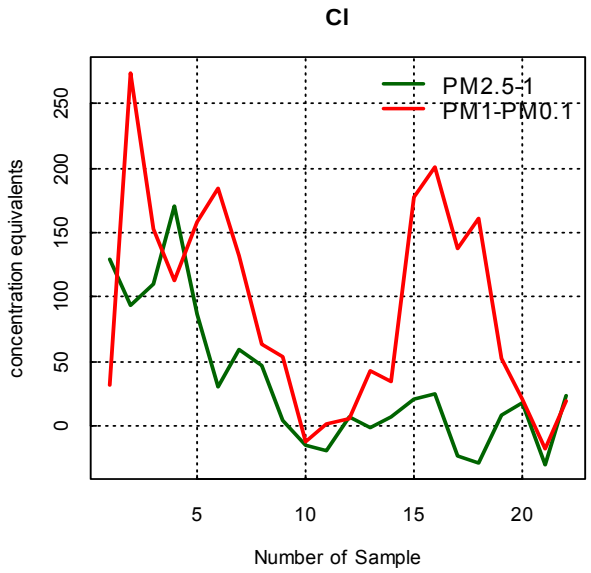


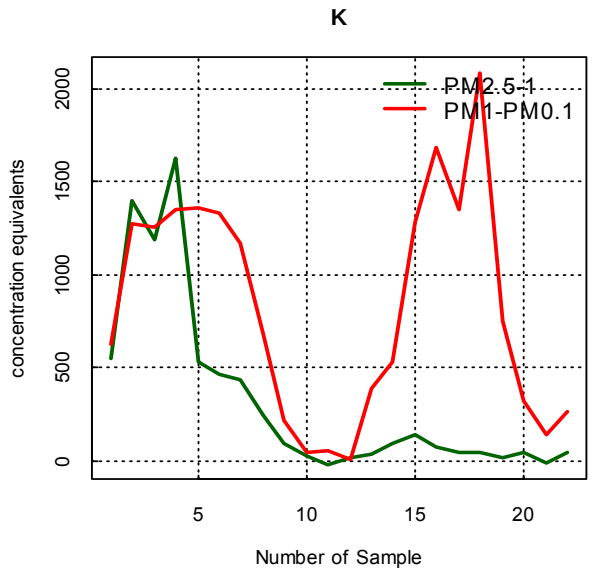


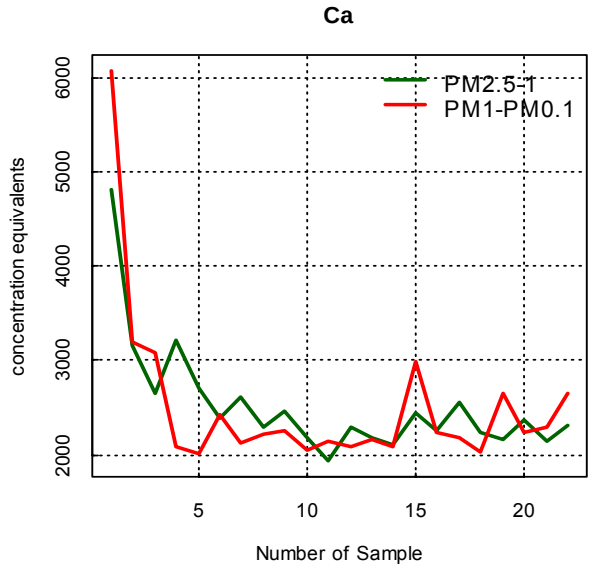
PM2.5-PM1

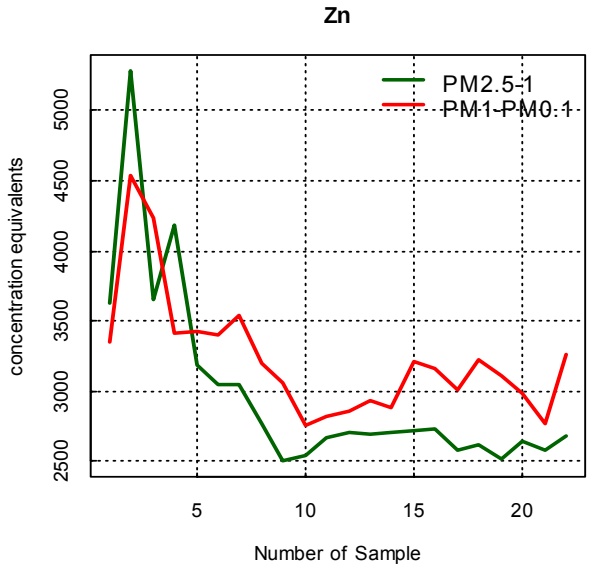
stove: preliminary results

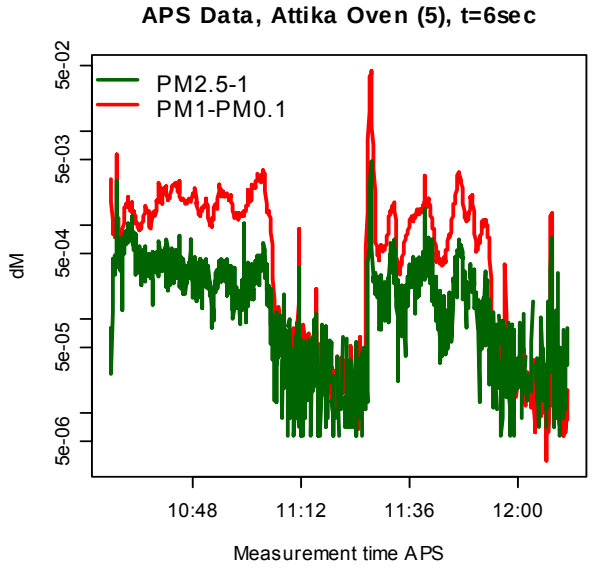




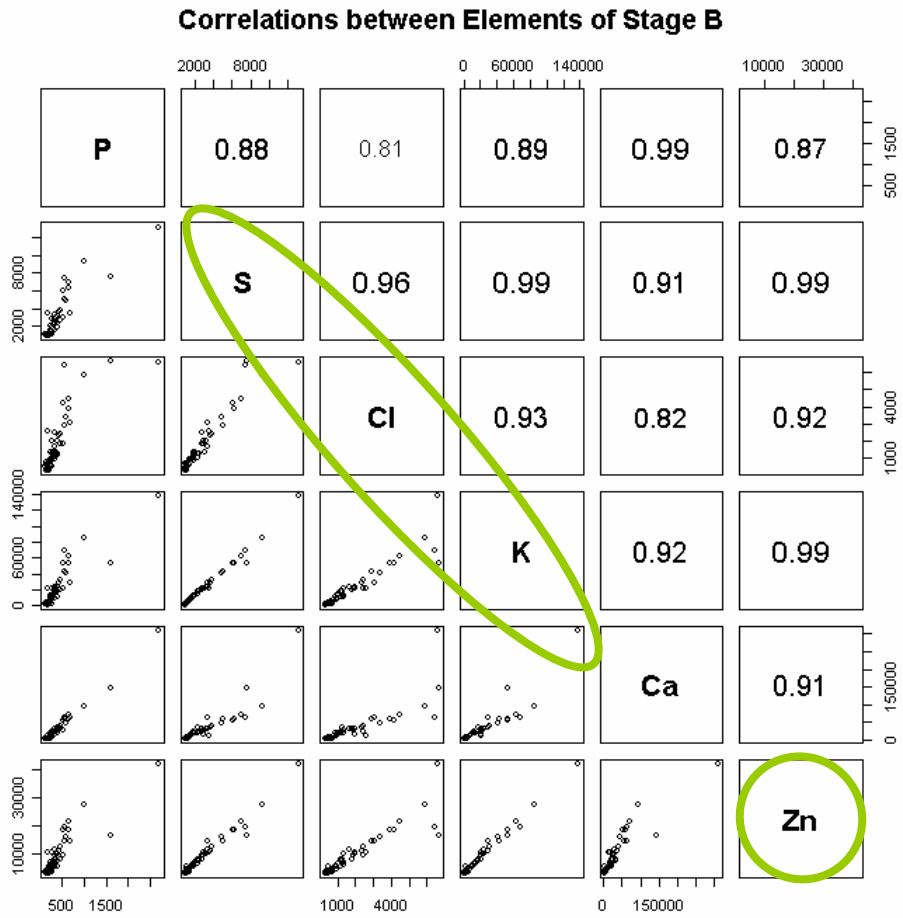


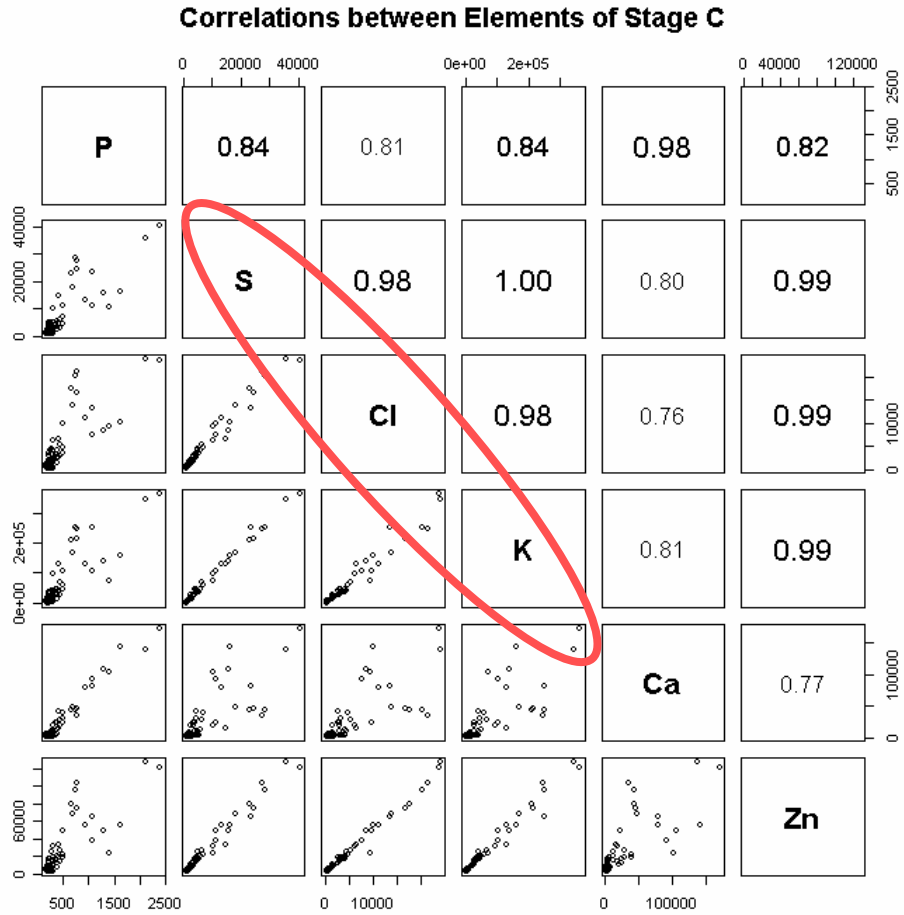






correlation of elements (pellet)



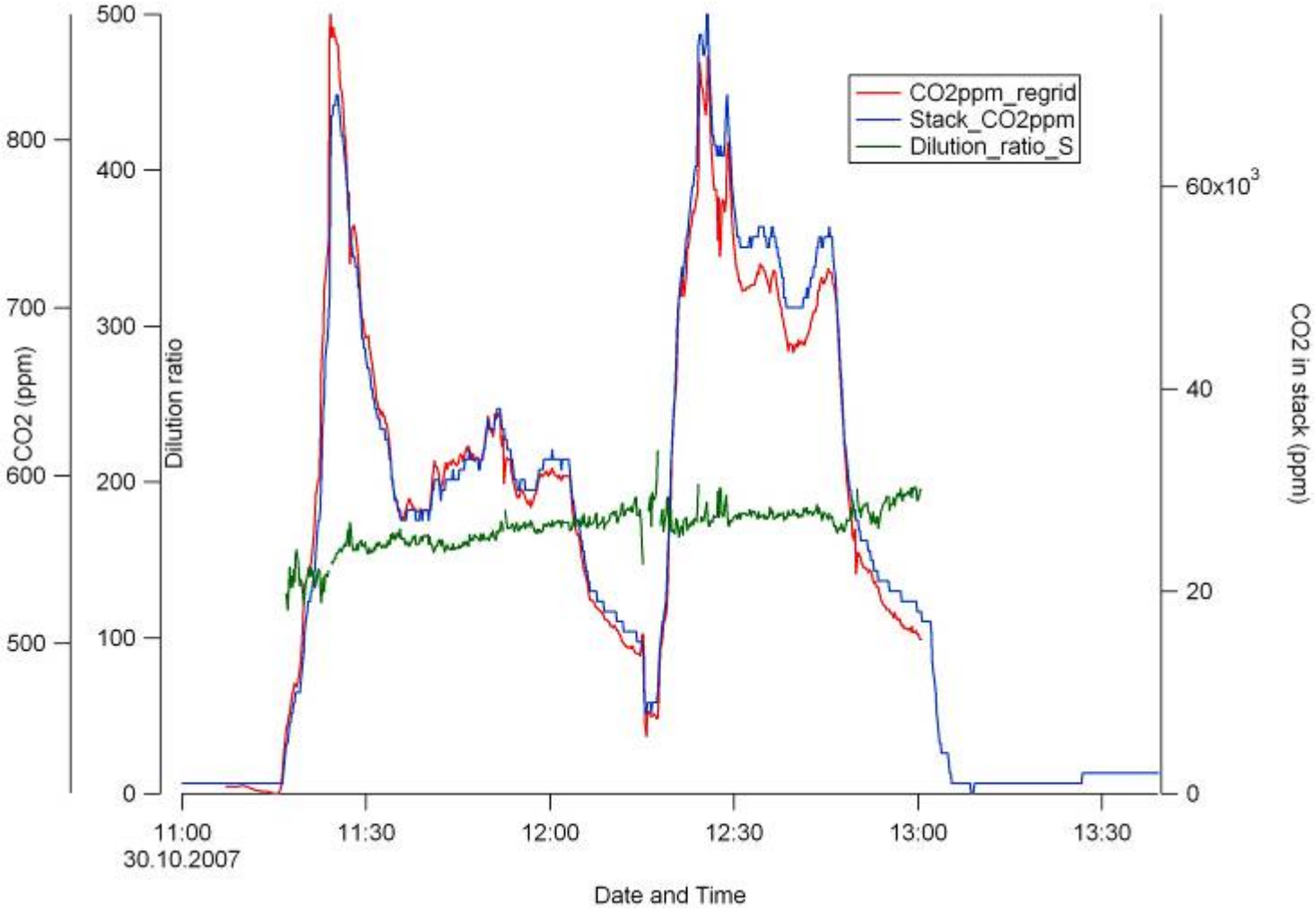


L.S. Johansson et al. / Biomass and Bioenergy 25(2003) 435– 446

E. Hedberg et al. / Atmospheric Environment 36 (2002) 4823–4837 **(+ Si)**

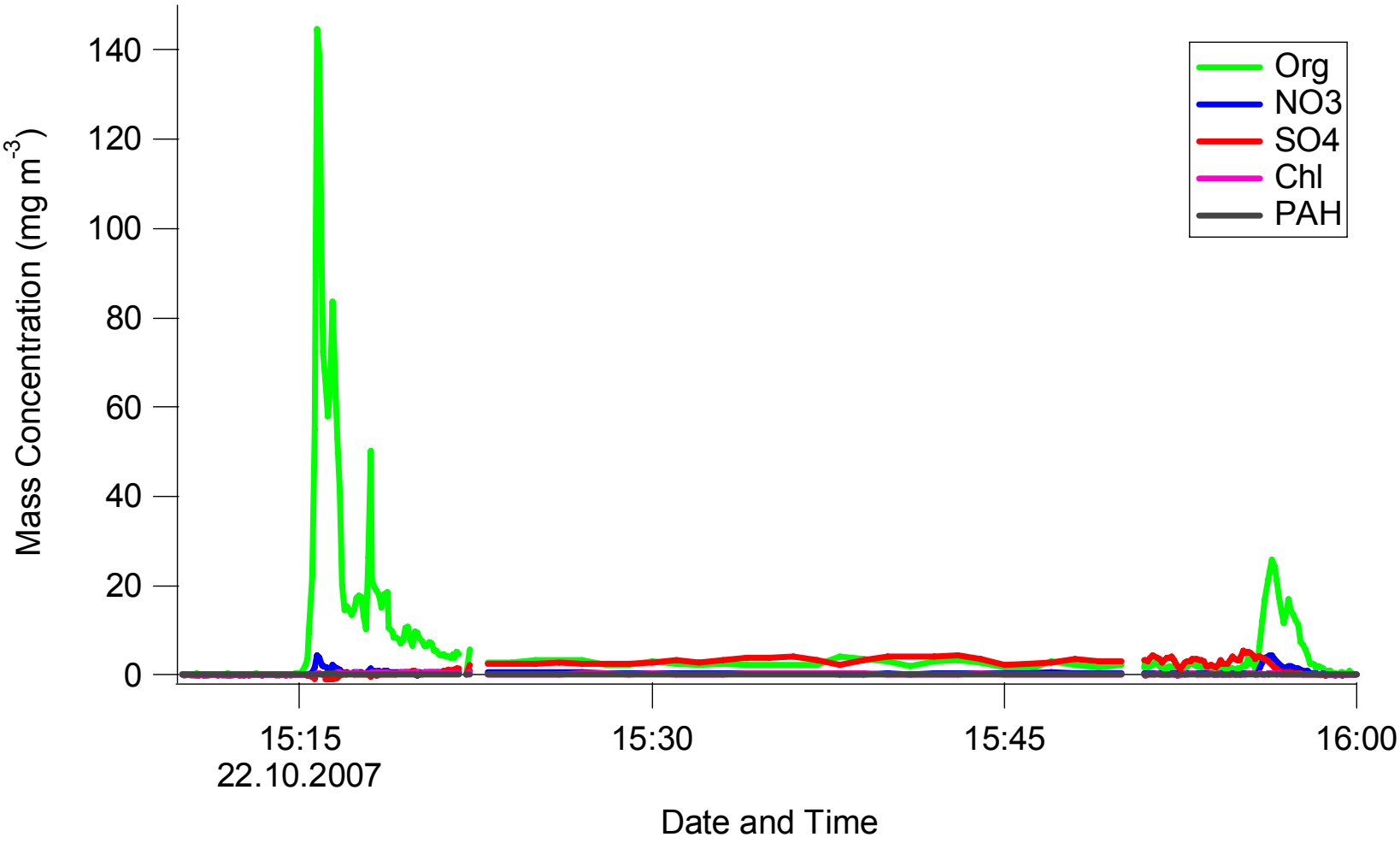
dilution ratio

example: insert oven, top down burn
=> ratio CO2 stack / CO2 @ AMS = 171



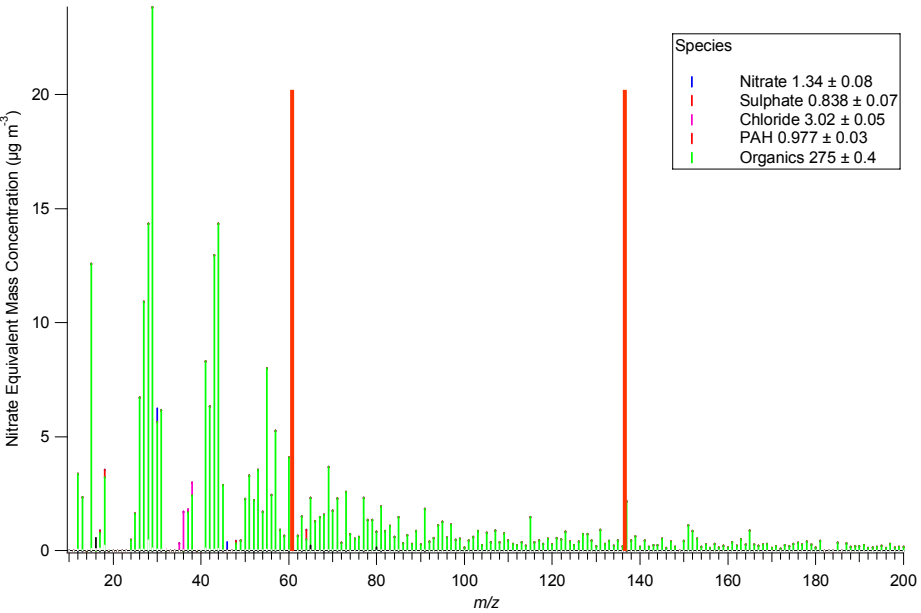
AMS results (pellet oven)

concentration is calculated back to concentration before dilution systems

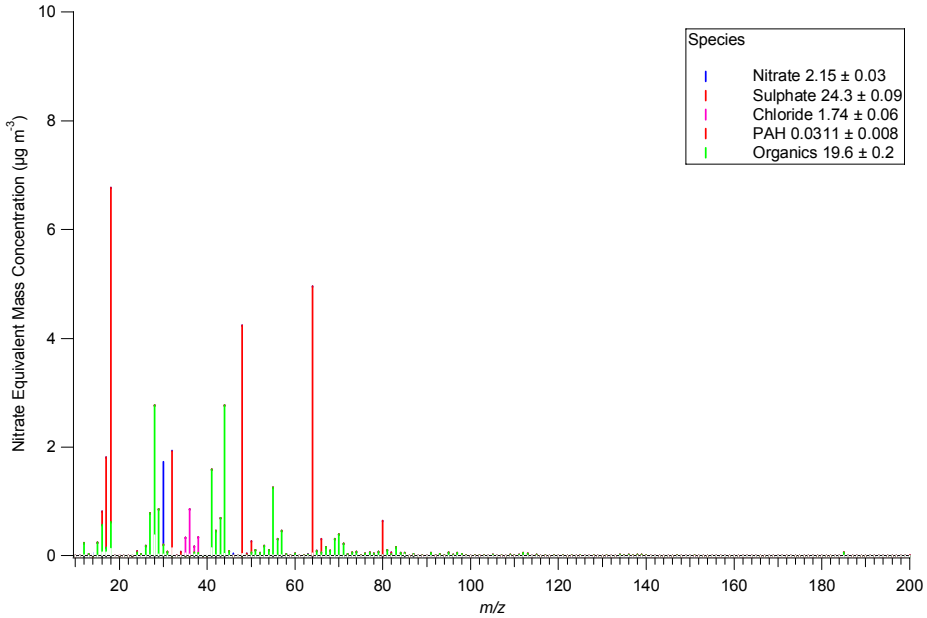


pellet automatic burner

start peak



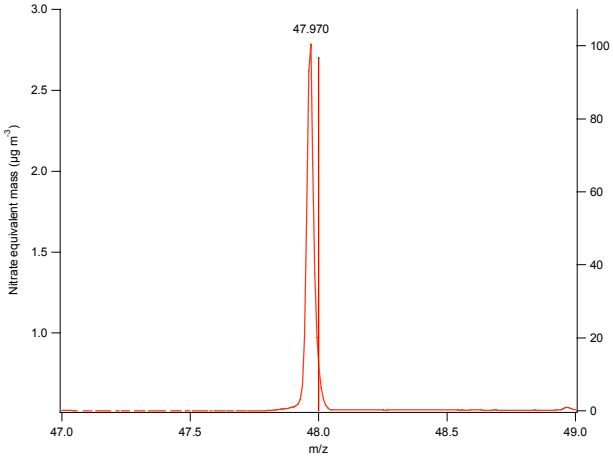
stable burning



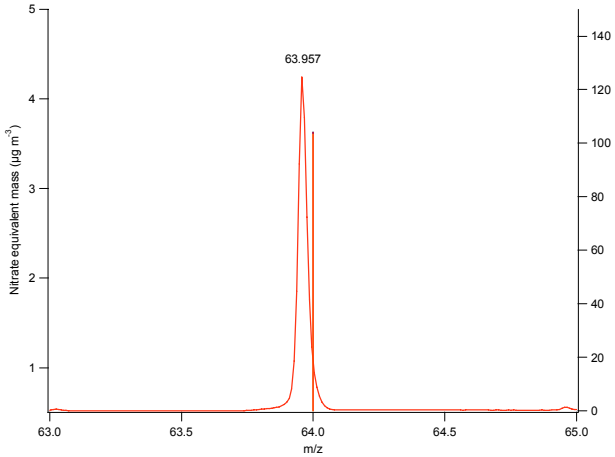
wood burning markers, m/z 60, 73 and 137, are mainly formed during the start

pellet oven, sulfate peaks

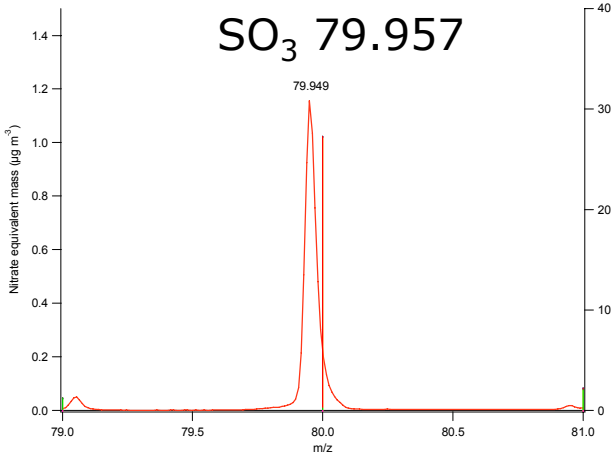
SO 47.967

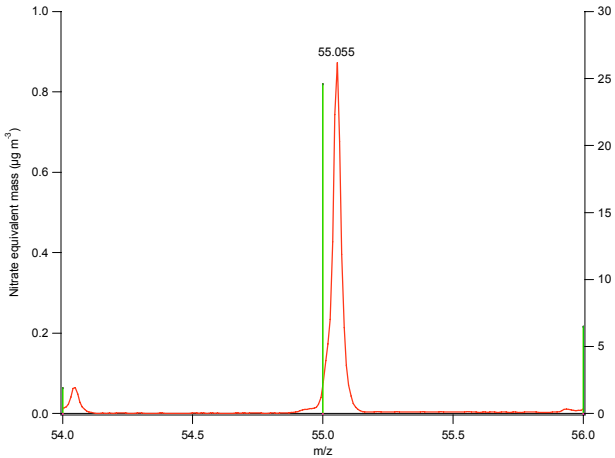


SO_2 63.962



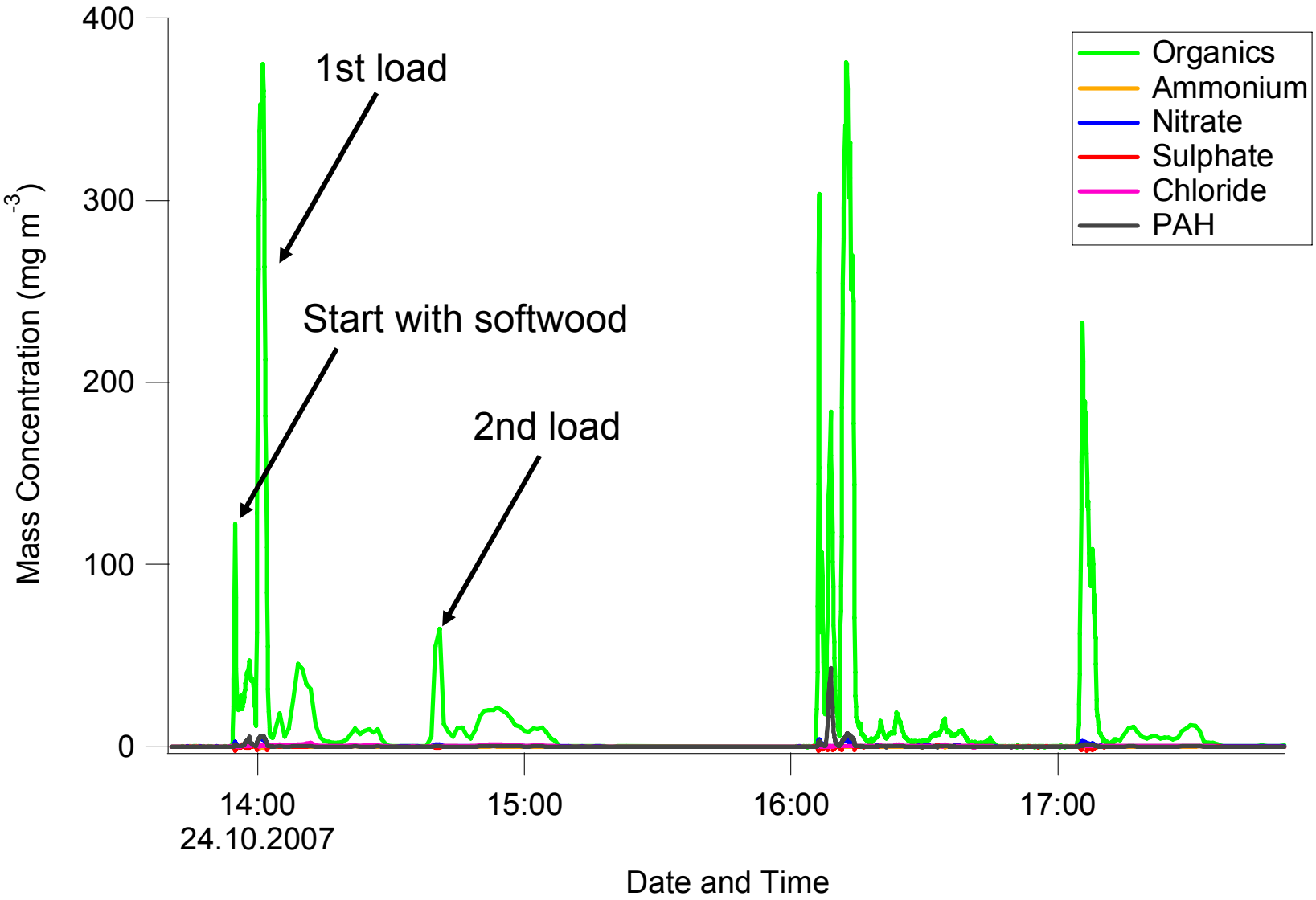
SO_3 79.957





C_4H_7 55.055 C_3H_3O 55.018

AMS results (logwood burner)



conclusions and outlook

RDI

- time resolved elemental analysis gives information about temporal variability of elements
- need for more burns to receive reproducible results

AMS

- the wood burning markers, m/z 60, 73 and 137, are mainly formed during the start
- high resolution data made it possible to discriminate peaks with the same nominal mass and to assign them to the correct species

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outlook:

- next campaign planned for October 2008 in co-operation with VERENUM, T. Nussbaumer et al. and FHWindisch


Thank you for your attention!