Pan-European Gas-AeroSOls-climate interaction Study, PEGASOS

***Astrid Kiendler-Scharr*, Patrick Schlag**

Introduction and motivation / Scientific objectives

Atmospheric aerosols have direct and indirect effects on the global climate. Particles alter the properties of clouds by acting as cloud condensation nuclei (CCN) and as surface for heterogeneous reactions. Particles are also known to have adverse health effects (Pope and Dockery, 2006).
These effects depend partly on the chemical composition of the particles.
Aerosol chemical composition can be measured online with high time resolution by the Aerodyne High Resolution Time-of Flight-Aerosol-Mass-Spectrometer (HR-ToF-AMS). Within the EU FP7 funded project PEGASOS intensive measurements of atmospheric chemical composition including gas and particle phase measurements were performed onboard of a Zeppelin airship. A campaign was scheduled for May 2012 targeting to explore the vertical distribution of aerosol composition in the planetary boundary layer over the Netherlands with specific focus on the area around Cabauw. The applicants operated a HR-ToF-AMS at the CESAR-tower in Cabauw, the Netherlands in May 2012, to provide ground based reference data on the aerosol chemical composition.
These measurements were made in parallel with measurements performed by the HR-ToF-AMS onboard a Zeppelin at several heights within and above the planetary boundary layer. The combined results will give an improved understanding of the vertical distribution of ambient aerosol composition. In particular this data set will provide insight into organic aerosol formation and aging processes. Thus, they will contribute to investigations on the effect of climate change on the number and mass concentration of atmospheric aerosols, and on the interaction of the atmospheric self-cleaning processes with the aerosol formation.

Additionally, the results are compared to data from the newly developed high resolution Thermo-Desorption-Proton-Transfer-Mass-Spectrometer (TD-PTR-ToF-MS) that was operated at Cabauw during this period. Previous comparisons of HR-ToF-AMS measurements with the quadrupole mass spectrometer TD-PTR-MS were performed by the group in summer 2008 and spring 2009 (Mensah et al., 2012) and with the TD-PTR-ToF-MS in November 2011. To investigate the seasonal cycle of aerosol chemical composition at Cabauw and to deepen the comparison of the two instruments, further measurements were needed to widen the range of meteorological and atmospheric chemical conditions covered in our observations.
Data analysis involves instrument comparison with the co-located TD-PTR-TOF-MS. Using high resolution mass spectrometric data from both instruments the relative contributions of N- and O-containing organic fragments to the aerosol composition are determined. This deepens the understanding of processes involved in organic aerosol mass formation and aging.

Reason for choosing station

Besides the fact that the previous campaigns mentioned above were also performed at Cabauw Experimental Site for Aerosol Research (CESAR), the Zeppelin made its flights mainly nearby the tower. This site is located at a rural site and is a representative for North-West Europe. Depending on the wind direction, the condition can be either of maritime or continental character.
Furthermore, the plain geography of the Netherlands and especially in the area of Cabauw reduces ambiguities in terms of air parcel convection and turbulences.

Method and experimental set-up

The Aerodyne HR-ToF-AMS sampled from a height of 5 m above the ground through a 7 m long, ½" Polyflo tubing. It measured the size distribution and chemical composition of particles and was operated 24 hours/ 7 days a week for 5 weeks with a time resolution of 4 minutes. Its transmission efficiency for particles in a range of 70 nm to 500 nm is almost 100 % (Jayne et al., 2000).
The sampling line was shared by a Monitor for AeRosol and GAses (MARGA). Close to the AMS inlet, a Condensation Particle Counter (CPC, Model TSI 3785) was deployed, resulting in an overall flow of 1080 mL min^{-1} between the MARGA inlet and the AMS inlet through 1.5 m stainless steel tubing with an inner diameter of 4 mm.
Once a week the instrument was maintained and calibrated for the ionization efficiency and particle size using a calibration rack containing an aerosol-generator and a Differential-Mobility-Analyzer (DMA), and a CPC (Model TSI 3025a). To determine the gas phase background signal, measurements with an in line particle filter was performed every 2-3 days.

The TD-PTR-ToF-MS also sampled from a height of 5 m above the ground through a separate, 10 m long copper tube with an inner diameter of 4mm. It is equipped with both a gas and an aerosol inlet and collects ambient particles in the size range of 70 nm to 2 µm at an air sample flow rate of 1L/min (Holzinger et al., 2010a; Holzinger et al., 2010b)

Preliminary results and conclusions

Figure 1 shows the temporal evolution of the total aerosol mass as well as of the most important aerosol species. Clearly seen are a number of periods with different mass loadings and different fractional abundances of these species, partly dominated by organics, sulfate or nitrate. These observations have to be referred to meteorological data like wind directions.
A first evaluation of the diurnal cycles shows night time maxima of the total mass and of the concentrations of organics and nitrate. The last can be explained by the heterogeneous conversion of nitrate in the night and the volatilization of nitrate, especially ammonium nitrate, during the day.
The average particulate mass loading measured by the HR-AMS during May and the beginning of June 2012 was 7.8 µg m^{-3} and lower than observed in previous campaigns in Cabauw in May 2008 (9.7 µg m^{-3}), but higher than the amounts of November 2011 (5.0 µg m^{-3}) and March 2009 (5.6 µg m^{-3}; Fig. 2). The organic fraction was the dominant species (37 %) in May 2012, followed by nitrate (25 %) and sulphate (20 %). The observed aerosol composition was thus overall similar to the results from autumn 2011 and summer 2008, whereas in spring 2009 a dominant nitrate fraction (42 %) was observed.

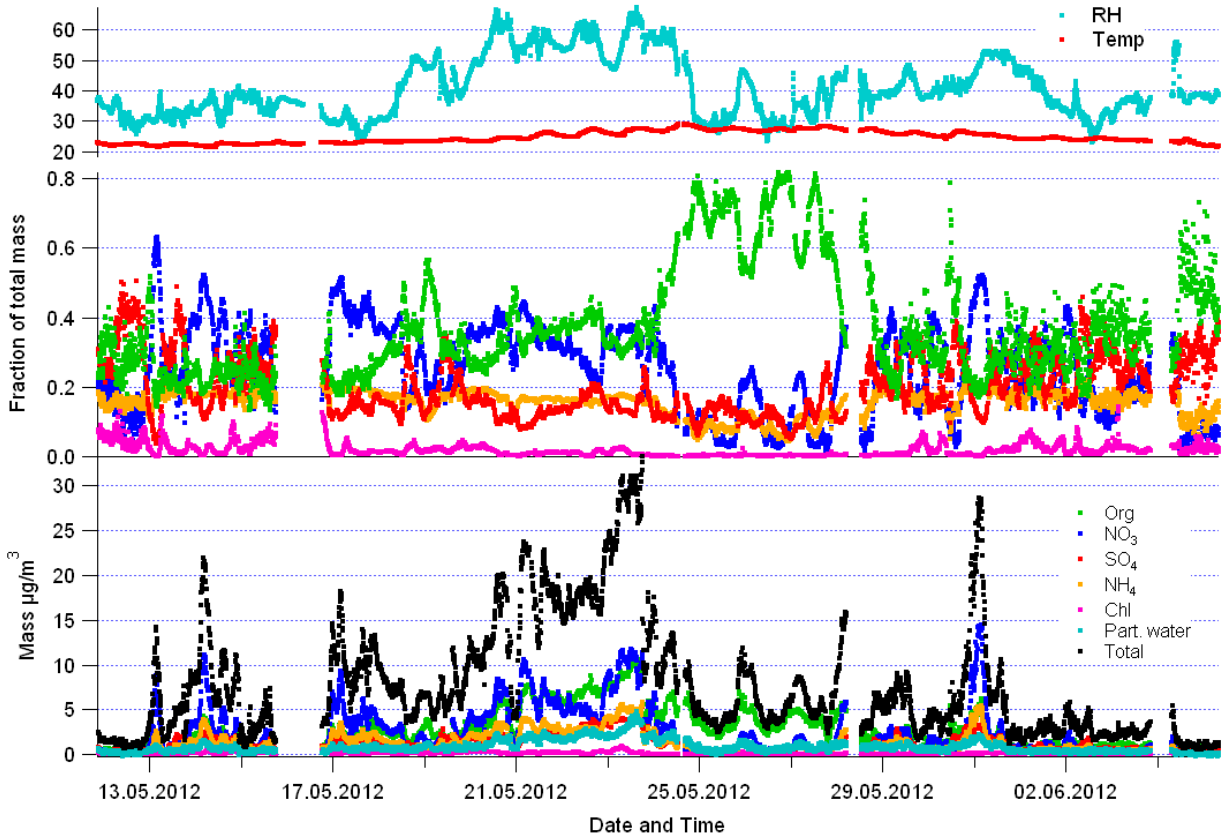

Fig. 1: Mass concentrations and fractional abundances of aerosol species; relative humidity (RH) and temperature (Temp)

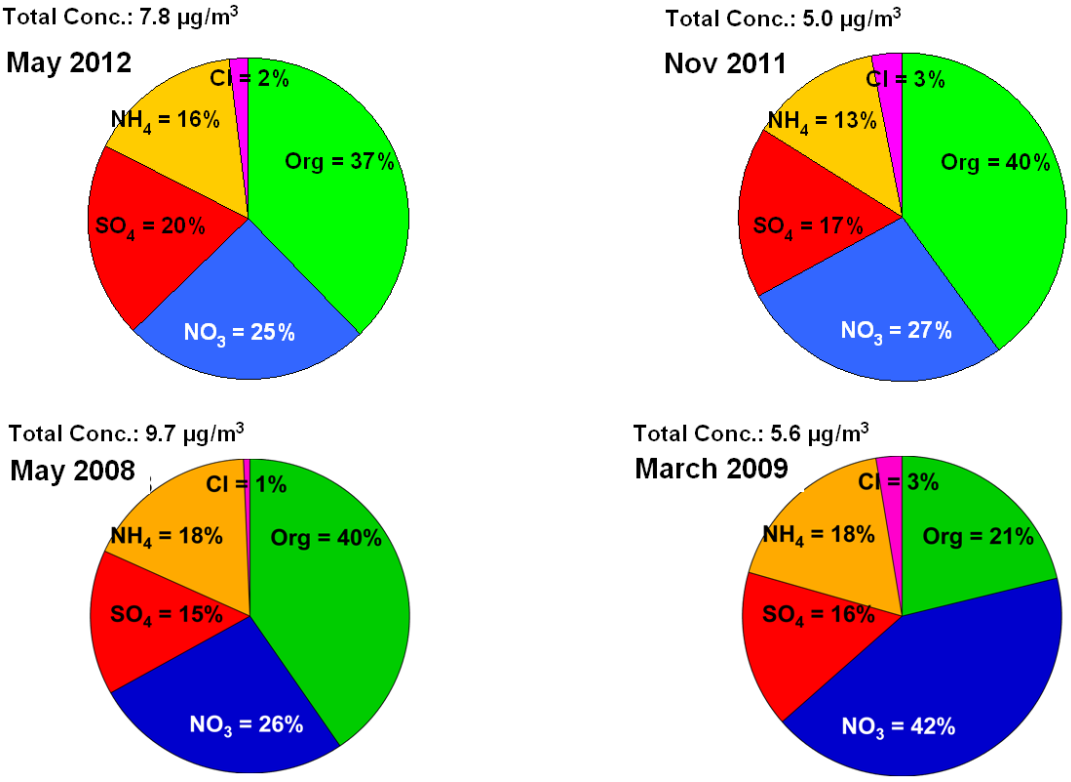

Fig. 2: Aerosol components average

A first evaluation of the aerosol measurements onboard the Zeppelin over Cabauw, which were performed in parallel to the data acquisition at ground level, shows vertical profiles of the chemical species, where especially the nitrate and organic amounts are lower above than within the planetary boundary layer. In contrast, sulphate concentrations had only little changes with altitude.

Outcome and future studies

Both AMS data sets measured onboard the Zeppelin and on the ground, will be used in detailed analysis of the vertical distribution of ambient aerosol composition.
Amongst others, further AMS data analysis will evaluate aerosol composition as function of size. For more detailed analysis of the organic fraction elemental analysis and Positive Matrix Factorization (PMF) will be performed.
Directly after the PEGASOS campaign, follow-up measurements at the CESAR tower were performed in June and July 2012 and were coordinated with a European intensive measurement period defined by EMEP.

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