Aerosol source-receptor studies

Hans Puxbaum

1. Overview

Dispersion models are based on source emission data and prediction of ambient concentrations making use of meteorological dispersion parameters. Conversely, source receptor models are based on a detailed knowledge of the chemical composition of the ambient aerosol and retrieval of the source impact by advanced statistical or refined tracer methods. The statistical methods make use of a series of ambient aerosol data and derive the source information by multivariate techniques such as PMF (Positive Matrix Factorisation). The tracer methods are based on knowledge of the typical source composition ("Source Profile" and derive the source information by a fitting procedure of sources from the ambient composition, designated as the "Chemical Mass Balance" (CMB). A related but simpler approach is the use of mass closure models, which make use of characteristic tracers for certain source categories. At Vienna University of Technolgy a mass closure model based on new characteristic tracers for major source categories was developed where most of the tracers are major components of the source profiles. This "Macro Tracer Model" was applied for a series of aerosol studies which are discussed in detail later in this chapter. The new tracers developed for the "Macro Tracer Model" comprised organic species such as cellulose – the main constituent in green plants (Kunit and Puxbaum 1996, Puxbaum and Tenze-Kunit 2003); "HULIS" ("humic like substances"), a predominantly secondary organic aerosol component (Limbeck et al. 2005); bacteria and fungal spores, the main components of the bio-aerosol (Bauer et al. 2002, Bauer et al. 2008) and levoglucosan, mannosan and galactosan, specific wood combustion tracers (Caseiro et al. 2007). Methods for the determination of the wood smoke tracers have been known before, however in the course of the macro tracer development it turned out, that in addition to the wood smoke tracers arabitol and mannitol were potent tracers for fungal spores and thus a specific method for the simultaneous determination of those tracers was desirable (Iinuma et al. 2009).

Also inorganic tracers were investigated: Si as main constituent of the mineralic aerosol (Peng et al. 2010) and platinum group elements as tracers for catalyst equipped automobile emissions (Limbeck et al. 2003). Finally, a method for carbonate, a missing component in the group of inorganic ions was introduced (Jankowski et al. 2008).

The Macro Tracer Method was applied in various forms in many international studies. Specific applications were for assessing the contribution of wood smoke to ambient PM levels (Puxbaum et al. 2007, Lanz et al. 2008, Sandrade-

wi et al. 2008, Pio et al. 2008). The formation of HULIS in smog chamber experiments was investigated in a study with the Paul Scherrer Institute (Baltensperger et al. 2008). The three main projects where the Macro Tracer Method has been applied (AUPHEP, CARBOSOL, AQUELLA) are discussed below; some of the data have also been used for the "European Aerosol Climatology" paper (Putaud et al. 2010).

In addition to these tracer methods, based on chemical analyses, source –receptor studies in Austria were also performed on purely meteorological methods. The trajectory statistics as developed by Scheifinger and Kaiser (2007) and used for gaseous pollutants by Kaiser (2009) found application in a number of studies performed for PM (e.g. Salvador et al. 2010, Offenthaler et al. 2009, Kaiser 2007). Results were also used with respect to health studies (Moshammer et al. 2009).

2. Mass balance studies

At an early stage of development mass balances were applied to derive information about major source impacts on the fine particles. A study in the savanna of South Africa was deployed to obtain information about the biogenic background component of the organic aerosol. Although situated at a background site considerable contributions from inorganic secondary aerosol (mainly sulfate) were observed. The main component in aerosol was soil dust (36%), followed by organic material (28%), and ammonium sulfate (27%). Aerosol humidity was 6%, BC 2% and plant debris 7%. A diurnal trend of the OC/EC ratio of 16 during day and 6 during night indicated a daytime source for organic components considered to originate from gas-particle conversion from biogenic emissions (Puxbaum et al. 2000).

The methods developed were also employed for aerosol studies in Bangladesh. The aerosol composition in the mega-city Dhaka was dominated by soil-type mineralic material, followed by carbonaceous material and ions (Salam et al. 2003a). From EC/OC relations and information from trace elements it was derived, that biomass combustion was not the dominant source for OC in the aerosol. Differences in the EC:TC and K:EC ratios in the 3 southeast Asia mega-cities (data available from Dhaka, Bangladesh; Lahore, Pakistan; Mumbai, India) indicated the aerosol source mix in southeast Asian cities varies considerably at national or even regional scales. A companion study at a background site in the Gulf of Bengal identified two different transport situations: In one period the EC/OC ratio was pointing to a predominantly fossil fuel dominated carbonaceous fraction in the aerosol; in the other period when he aerosol arrived from Indian coastal regions around 50% of the carbonaceous material was from biomass combustion (Salam et al. 2003b).

A further step, also based on mass balances was the TU Vienna contribution to the AUPHEP study (for a detailed presentation of the project see this compendium, pp. 130 ff). The gas and aerosol part was designed as a dual-site study with one site in the 9th district in Vienna, and one background site about 40 km upwind northwest of Vienna. Collocated PM2.5 and PM10 samplers were used for collecting aerosol filters analyzed for carbonaceous material, ions, and trace metals. The PM2.5 and PM10 measurements at the 2 sites indicated only low aerosol generation activity in the city on an annual basis. Defining the term urban impact as the difference between observations at an urban and a regional background site, an annually averaged urban impact for PM2.5 was 3.4 µg/m3 and for PMC was 3.3 µg/m3 (coarse fraction PMC = PM10-PM2.5). The relative increase of particulate matter (PM) concentration at the urban vs. background sites was annually averaged to be 19% for PM2.5 and 60% for PMC, indicating a relatively strong mineralic aerosol source in the city. Major chemical constituents of the PM2.5 urban impact were black carbon (BC), organic carbon (OC), and SO_4^{2-}; main constituents of the PMC urban impact were OC and indicators for mineral aerosol (Fe, Ca, Mg, Na, K). The BC:TC ratio of PM2.5 urban impact was typical for combustion sources, e.g., automotive traffic, oil or coal combustion. Urban coarse OC was tentatively attributed to non-pyrogenic sources. From trace metals (As, Cd, Co, Cu, Cr, Mn, Ni, Pb, V, Zn), only Cd, Ni, Pb, and Zn exhibited a slight cold season enrichment in the urban airshed. From the weak seasonal signal for oil or coal combustion indicators, it was concluded that local domestic heating sources are using clean fuels (Puxbaum et al. 2004).

At this stage it became clear, that for a better understanding of the sources of the mineralic aerosol the main components Si and Al had to be included in the list of determined components. This request posed a considerable challenge for the sampling procedure, as the standard filter for chemical analysis was generally a pre-cleaned quartz-fibre filter. Thus, for the analysis of mineralic constituents a further sampler equipped with cellulosic filter material was applied, the filter being analysed by WLD-XRF (wavelength dispersive X-ray fluorescence) (Peng et al. 2010). From a half year data set from Goess (Styria) interesting details of the atmospheric occurrence of mineralic material emerged: Silicious material predominantly from crustal material, e.g. street abrasion, aeolian dust (analytical tracers Si, Al) showed in the temporal trends a decoupled behaviour from "carbonates" (analytical tracers soluble Ca, Mg). A decrease in Si and Al and an increase in carbonates, Na, K, Zn and P were observed during the cold season. Si and Al were highly correlated in all seasons with a Al/Si ratio of 0.27, identical with the respective ratio observed for the continental upper crust (Fig. 1a). Ca shows a wide variation in the relation to Si with a lower limit of Ca/Si 0.3, which is higher than the equivalent to the natural ratio in the upper crust (0.13). The Ca/Si ratio of 0.3 – dotted line in Fig. 1b – is considered to "represent" the street abrasion material (Fig. 1b). The in-

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crease in variability and of absolute levels of carbonates during the cold season was most likely deriving from dolomite gritting material. A marked increase in winter was also observed for Na and Cl originating from the use of thaw salt. A winter increase of K, Zn and P pointed to domestic wood combustion as common source. PM10 levels were 12 μg/m³ higher in December than in July, due to increased carbonates (gritting), thaw salt, wood smoke and inorganic secondary material, predominantly nitrates (Peng et al. 2010).
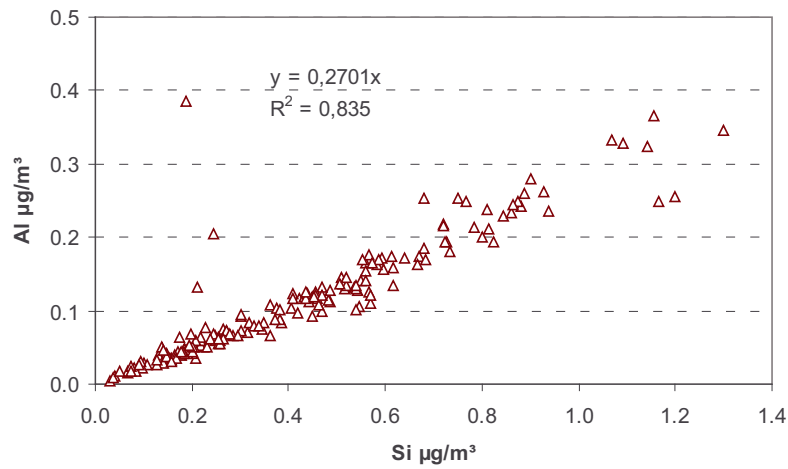

Figure 1a: Al and Si concentrations from the half year data set from Goess (Styria). (Data from July 2006–Jan 2007, from Peng et al. 2010).
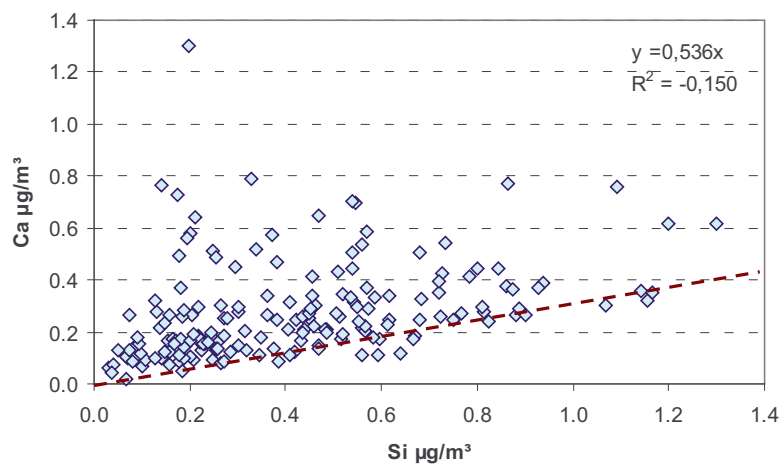

Figure 1b: Ca and Si concentrations from the half year data set from Goess (Styria). (Data from July 2006–Jan 2007, from Peng et al. 2010). The dashed line marks the Ca/Si ratio 0.31 considered the contribution of Ca from street abrasion.

3. AQUELLA

The Austrian project AQUELLA has been launched in 2003, with the participation of the countries Vienna, Styria, Salzburg, and from 2004 also with Carinthia, Lower Austria and Burgenland, to investigate the causes of the violations of the PM10 short term criteria (Figure 2). AQUELLA was based originally on the CBM technique, but then a specifically developed macro-tracer concept was applied fore the source analysis. Sampling was performed at multiple sites in and around the main urban settlements, using 2 parallel operated PM10 samplers, a Hi Vol with quartz and a Lo Vol with cellulose-ester filters. The project involved 23 cooperating scientists and students, operating 11 different analytical methods. The macro-tracers involved and the factors for deriving source contributions are listed in Table 1. The introduction of a series of tracers present in the aerosol at concentrations >0.1%, e.g. Levoglucosan (wood combustion tracer), Arabitol (tracer for fungal spores), cellulose (tracer for plant debris) and "HULIS" ("humic like substances", a group of macromolecular compounds considered to originate from secondary formation processes) in addition to ions and mineralic constituents (silicates, carbonates) allowed a discrimination of 8 influencing sources composing about 90% of the fine particles (PM10 and PM2.5).
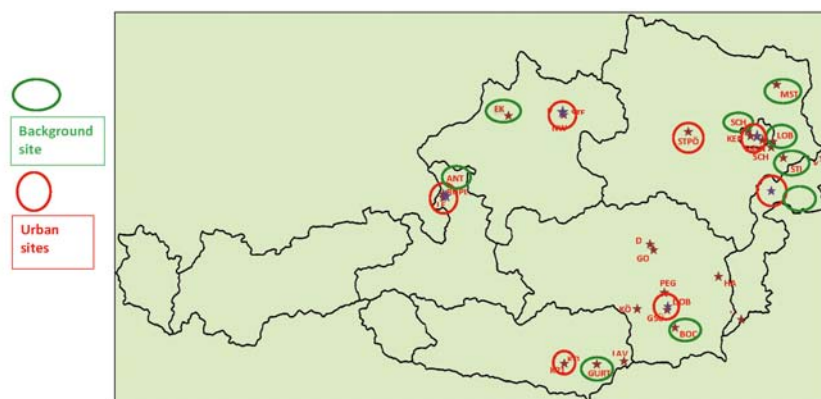

Figure 2: AQUELLA sites in Austria.

Table 1: Macro-tracers used to derive source contributions, associated source groups and main sources.

Analyte	Source Group	Sources
Levoglucosan	Biomass-Smoke	Wood combustion (domestic)
		Wild fires, open fires
Arabitol	Primary Biological Particles	Fungal spores
HULIS (Humic Like Substances)	Organic Secondary Aerosol	Winter: Wood combustion Summer: natural Sources
Cellulose	Plant Debris / Detritus	Plant Debris
Soot (BC or EC)	Combustion	Diesel-Emissions Wood-Combustion Coal-Combustion Fuel Oil-Combustion
Si, Ca	Mineralic Dust	Street Abrasion Winter Sanding (Gravel) Sidewalk Sanding Construction Sites Dusty Vehicles Aeolian Dust
NaCl	Thaw Salt	Thaw Salt Sea Spray
Ammonium, Nitrate, Sulfate	Inorganic Secondary Aerosol	NH_3: Agriculture SO_2: Coal, Smelters NO_x: (Hot) Combustion
Unidentified		Further secondary OM Aerosol humidity

The results of the AQUELLA study are subject of reports to the country governments. Reports are available from the official information systems of the provinces Styria (Bauer et al. 2007a, 2008a, 2009a, b), Carinthia (Bauer et al. 2007b) and Upper Austria (Jankowski et al. 2009).

From the seasonal variation of the PM10 daily averages it became evident, that exceeding of the PM10 air quality standard value of 50 μg/m³ took place generally during the cold season. Thus a first step included an analysis of different influences on the aerosol concentration during the warm and the cold season. Two possibilities had to be taken in consideration:

a. During the cold season due to more frequent low level inversions ventilation is generally lower than during the warm season.

b. During the cold season additional sources (e.g. domestic heating) are active compared to sources which are relatively constant throughout the year.

The influence of the ventilation was investigated by means of a traffic tracer, since traffic is a relatively constant aerosol emission source throughout the year. The main particulate exhaust emission from mobile sources is carbonaceous material. This, however, is also emitted from biomass and fossil fuel combustion. New tracer compounds from vehicular emissions are platinum elements used in the catalysts. Although emitted in very minor amounts, they can be used to investigate the ventilation issue. Limbeck et al. (2004) observed only a weak seasonal dependence of the Pd concentration in two cities explaining about 20% of the enhanced PM10 levels in the cold season to originate from less ventilation (Klagenfurt and Salzburg).
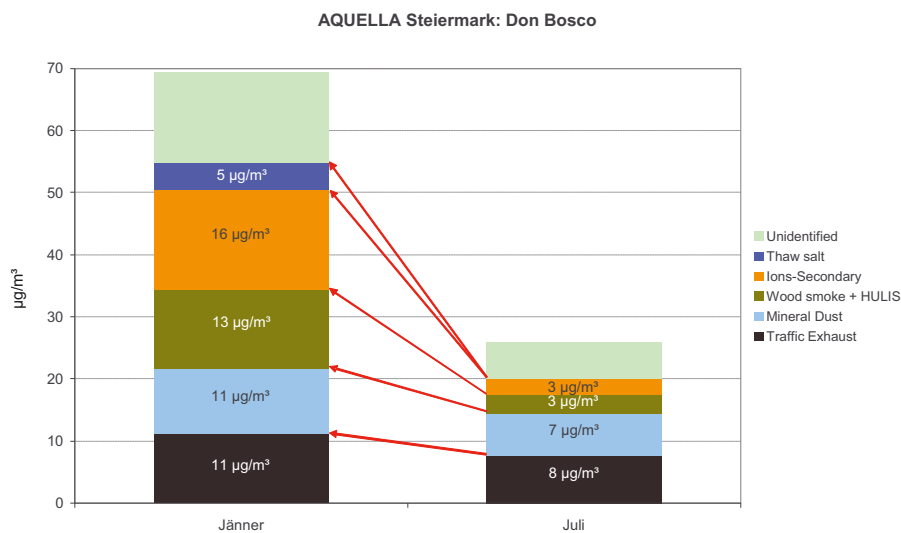
AQUELLA Steiermark: Don Bosco

Figure 3: Comparison of source contributions for January and July 2004 at the urban site Don Bosco, Graz. From Bauer et al. (2007a).

The actual increase from a summer to a winter month was around 100-200%. As the macro-tracer results reveal, this increase is a result of specific "winter" sources. While Diesel-soot increases by the factor of wintry reduced ventilation (20-30%), the highest primary aerosol impact is from wood smoke and from thaw salt. A less discussed effect is the dramatic increase of ammonium nitrate, which is a result of the temperature dependent stability of the salt, with low formation tendency in the warm season, but stable conditions in cold and humid situations. Thus the cold season increase of ammonium nitrate is not an

emission dependent effect, it is a result of a shifted thermodynamic equilibrium. Figure 3 shows the comparison of the aerosol source contributions at the urban site "Don Bosco" in Graz for PM10 for January and July.

For all urban sites studied, four main sources or source types responsible for the elevated PM10 levels in the cold season were identified. The four main sources however show with quite variable contributions at the different sites and during the different exceeding periods:

- Street dust from grinded gravel and thaw salt used for streets under snow/ice conditions ("wintery street conditions"). Main emission take place during long lasting cold and dry periods, frequently occurring in the late winter.

- Ammonium sulfate and nitrate aerosol regional accumulation and transport from areas outside of the city area

- Smoke from wood combustion regional accumulation and transport from areas outside of the city area

- Diesel soot from traffic and off road sources

The AQUELLA experiments were planned as multiple site studies, thus, it was also possible to discriminate the rural concentrations – representing the background to the urban loadings. A surprising result of AQUELLA was that in many cases during pollution episodes a high background concentrations was present, which limited dramatically the additional "urban contribution" of an aerosol concentration which would keep the total PM10 level (background plus "urban increment") below the limiting value. On the average during "exceeding days" (days exceeding the 50 $\mu g/m^3$ threshold for PM10 concentrations at the most exposed site) the background level contributed around two thirds, the urban increment one third to the PM levels measured at the exposed urban sites.

It became evident that in many of the exceeding periods the regional background levels were high due to two main aerosol sources:

- Wood smoke (including secondary material from wood smoke ("HULIS")

- Ammonium nitrate and sulfate, particularly pronounced at the sites north and east of the Alps as a result of long range transport

The dominant impact of wood smoke and inorganic secondary material at AQUELLA background sites is evident from Figure 4.

Schafberg: Vienna   Anthering: Salzburg   Bockberg: Styria/Graz
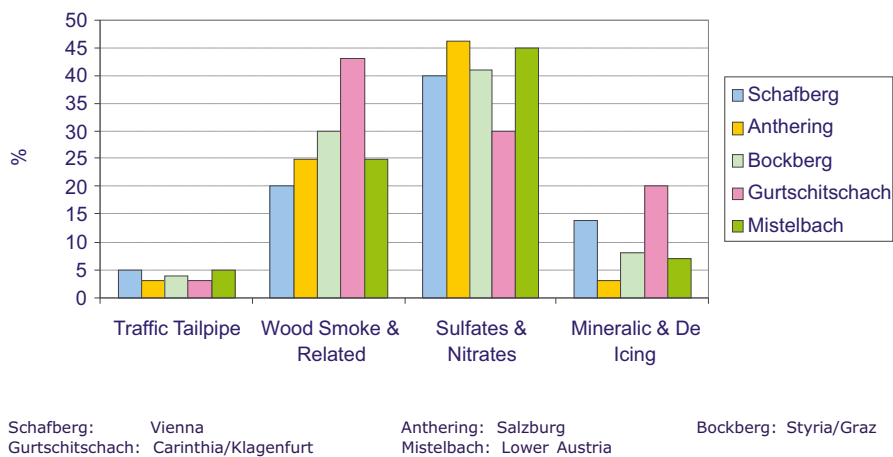Gurtschitschach: Carinthia/Klagenfurt   Mistelbach: Lower Austria

Figure 4: Source contributions at AQUELLA background sites in Vienna, Salzburg, Styria, Carinthia and Lower Austria during "exceeding periods" (Compiled from reports).

However, also at sites south of the main Alpine ridge the impact of secondary inorganic aerosol in the regional background is obvious. The overwhelming impact of the regional background (Lavamünd, Gurtschitschach) on PM10 levels at the urban sites Koschatstrasse and Völkermarkterstrasse (in the city of Klagenfurt) is visualized for a three month period given as averages of "exceeding days" in Figure 5. The Klagenfurt basin is "filled" with a "lake" of wood smoke and inorganic secondary material, extending over a distance of more than 60 km. Within the city mainly mineral dust, carbonates, thaw salt and exhaust emissions increase.

Within a region considerable different types of episodes may occur. An example is given for Graz. In Figure 6 aerosol source contributions are shown for three different episodes, where in one case mineralic material, carbonates and thaw salt dominate the PM10 composition; in the further case the aerosol is dominated by inorganic secondary aerosol (ammonium nitrates and sulfates; Episode from 25-28 Nov. 2004 is and a third case is dominated by wood smoke and traffic exhaust, indicating also a city source for wood smoke.
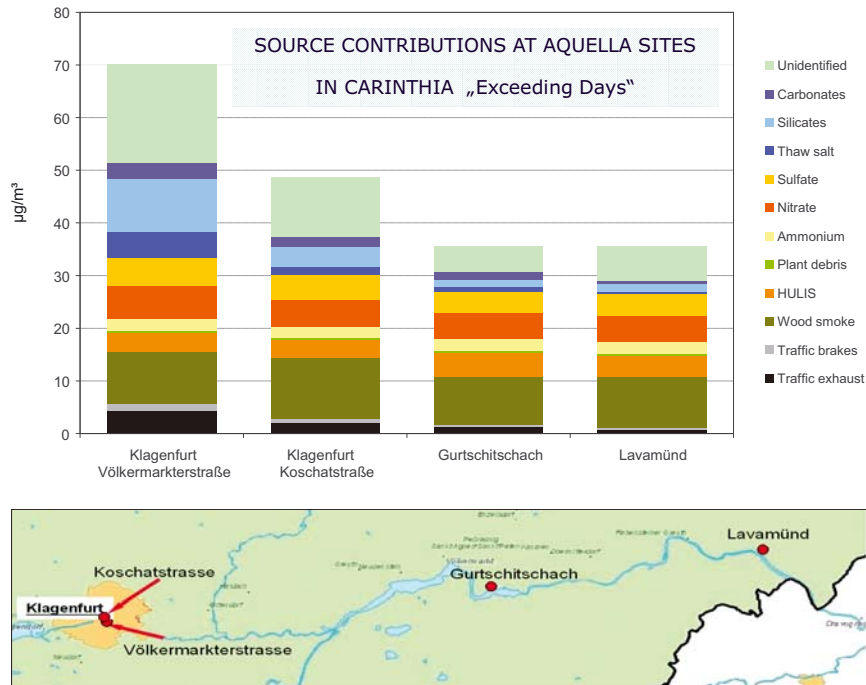
Figure 5: Souce contributions at AQUELLA sites in Carinthia – averages for "exceeding days". (From Bauer et al. 2007b).

As a consequence from the AQUELLA results recommendations were derived for reducing PM10 emissions:

- Reductions of the aerosol source strength of around 30-50% are required in a similar magnitude for the regional background as well as for the urban sites.

- For reducing the regional background reductions have to be considered for NO_x and ammonia in the air quality region, as well as reductions of NH_3, SO_2 and NO_x should proceed in European countries with respective high emissions.

- The use of log wood for combustion in "old" stoves should be restricted during PM10 "alarm" situations, stove/boiler changes be considered. This applies to the countryside as well as to the urban districts.

- High mineral dust and thaw salt aerosol is a result of long lasting cold and dry winter periods. A task force for reducing this dust source is recommended to identify possibilities of less dustier wintry conditions.

- The reduction of exhaust PM and NO_X emissions as a consequence of the Euro5 and Euro6 emission regulations for new cars it is expected to miti-

gate the soot emission of cars and as well as of the NOx emissions, which are currently an important precursor for the wintry ammonium nitrate aerosol.
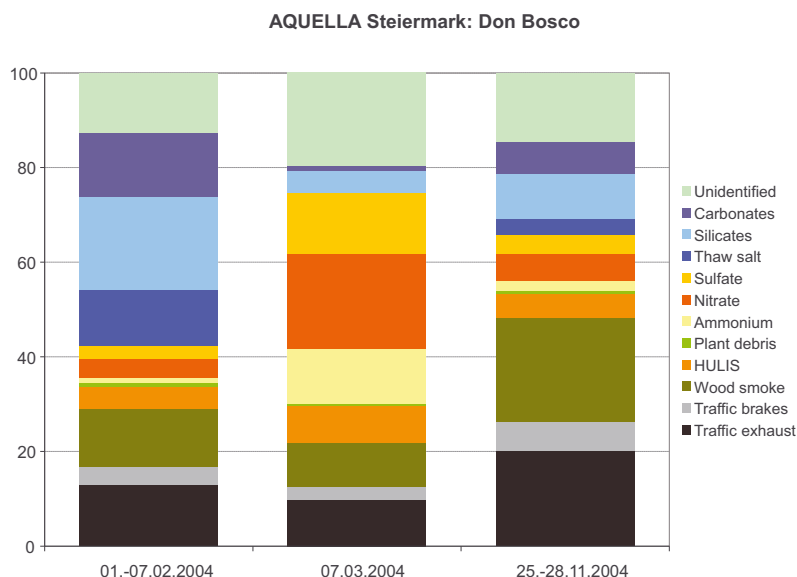
AQUELLA Steiermark: Don Bosco

Figure 6: Different source signatures during three different PM10 "episodes" in the greater area of Graz: Episode from 1-7 Feb. 2004 is dominated by mineralic material, carbonates and thaw salt; Episode from 7 March 2004 is dominated from inorganic secondary aerosol (ammonium nitrates and sulfates); Episode from 25-28 Nov. 2004 is dominated by wood smoke and traffic exhaust.

4. CARBOSOL (Aerosol chemistry and ice core aerosol trends)

The EU funded CARBOSOL project was a joint study of a network of international laboratories to investigate the present and retrospective state of organic versus inorganic aerosol over Europe. The coordinator was Michel Legrand of the LGGE (Laboratoire de Glaciologie et Géophysique de l'Environnement du CNRS, St Martin d'Hères, France).

"In this project we combine the study of spatial distribution and composition of the present-day carbonaceous aerosol in air and wet deposition in western/central Europe with the composition trends of climatically relevant carbonaceous material in Alpine ice cores spanning the last 150 years. The investigations include organic and inorganic constituents, EC/OC, WSOC and WinOC. Source apportionment (natural versus anthropogenic, primary emissions versus secondary production) is performed via selected organic tracers and ^{14}C *analyses. From Alpine ice cores we reconstruct past*

> *atmospheric changes of relevant aerosol components over Europe by establishing air-snow relationships at the glacier sites. For some organic fractions this is the first report about the occurrence in early 20th century. Transport/chemistry models are used to investigate the net radiative effect of the change in aerosol load and composition (inorganic/organic) in Europe from the pre-industrial era to the present. Thus, the inversion of ice core aerosol signal is used in a regional chemical-transport model to estimate the underlying regional climate impact."* (Legrand and Puxbaum 2007)

CARBOSOL ice-cores under investigation were from glacier fields at Monte Rosa and Mt. Blanc. The present day aerosol and precipitation was studied over a time span of two years at a set of 6 sites arranged over a west-east transect from the Azores (Atlantic Ocean) to K-Puszta (Hungary), including Azores (50 m elevation), Aveiro (50 m), Puy de Dome (1450 m), Schauinsland (1205 m), Sonnblick (3106 m) and K-Puszta. A regional circulation model coupled with a radiative transfer model in which aerosols are treated on-line was deployed to reconstruct the past evolution of aerosol emissions, concentrations and radiative forcing in Europe.

The present-day climatology of the aerosol gained during CARBOSOL was compiled by Pio et al. (2007). Carbonaceous aerosol concentrations showed a gradient of more than 20 times between the AZO marine background and the lowland continental sites of AVE and KPZ, with intermediate levels at continental mountain sites.

In a pioneering study source apportionment of the carbonaceous aerosol was achieved by combining measurements of organic macro tracers (levoglucosan for biomass combustion and arabitol for fungal spores, cellulose for plant debris) and a high sensitivity ^{14}C radiocarbon method for interpreting sources of EC and OC (Gelencser et al. 2007). Source types were lumped into primary emissions from fossil fuel combustion and biomass burning, plant-derived bio-aerosol, and secondary productions from precursors emitted by fossil and non-fossil sources. It has been found that while fossil-related sources predominate EC throughout the year at all sites, the sources of OC are mainly biogenic. In winter, biomass burning is the main OC source, with sizable additional contribution from fossil fuel combustion. In contrast, secondary organic aerosol from non-fossil sources becomes predominant in summer (Figure 7). From the levoglucosan data it is shown that biomass smoke is a very important constituent of the organic material in the mid and west European background in winter with contributions to organic matter of ~20% at the elevated mountain sites and 50-70% at polluted continental sites, not including secondary organic aerosol related to biomass combustion sources (Puxbaum et al. 2007).
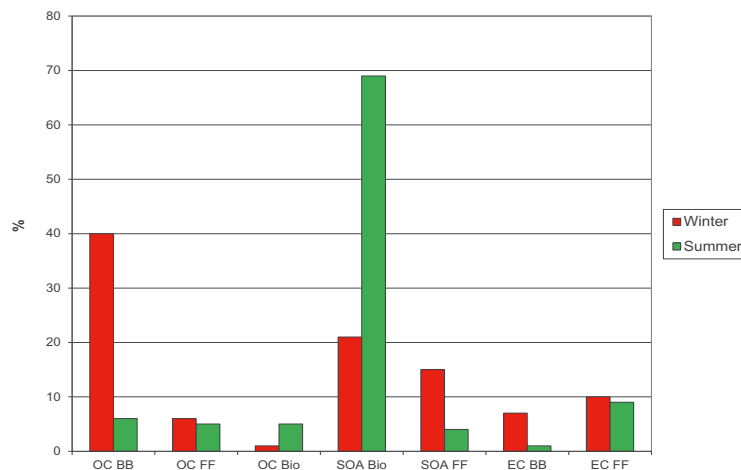
Figure 7: Source apportionment of EC, OC (primary) and SOA (secondary OC) at the site K-Puszta (Background site in Hungary) during the winter and summer season. BB .. biomass burn, Bio .. biogenic particles, FF .. fossil fuel. Compiled from data in Gelencsér et al. (2007).

More than 200 compounds were identified and quantified by GC/MS, including alkanes, polyaromatic hydrocarbons, aldehydes, ketones, alcohols, sterols and organic acids in CARBOSOL aerosol samples. The organic compounds identified were used to evaluate the different sources with geochemical parameters and lipid source assignment. The GC/MS results confirmed the strong impact of biomass burning during winter at lowland sites (Oliveira et al. 2007). The weak amount of OC identified by such organic speciation (only 2-4% of the organic matter) stimulated the study of the oligomeric or polymeric matter. This was done for cellulose which, as expected, reveals higher concentrations in the growing season than in winter at continental surface sites. More surprising is the high relative concentrations of plant debris in organic matter observed at the elevated sites (5% in summer and 6-22% depending on elevation in winter) (Sánchez-Ochoa et al. 2007). Ubiquitous in organic aerosol, HULIS are a still ill-defined subset of organic aerosol mass and in fact no standard method is available for their determination. Using two different methods their climatology was investigated (Lukács et al. 2007, Feczko et al. 2007). Organic C2-C5 diacids were found to be significant contributors to "water soluble organic carbon" (from 3 to 7% at continental sites and up to 14% at Azores in summer) (Legrand et al. 2007a).

Carbonaceous species including filterable elemental and organic fractions, dissolved organic carbon, HULIS, and single organic compounds like carboxylic acids, levoglucosan, and cellulose were investigated together for the first time in high alpine snow deposits (Legrand et al. 2007b). An extended version of the EMEP 3-D chemistry-transport model was developed towards EC and OC and simulations were compared with observations in Europe. In additi-

on to primary emissions from fossil-fuel and wood burning, the model includes secondary production of anthropogenic and biogenic organic aerosol. The EMEP model reproduces well pollutants with well-know emissions and chemistry like sulfate but significantly under-predict EC, especially in winter (Simpson et al. 2007). For OC the performance of the EMEP model was evaluated not only with respect to observed OC but also for the first time against the levoglucosan analysis of Puxbaum et al. (2007) and the source apportionment calculations from Gelencsér et al. (2007). This powerful exercise indicates that underpredicted OC concentrations in winter (by a factor of 3 to 5) mainly results from an underestimated wood burning contribution in the model whereas in summer most of the departure (an underestimation by a factor of 2 to 3) is related to secondary production from both fossil-fuel and non-fossil fuel precursors not accounted for in the model.

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Address of the Author:

ao. Univ.-Prof. Dr. Hans Puxbaum
Institute of Chemical Technologies and Analytics,
Vienna University of Technology
Getreidemarkt 9/164, A-1060 Vienna