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Study of emission episodes of urban aerosols by ion beam analytical techniques

A. Angyal [a,b,*], Zs. Kertész [a], Z. Szikszai [a], Z. Szoboszlai [a,b], E. Furu [a], L. Csedreki [a], L. Daróczi [c]

[a] Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), H-4026 Debrecen, Bem tér 18/c, Hungary
[b] University of Debrecen – ATOMKI, Department of Environmental Physics, H-4001 Debrecen, P.O. Box 51, Hungary
[c] University of Debrecen, Department of Solid State Physics, H-4026 Debrecen, Bem tér 18/b, Hungary

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ABSTRACT

The quantitative elemental composition and morphology of over 500 atmospheric aerosol particles were determined by nuclear microscopy and scanning electron microscopy (SEM). The samples originated from eight sampling campaigns, when hourly variation and sources of the urban aerosol elemental components were studied in Debrecen between 2007 and 2010. Aerosol which could be connected to heavy metal pollution episodes and high aerosol pollution levels deposits were selected for the nuclear microprobe study.

Ion beam analytical methods (micro-PIXE and STIM) provided the elemental composition of coarse (particles with aerodynamic diameter between 2.5 and 10 μm) aerosols while the morphology of the different particle types was determined by SEM.

Through the elemental composition, elemental correlations and morphology different particle types were identified and attributed to different anthropogenic sources like biomass burning, oil combustion, traffic or industry.

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1. Introduction

Aerosol pollution is one of the biggest problems in urban environments because high pollution levels present significant health hazards [1,2]. Thus the characterization of atmospheric aerosol particles and its sources is a relevant task in densely populated areas like Debrecen.

The hourly variation of the elemental components of urban aerosols has been studied as part of the aerosol research program running at the Laboratory of Ion Beam Applications of ATOMKI [3]. During the eight sampling campaigns which were carried out between 2007 and 2010 several emission episodes were detected in both the fine (particles with aerodynamic diameter <2.5 μm) and coarse (particles with aerodynamic diameter between 2.5 and 10 μm) size fractions. Sources of the urban aerosols were also determined based on the elemental composition data obtained from bulk PIXE analysis [4]. However, the origin of some sources and emission episodes remained unclear after the bulk analysis.

In our previous work we have shown that single particle analysis using nuclear microscopy methods provides a useful contribution to the source characterization [5,6]. For example, with the help of a nuclear microprobe study and statistical methods four different sources of Cl-containing coarse particles were determined [6].

In the present work emission episodes and sources related to potentially toxic elements and heavy metals were in the focus. Single particle analysis on selected samples was done at the Scanning Nuclear Microprobe Facility in Debrecen. The samples originated from sampling campaigns carried out with 2-h time resolution from 2007 to 2010, and the selection was made according to the elemental composition obtained from bulk PIXE analysis. The quantitative elemental composition of over 500 particles was determined using PIXE and STIM analytical methods. The morphology of particles was also investigated by scanning electron microscopy (SEM) at the Department of Solid State Physics of the University of Debrecen.

2. Experimental and methods

2.1. Aerosol samples

A two-stage sequential PIXE International streaker [7] is used to study the hourly evolution of atmospheric aerosol concentration. The sampling device consists of a pre-impactor to stop particles with an aerodynamic diameter (D_{ae} in the following) greater than 10 μm, a Nuclepore polycarbonate filter with 0.3 μm pore diameter to collect the fine fraction (D_{ae} < 2.5 μm) and a kapton foil coated with paraffin to gather the coarse fraction (D_{ae} = 2.5–10 μm). Sampling has been done with 2 or 3-h-time resolution every season of the year since October 2007. Seven sampling campaigns have been

* Corresponding author at: Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), H-4026 Debrecen, Bem tér 18/c, Hungary.
E-mail address: angyal@atomki.hu (A. Angyal).

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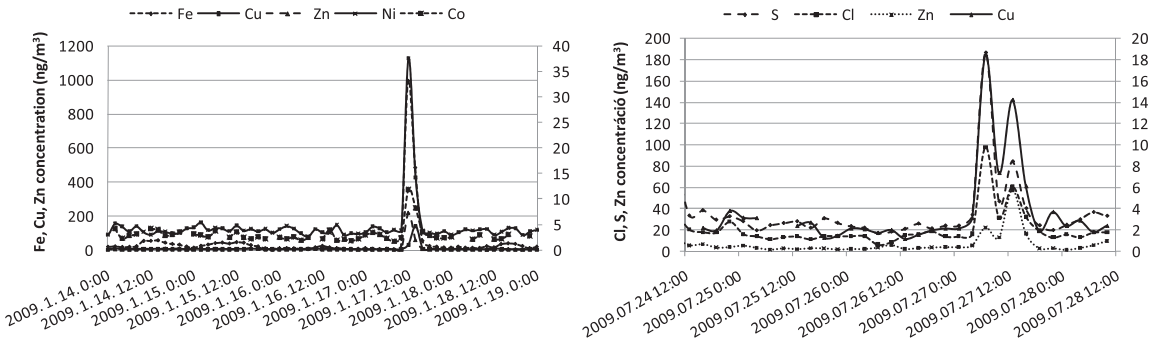

Fig. 1. Fe, Cu, Zn, Ni, Co concentrations between 14–19th January 2009 (left) and S, Cl, Zn, Cu concentrations recorded on 24–28th July 2009 (right) in the coarse fraction.

carried out at an urban background site, in the garden of ATOMKI, which is located close to the centre of Debrecen. The dates of campaigns were the following: 10–19 October 2007, 24–30 January 2008, 15–24 May 2008, 18–27 August, 13–19 January 2009, 22–28 July 2009, 25 August–3 September 2009.

2.2. Nuclear microprobe measurement

As far as the fine fraction is concerned the micro-PIXE technique did not provide additional information about the composition and origin of aerosols, therefore single particle analysis has only been done on the coarse samples. For the nuclear microprobe study 23 coarse samples were selected. These samples were either enriched in heavy metals or originated from an especially high pollution level episode in January 2009 (Fig. 1).

Single particle analysis was carried out at the Debrecen Scanning Nuclear Microprobe installed on the 0° beamline of the 5 MV Van de Graaff accelerator of the IBA laboratory of the Institute of Nuclear Research of the Hungary Academy of Sciences [8–10]. A 1.2 μm × 1.2 μm H^+ beam of 2 MeV energy and 100 pA current was used for the study. In each sample, measurements were done by scanning several 75 × 75 and 100 × 100 μm^2 regions. The accumulated charge was 0.5–0.7 μC and the irradiation time of one scan area was approximately 1–1.5 h. The morphology and sizes of particles were determined by Scanning Transmission Ion Microscopy (STIM). The PIXE–PIXE ion beam analytical technique, based on the simultaneous use of an ultra thin windowed (UTW) and a conventional Be windowed Si(Li) X-ray detector, was used to determine the elemental composition for $Z \geqslant 6$ [5,11]. The X-ray lines with low and medium energy (0.2–9 keV) were identified by the UTW detector and thus the measurement of C–Fe was possible while similarly the medium and high energy X-rays (>2 keV) were distinguished by the Be-windowed detector and thus enabled measurement of the S–U region. The accumulated charge was measured by a Faraday cup placed at 0° behind the sample.

2.3. Data evaluation

Particles were identified according to the different energy maps (STIM) and elemental maps. PIXE spectra, the particle thickness (mean energy loss) and the accumulated charge were extracted manually for each identified particle from the list mode data. Absolute concentration values of C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Br, Sr, Ba, and Pb of over 500 particles were determined using the PIXEKLM [12,13] and PIXEKLM TPI [14] program packages. Elemental maps of a typical particle deposit are shown on Fig. 2. However, the concentration of C was not used for further investigation due to the contribution of the supporting kapton foil.

On-axis STIM with ~500 nm lateral resolution provided adequate information about the particle size, and most of the cases the particle shape could also be determined (spherical or odd-shaped). However, Scanning Electron Microscopy (SEM) proved to be more suitable for detailed morphological characterization of individual particles (shape, agglomerates, external-internal mixing, etc.).

3. Result and discussion

3.1. Emission episodes

The hourly evolution of the elemental components showed several emission episodes while concentration values of one or more elements significantly increased for a few-hour-long period in both size fractions [15]. Some of these events could be related to heavy

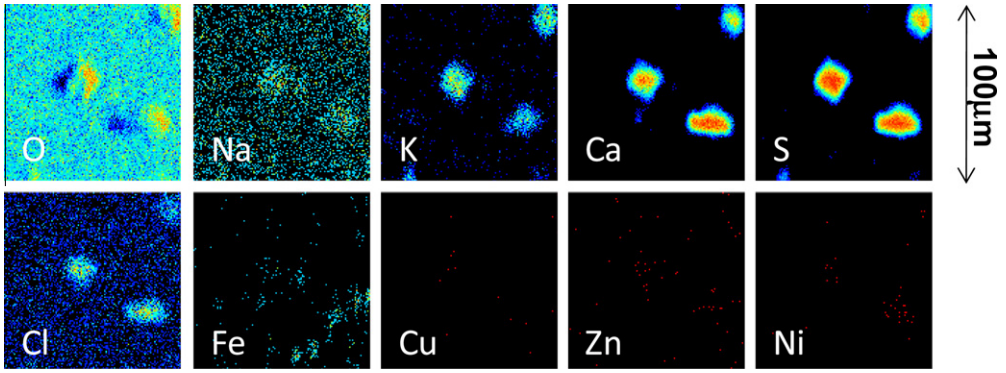

Fig. 2. PIXE elemental maps of individual aerosol particles. Scan size: 100 × 100 μm^2.

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