Nuclear Instruments and Methods in Physics Research B 318 (2014) 125–129

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

PIXE–PIGE analysis of size-segregated aerosol samples from remote areas

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ARTICLE INFO

Article history:
Received 15 March 2013
Received in revised form 7 May 2013
Accepted 27 May 2013
Available online 22 July 2013

Keywords:
PIXE
PIGE
Sodium
Atmospheric aerosol
Size segregated sample
Cascade impactor

ABSTRACT

The chemical characterization of size-segregated samples is helpful to study the aerosol effects on both human health and environment. The sampling with multi-stage cascade impactors (e.g., Small Deposit area Impactor, SDI) produces inhomogeneous samples, with a multi-spot geometry and a non-negligible particle stratification.

At LABEC (Laboratory of nuclear techniques for the Environment and the Cultural Heritage), an external beam line is fully dedicated to PIXE–PIGE analysis of aerosol samples. PIGE is routinely used as a sidekick of PIXE to correct the underestimation of PIXE in quantifying the concentration of the lightest detectable elements, like Na or Al, due to X-ray absorption inside the individual aerosol particles. In this work PIGE has been used to study proper attenuation correction factors for SDI samples: relevant attenuation effects have been observed also for stages collecting smaller particles, and consequent implications on the retrieved aerosol modal structure have been evidenced.

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1. Introduction

Atmospheric aerosols have important effects on human health and natural environment. Many properties of aerosol particles play a role in determining the nature and the relevance of the aforementioned effects; in particular, chemical composition and particle dimensions are crucial aspects. In fact, as concerns the effects on human health, smaller particles have more relevant effects due to their potential to get deeper into the breathing apparatus and from there into the circulating apparatus; particles containing harmful chemical components are indeed dangerous for human beings. Further, both chemical composition and particle dimensions influence the interaction between particles and solar/terrestrial radiation, thus affecting the Earth radiation budget. Moreover, the study of the chemical composition of aerosols as a function of particle dimensions can help in the identification of the main aerosol sources and can give information on the atmospheric processes occurring during atmospheric transportation. In fact, atmospheric aerosols display multi-modal distributions of particle mass by particle size, which may vary significantly depending on location, atmospheric conditions and aerosol sources.

The collection of aerosol in different samples depending on the particle dimensions (size-segregated samples) is generally achieved with cascade impactors: particles impact on different stages according to their aerodynamic properties (with the particle size decreasing getting deeper into the impactor); the cut-off diameter of a stage is defined as the one corresponding to particles therein collected with a 50% efficiency. For this work, a Small Deposit area Impactor (SDI, [1]) by Dekati Ltd has been used.

Analytical techniques allowing quantitative and fast measurements of a wide range of elements, characteristic of the different sources, are valuable for the study of aerosol. In particular, Particle Induced X-ray Emission (PIXE) technique is excellently applied in this research field due to its capability to detect with high efficiency in a single measurement all the elements with $Z > 10$. Furthermore, PIXE is an unrivaled technique for the study of the dust component of aerosol, due to its high sensitivity to crustal markers (in particular Si, which is in principle detectable with chemical techniques such as Inductively Coupled Plasma – Atomic Emission Spectroscopy, ICP-AES, but whose quantification might be poor due to the volatility of SiH_6, formed under the HF attack of the silicon matrix). In remote areas, not influenced by local pollution sources, the assessment of the impact of long-range transports of dust, sea-salt and anthropogenic pollutants is fundamental, hence giving a prominent role in this field to PIXE. Nevertheless, the quantification of light elements (namely, Na, Mg, Al and Si) may give underestimated values due to the

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0168-583X/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2013.05.097

absorption of the less energetic X-rays inside the single aerosol particle and/or inside the deposition spot (when particle stratification is not negligible).

Correction factors for the attenuation in (non-size-segregated) aerosol samples cannot be accurately calculated since these samples contain particles with different composition and with an "a priori" unknown distribution of sizes [2,3]. In size-segregated samples, every single stage sample contains particles in a defined size class indeed, but particle stratification further limits the possibility to calculate correction factors on the basis of simple assumptions on the matrix composition. Some authors have already discussed the issue of matrix corrections for samples collected with different types of cascade impactors characterized by non-uniform "thick" deposits [4–6], suggesting some approaches for calculating the X-ray attenuation based on the use of different codes and methods for the evaluation of the deposit thickness. Nevertheless, such approaches are limited by high uncertainties on the obtained correction factors (due to assumptions on the matrix composition and on the schematization of the deposit geometrical structure) and by long calculation times of the procedure itself.

As concerns SDI samples, they have a multi-spot geometry, determined by the pattern of the different and closely clustered flow jets (see Fig. 1): on some stages, despite the small size of the collected particles, relevant correction factors for X-ray attenuation may be expected as the aerosol is concentrated in small spots where particles pile up.

At the INFN-LABEC laboratory, PIGE (Particle Induced Gamma-ray Emission) with proton beams has been advantageously used as a complementary technique to PIXE for the analysis of light elements in many application fields (e.g., atmospheric aerosol [3], sediments [7], cultural heritage [8]). In fact, PIGE does not suffer from the attenuation problem thanks to the higher energy of the detected radiation, consisting in prompt γ-rays emitted from (p,γ), (p,p'γ) and (p,$\alpha\gamma$) reactions.

This paper is focused on the assessment of proper matrix correction factors for Na, the lightest element we detect, by simultaneous PIXE–PIGE measurements of the size-segregated samples collected with a SDI in Ny Ålesund (78.6°N, 11.6°E, Svalbard Islands) in the framework of a campaign for the study of the arctic aerosol.

2. Methods

2.1. Sampling site and aerosol sampling

Aerosol sampling was performed at the Gruvebadet laboratory, a recently installed station for the research on Arctic atmospheric aerosol. It is located in the international research center of Ny Ålesund, on a western fjord of Svalbard Islands; these islands are an ideal site for the study of the interaction between the climate change and the atmosphere, ocean and land variations, due to their geographical position, coinciding with the northernmost point influenced by the warm West Spitsbergen Current. At Ny Ålesund, long time series of environmental data have been collected, thus enhancing the understanding of such phenomena on a long-term base; concerning the size-segregated aerosol samples, aerosol samplings with a cascade impactor (whose samples were also analysed by PIXE) were performed there already two decades ago [9]. At Gruvebadet, the joint effort of many research institutions has allowed the installation of a wide set of instruments for the study of different aerosol properties, as number and aerosol mass size distributions, chemical composition (accounting for a number of chemical species) and optical properties such as light absorption (by a Particle Soot Absorption Photometer, PSAP) and scattering (by a Nephelometer).

In this framework, we performed samplings of size-segregated samples with a SDI impactor, with a 4-days resolution (in order to collect a significant number of particles for the following analysis). The SDI impactor allows the collection of aerosol particles on 12 different stages, characterized by the following cut-off aerodynamic diameters: 8.57, 4.12, 2.70, 1.68, 1.07, 0.90, 0.60, 0.35, 0.23, 0.15, 0.09, 0.05 μm (stages 12 to 1). With SDI, aerosol particles are collected in multi-spot, point-like deposits enclosed in an 8 mm diameter area on impaction foils; the pattern of the deposit and the number of spots where particles accumulate are different from stage to stage, depending on the number of air jets and on the stage geometry. Kapton (and, to a lesser extent, Nuclepore) foils were used for aerosol particles collection during the 2010 and 2011 sampling campaigns. During the two campaigns, a total amount of 81 samplings was performed. This paper focuses on the methodological work performed on 11 samplings (corresponding to 132 single stage samples) fully analyzed by PIXE and PIGE measurements.

2.2. PIXE–PIGE measurements

PIXE–PIGE measurements were performed at the 3MV Tandetron accelerator of the INFN-LABEC laboratory in Florence (Italy), using the therein installed external beam line fully dedicated to PIXE–PIGE analysis of aerosol samples; the set-up is extensively described elsewhere [10]. Briefly, the proton beam is extracted in air through a 500 nm thick Si_3N_4 window; aerosol samples are positioned at a distance of about 1 cm from the extraction window, with the volume of atmosphere between them saturated with helium, to reduce the absorption of the emitted radiation. A collimator located in the end of the in-vacuum beam line sets the beam size (usually to 1.0×2.0 mm^2). The use of two X-ray detectors (Silicon Drift Detectors, SDDs) optimized for low and medium–high X-ray energies, respectively, allows an effective simultaneous detection of all the elements of interest. The set-up includes a 60×23 mm Ge detector for γ -rays detection for PIGE analysis.

As previously mentioned, SDI samples present a non-uniform deposit, with many point-like spots enclosed in 8 mm diameter

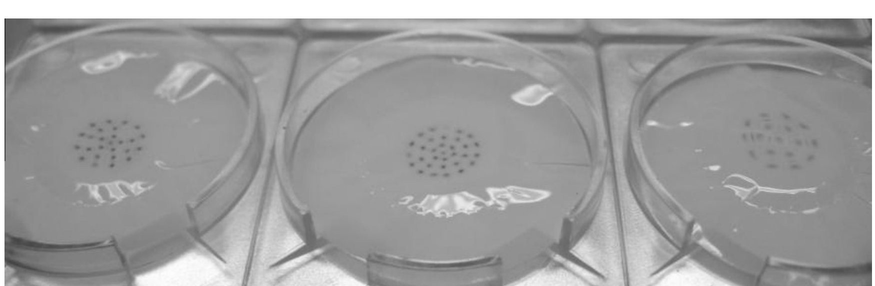

Fig. 1. Examples of samples collected with the SDI impactor (namely, stages 4–6).

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