Abstract

In the scope of this thesis, multi-elemental quantitative data of coarse (PM_{10}) and fine ($PM_{2.5}$) atmospheric particulate matter (PM) samples originating from Ulaanbaatar, Mongolia, including the independent measurement of black carbon concentration have been utilized so far to support source apportionment using statistical tools such as the Positive Matrix Factorization (PMF) model. In addition, the speciation of two element of interest (Cr and Zn) was investigated by the synchrotron induced X-ray absorption method.

In the first set of aims, secondary target excitation X-Ray Fluorescence (XRF) technique with an optimized hardware design was used for elemental characterization on a large data set of 184 samples each of fine and coarse fractions collected over a three-year period (2014–2016). PMF was applied using the concentrations of 16 elements measured by an energy-dispersive XRF spectrometer along with the black carbon content measured by a light reflectometer as input data. The results revealed that whereas mixed sources dominate the coarse fraction, soil and traffic sources are the principle contributors to the fine fraction. The source profiles and the seasonal variations of their contributions indicate that fly ash emanating from coal combustion mixes with traffic emissions and re-suspended soil, resulting in variable chemical source profiles. Additionally, the probable source contributions from long-range transport events were assessed via concentration-weighted trajectory analysis.

In the second part of work, particle induced X-ray emission (PIXE) technique was applied to demonstrate and compare the analytical capabilities, strengths and weaknesses of both the PIXE and XRF techniques on size-fractioned PM. The PIXE measurements were carried out at a 3 MeV Tandem accelerator. Elemental sensitivities were experimentally determined using thin-film, single-element targets, whereas the accuracy in the quantification procedure was evaluated using the NIST standard SRM2783. A critical comparison of the merits and drawbacks of each technique is presented with respect to analytical range, precision, accuracy and detection limits obtained.

Finally, X-ray absorption near edge spectroscopy (XANES) was carried out on size-fractioned PM successfully for the K-edges of Zn and Cr at Elettra Sincrotrone Trieste. Measurements were focused on gaining information about chemical speciation of these two elements. The spectroscopic results identified trivalent chromium sulfate [$Cr_2(SO_4)_3$] and chromium oxide (Cr_2O_3) compounds as the major chemical forms of Cr in both the coarse and the fine PM fraction. Furthermore, both fractions contained abundances of Zn sulfate ($ZnSO_4$) and silicate (Zn_2SiO_4) compounds, but only the fine mode contained Zn oxalate (ZnC_2O_4), whereas only the coarse mode contained Zn chloride ($ZnCl_2$). These Cr and Zn species seemed to originate from local anthropogenic sources, e.g., combustion products or traffic-related re-suspended road dust.