SUMMARY OF PHD THESIS

DEVELOPMENT OF A PHOTOACOUSTIC INSTRUMENT FOR SPECTRAL CHARACTERISATION OF ATMOSPHERIC AEROSOL

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1. INTRODUCTION

Aerosol light absorption plays an important role in the earth's atmosphere in terms of direct and semi-direct radiative forcing, visibility impairment as well as on various and adverse health effects. Although the absorption spectra of the atmospheric bulk is not a conservative indicator of the chemical composition, recently discovered links between the chemical and absorption features of the major constituent of the ambient open up a new perspective for the reliable on-line source apportionment. The importance of the on-line source apportionment is also significant because the toxicity factor of aerosol is strongly depends on its chemical composition. Moreover, the EU also forces the on-line (selective) source apportionment through its regulations (including decree of 2008/50/EK). The aim is the localization of the major anthropogenic emission sources. Despite of its importance, the absorption spectra of even the major anthropogenic constituents are very poorly characterised partly due to the recently discovered BrC fraction having various and versatile spectral feature

and origin, and partly due to the lack of the proper instrument. Although photoacoustic spectroscopy is the only measurement technique which can measure light absorption by aerosol directly without any measurement artefact associated to both the DM and IPT techniques, its application is not widespread yet. This can be explained by several reasons including the lack of well established and reliable calibration method, as well as proper light sources.

2. Objectives and methods

The aim of my work was to develop a photoacoustic instrument for spectral characterisation of atmospheric aerosol in the UV-VIS-NIR spectral region. A special light source and a wavelength independent calibration procedure were created for this purpose.

My aim was to construct a photoacoustic instrument for monitoring light absorption by aerosol in the UV-VIS-NIR spectral range and to prove the practical applicability of the instrument both under laboratory and under field conditions.

The developed instrument is based on the successful combination of a novel light source and photoacoustic detection scheme. A light source based on an Nd:YAG disc laser and its frequency converted higher harmonics has also been developed in the frame of this work. Furthermore, a wavelength independent calibration procedure has been evolved and implemented into the

instrument. I have determined the characteristic performances and demonstrated the applicability of the presented instrument for spectral characterisation of the atmospheric aerosol both under laboratory and under field conditions. In this work I presented the first UV photoacoustic measurement of atmospheric aerosol both under laboratory and field circumstances.

3. New scientific results

1 I have developed and optimized a novel light source based on an air cooled Nd:YAG and its frequency converted higher harmonics for photoacoustic aerosol measurements. This light source has been built into a photoacoustic aerosol measuring system, which has been thoroughly characterised for aerosol light absorption measurements. Applying independent reference method I have proved that the characteristic performances of the instrument are suitable for light absorption measurements under laboratory circumstances.

2 I have developed a wavelength independent calibration procedure based on gas-phase absorption. The gas-phase calibration protocol has been proved to be adequate to accurate measurement of aerosol light absorption

3 Spectral properties of laser generated coal aerosols with different origin/chemical composition have been analyzed by the presented instrument. It has been proven that the absorption spectrum of the investigated

coal sample is characteristic for its chemical composition. Moreover, I have demonstrated that the identification of the chemical composition through the absorption spectra of the aerosol is more efficient in the UV wavelength region.

4 A compact version of the presented photoacoustic instrument has been built for field measurements. The field-deployable instrument is free from gas-phase cross sensitivity. The characteristic performances of the instrument allow reliable ambient aerosol absorption measurements under climate relevant conditions (above 10 Mm^{-1} and 100ng/m^3 EBC).

5 I have tested the instrument for ambient aerosol measurement under various meteorological conditions at different places. As a result, I have demonstrated that accurate spectral characterisation of ambient aerosol is feasible by the instrument. Moreover, comparative measurements with an Aethalometer have proved that the estimation of the UV absorption from measurement data originated from the VIS spectral region can lead to measurement errors in certain cases. Corrections of such

estimation strongly depend both on the Ångström exponent and on the applied wavelengths.

4. Publications

My results are based on the following publications:

[1] Tibor, Ajtai, Ágnes Filep, Martin Schnaiter, Claudia Linke, MarlenVragel, Zoltán Bozóki, Gábor Szabó, Thomas Leisner: „A novel multi-wavelength photoacoustic spectrometer for the measurement of the UV–vis-NIR spectral absorption coefficient of atmospheric aerosols” Journal of Aerosol Sciences DOI:10.1016/j.jaerosci.2010.07.008. IF: 2,192

[2] Tibor Ajtai, Ágnes Filep, Gabriella Kecskeméti, Béla Hopp, Zoltán Bozóki, Gábor Szabó: „Wavelength dependent mass-specific optical absorption coefficients of laser generated coal aerosol determinedfrom multi-wavelength photoacoustic measurement”Appl. Phys. A. DOI 10.1007/s00339-010-6068-3. IF: 1,760

[3] T. Ajtai, Ágnes Filep, Noémi Utry, Martin Schnaiter, Claudia linke, Zoltán Bozóki, Gábor Szabó, Thomas Leisner. Inter-comparison of optical absorption coefficient of atmospheric aerosols determined by a multi-wavelength photoacoustic spectrometer and an Aethalometer under sub urban wintry conditions. Journal of Aerosol Science; 42 (2011) 859-866. IF: 2,192

Further publications:

[4] J H Miller, Yury A Bakhirkin, **T Ajtai**, Frank K Tittel, C J Hill, R Q Yang, "Detection Of Formaldehyde Using Off-Axis Integrated Cavity Output Spectroscopy With An Interband Cascade Laser." *Appl. Phys. B* **85**: 391-396 (2006).

[5] Zoltán Filus, **Tibor Ajta**i, Zoltán L. Horváth, Zoltán Bozóki, Gábor Pap, Tibor Nagy, Tamás Katona, Gábor Szabó: A novel apparatus based on a photoacoustic gas detection system for measuring permeation parameters of polymer samples.*Polymer Testing*, **26** (2007) 606-613. IF 1.357

[6] T. Ajtai, Á. Filep, A. Varga, G. Motika, Z. Bozóki and G. Szabó: „Ozone concentration monitoring photoacoustic system based on a frequency quadrupled Nd:YAG laser" Applied Physics B: Lasers and Optics DOI: 10.1007/s00340-010-4174-8. IF: 2.239