Atmos. Meas. Tech. Discuss., 7, C4841–C4846, 2015
www.atmos-meas-tech-discuss.net/7/C4841/2015/

Atmospheric Measurement Techniques
Discussions

Interactive comment on "A novel inlet system for on-line chemical analysis of semi-volatile submicron particulate matter" *by* P. Eichler et al.

P. Eichler et al.

armin.wisthaler@uibk.ac.at

Received and published: 8 February 2015

We thank the reviewer for his thorough review and his valuable comments. Our specific responses are detailed below.

Reviewer: My principal criticism of the manuscript is that, given its technical focus, insufficient details are presented to allow the interested reader to fully understand and evaluate the design of the inlet. Figure 1 presents a flow-chart representation of the inlet, which adds little to the textual description, when what would be truly useful are technical drawings of the inlet design and the experimental arrangement (e.g. aerosol vs gas sampling). I would encourage the authors to replace Figure 1 (or add an additional figure if they feel the current Figure 1 merits inclusion) with schematics/technical

drawings and include dimensional information to bring the level of the physical description of the inlet into line with the technical description of the performance evaluations performed.

Response: This is a valid argument which we have addressed as follows: i) As suggested by the reviewer, Figure 1 was replaced with a more technical depiction (see below), ii) A Supplementary Figure S1 has been added which shows the detailed experimental arrangement (particle sampling, gas sampling, zeroing).

Reviewer: The abstract feels a little weighted toward the inlet+PTR-ToF-MS instead of focused on the inlet that is the topic of the manuscript. Some of the inlet performance details should be included in the abstract: flow rate, enrichment, gas-phase removal efficiency, etc.

Response: We have modified the abstract as suggested by the reviewer. Key performance parameters (gas-phase removal efficiency, particle transmission efficiency, enrichment fact, limit of detection) have been included. We do, however, not report technical details (such as flow rate) in the abstract. The abstract now reads as follows: "We herein present a novel modular inlet system designed to be coupled to low-pressure gas analyzers for on-line chemical characterization of semi-volatile submicron particles. The "Chemical analysis of aerosol on-line" (CHARON) inlet consists of a gas-phase denuder for stripping off gas-phase analytes, an aerodynamic lens for particle collimation combined with an inertial sampler for the particle-enriched flow, and a thermodesorption unit for particle volatilization prior to chemical analysis. The denuder was measured to remove gas-phase organics with an efficiency >99.999% and to transmit particles in the 100-to-750 nm size range with a 75-to-90% efficiency. The measured average particle enrichment factor in the subsampling flow from the aerodynamic lens was 25.6 which is a factor of 3 lower than the calculated theoretical optimum. We coupled the CHARON inlet to a proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS) which quantitatively detects most organic analytes and ammonia. The combined CHARON-PTR-ToF-MS set-up is thus capable of measuring both the organic and the ammonium fraction in submicron particles in real-time. Individual organic compounds can be detected down to levels of 10-to-20 ng/m3. Two proof-of-principle studies were carried out for demonstrating the analytical power of this new instrumental set-up: i) oxygenated organics and their partitioning between the gas and the particulate phase were observed from the reaction of limonene with ozone and ii) nicotine was detected in cigarette smoke particles demonstrating that selected organic target compounds can be measured in submicron particles in real-time."

Reviewer - P10110, L1: The authors present more than the concept of the inlet, but an evaluation of a constructed inlet suggest rewording to remove “concept”. “Novel” may be a bit strong given the last decade of aerosol inlet development. The new feature is the use of the aerodynamic lens to produce an enhancement in the sampled aerosol mass. The denuder and thermo-desorption (evaporation?) have been used in concert before (e.g. Rollins et al., ES&T, 2010 among others cited by the authors) and an enhancement technique (mVACES) is reported in Vogel et al. and others.

Response: We agree that the term “concept” should not be used. We do, however, think that the use of the term “novel” is justified. The combined use of a GPD, an ADL with inertial sampler, and a TD is novel. An ADL that is operated at an outlet pressure of a few mbar and that can be coupled to gas analyzers operating in this pressure regime is also novel.

Reviewer - P10110, L9: The sentence “The combined set-up. . .” seems limited since experiments with organics are reported as well.

Response: The phrasing used in the manuscript may indeed lead to a misinterpretation and has thus been changed: “The combined CHARON-PTR-ToF-MS set-up is thus capable of measuring both the organic and the ammonium fraction in submicron particles in real-time.”

Reviewer - P10114, L1 (P10113, L1): The statement that the individual compound mixing ratios were varied was initially a little confusing. The dynamic dilution of the

mixture resulted in the mixing ratios of individual compounds ranging from 0 to 30 ppb (and of the total organic loading from 0 – 11*30=330 ppb).

Response: We have rephrased the paragraph to avoid any potential confusion:“To test the VOC removal efficiency, the GPD was challenged with a certified gas mixture (Apel Riemer Environmental Inc., Broomfield, USA) containing a set of 11 pure and oxygenated hydrocarbons in equimolar concentrations (methanol, acetonitrile, acetaldehyde, acetone, isoprene, methylethylketone, benzene, toluene, xylene, 1,3,5-trimethylbenzene, α-pinene). Dynamic dilution of the mixture with catalytically (Pt/Pd at 325°C) cleaned laboratory air resulted in mixing ratios of individual compounds in the 0-to-30 ppbV range. The total organic load onto the GPD thus ranged from 0 to 330 ppbV.”

Reviewer - P10114, L20: The aerodynamic lens by itself does not produce an enrichment in aerosol concentration, rather it is the combination of the lens to produce a collimated aerosol jet along with what is effectively a virtual impactor that produces the enrichment in the minor flow.

Response: The reviewer is right and we have changed our wording: “an aerodynamic lens (ADL) for particle collimation which is combined with an inertial sampler for the emanating particle-enriched flow”

Reviewer - P10118, L8: I am not sure what “mainstream” cigarette smoke is. Is this a technical term?

Response: Mainstream smoke emanates from the filter end of the cigarette whereas sidestream smoke has its source at the burning cigarette tip. Given that the sampling procedure is described in the text, we are no longer using this term.

Reviewer: It would be nice to have a summative discussion of the estimated enrichment of aerosol organics (combining all of the factors discussed individually) and how that might relate to measurement of compounds in ambient aerosol. There is a brief mention in section 3.3, but a more complete description of how the inlet would improve the analytical detection limit (e.g. of the PTR-ToF-MS) for aerosol organic compounds would be useful. This quantity could also be stated in the abstract. The authors could also include the improvement MDL/MQL expected from further inlet improvements

Response: We agree that such a discussion would be valuable and useful. Since the submission of the manuscript we have tested "improved" configurations on different PTR-ToF-MS instruments and these tests have shown that it is very difficult to predict the performance of modified systems. Also, the CHARON has not yet been tested for ambient aerosol. We thus prefer to strictly limit our description to what has actually been measured rather than giving an outlook to what is very difficult to predict at the moment. The detection limit for individual organic species of the current set-up is now mentioned both in the abstract and in the main body of the text.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/7/C4841/2015/amtd-7-C4841-2015-supplement.pdf

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 10109, 2014.

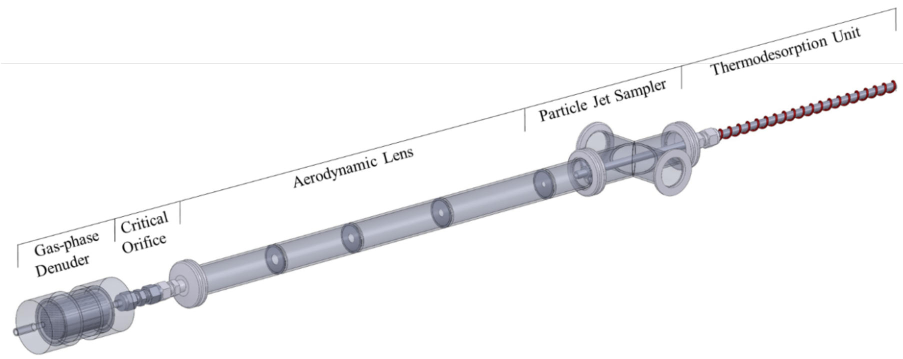

Fig. 1.