

Response to Referee #2

The authors would like to thank Referee #2 for his/her detailed and valuable comments to further improve and clarify the MS. We have considered all recommendations, and made the appropriate alterations. The changes can be explicitly tracked in the annotated version of the MS. Our specific responses to the comments are as follows.

Major comment

Although authors showed the data, it was not clear to me how the data of the present study could be compared with former studies at other urban areas. If I understand it correctly, the uniqueness and novelty of the study is that the authors have conducted an atmospheric observation at Budapest in winter. So, it would be important to compare if the data at Budapest is similar to/different from those in other cities.

We revised thoroughly and improved the Results and discussion section at several places and from several aspects with more detailed arguments. We also modified the body text to make our intentions and statements more detailed, specific, and further literature sources were also included to support our conclusions.

Specific comments

Title: the title of the manuscript could be modified so that it contains more detailed information (e.g., adding information about the observation site).

The hygroscopic and volatile properties of particles in winter and the results/conclusions obtained from them contribute to the improved understanding of the urban-type atmospheric environment and not just of a specific city. The present title emphasises the value of the VH-TDMA measurements in cities in general. This was our motivation for the present formulation of the title. The details of the study including the observation site are given exactly already in the abstract, and further in body text.

Abstract: The current abstract is too long (more than 450 words). If I remember correctly, this journal does not have any length limit for abstract. However, I believe that the abstract could be shortened by almost 50% if it is described concisely.

The abstract was revised and restructured substantially, and it was also shorten as requested.

HTDMA measurements: The authors state that the RH stability of the measurement was $90 \pm 2\%$ (standard deviation). This value is not so small, compared with other HTDMA systems (e.g., [Duplissy et al., 2009]). It would be ideal to have a description on how the fluctuation in RH could influence interpretation of observation data.

As a result of this comment, we realised by double checking that the standard deviation of the mean RH and mean denuder T were given in an incorrect way as a result of faulty data handling (as the maximal deviations). The correct values were given. They comply with the ordinary uncertainty range described in Duplissy et al., 2009.

Application of TDMA_{inv} on VTDMA data: I understand that the method was developed mainly for analysis of HTDMA data. Unlike HTDMA, a part of particles passing through DMA1 completely evaporate (i.e., disappear) by heat during thermal desorption process [Kuwata and Kondo, 2008]. I wonder how this type of particles was considered during the data inversion process.

The volatility properties of particles with a diameter of 20 nm were not interpreted because they appeared at the lower end of the VGF range after the shrinkage, and thus their diameters were close to the detection limit of the CPC used as the detector, and they were also subjected to enhanced diffusional losses. The complete evaporation of particles with a diameter >50 nm was not considered in this study. A comment on this, and the estimation of the magnitude of the loss was briefly discussed.

VFR: Although VFR is frequently used for volatility study on bulk aerosol particles, I am not sure if it is commonly used for VTDMA study. Would it be possible to explain why this metric is useful in obtaining a physically meaningful parameter?

The two terms, namely the volatility growth factor and the volume fraction remaining have related meanings. The former quantity shows the diameter change, while the latter property represents a very expressive picture on the physical appearance of the coating and core of particles, and on their volume ratio. Therefore, we would like to keep this in the MS as well as an auxiliary property.

P8L4: ‘while their coating with succinic acid, sulphuric acid or polyaromatic hydrocarbons (PAHs) influenced the hygroscopic growth in a complex way.’ PAHs are not hygroscopic at all. Would it be possible to clarify how they could influence hygroscopic growth?

According to the cited reference, “PAHs are slightly soluble or even insoluble in water and the solubility decreases with increasing molecular mass. The comparison of the mass spectra of untreated soot and sulfuric acid coated soot showed that the fraction of mass peaks with $m/z > 150$ dropped and the fraction with $m/z < 150$ increased. Probably the sulfuric acid reacts

with the PAHs and forms products with lower molecular mass than the initial PAHs. (...) Reactions of sulfuric acid with PAH have been previously observed. (...) These products could have a higher solubility in water than the initial PAHs. As a consequence, the hygroscopic growth and activated fraction increased due to (a) the products of this reaction and (b) the unconsumed coating fraction itself.” We appreciate the comment, and added this specific information as a further explanation and clarification.

P9L23: ‘Since the rush hours also coincided with the sunrise and sunset in winter’ Is there any supporting evidence for this statement?

The sentence was removed.

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14-02-2018