Review of thesis by Nicolas Talbot entitled "A detailed study on aerosol particle size distribution in indoor and outdoor environments with attention to ammonium nitrate transformation"

The thesis deals with ammonium nitrate volatility/hygroscopicity in atmospheric size-segregated aerosol particles with respect to gradients of gaseous aerosol component - water vapour, and ambient air temperature. The gradients of temperature and humidity are frequently developed between indoor and outdoor environments. Therefore some changes in aerosol particle size distribution, occurring during outdoor/indoor aerosol penetration, can be apportioned to particulate ammonium nitrate behaviour. Careful study and precise quantification of such influence is an important not only for explanation indoor/outdoor size distribution changes but also for reference method of aerosol mass determination.

Regarding the formal requirements of a thesis, author must clearly specify his own contribution to overall work presented in the thesis. This statement is an indispensable part of the thesis and has to be added to the text of thesis.

As far the thesis composition, it is composed of introductory Chapters 1-7, Chapters 8-12 presenting full text of five articles, and Chapters 13 and 14 being Conclusion and prospective to Future work respectively.

Since, of total number five manuscripts, two manuscripts were accepted for publication (No.3 and 5), one accepted for review (No.4), one submitted to journal (No2.) and all had already went through quality demanding editorial process, I focused my attention to Chapters 1-7 and 13, 14 of the thesis and manuscript No 1., which was not sent to any journal at time of review.

Concerning Chapter 1, atmospheric aerosol definition omitting gaseous phase is misleading particularly with respect to the scope of the thesis, aerosol phase changes. Nevertheless, it is peculiar why correct aerosol definition, in subchapter Terminology at page 5, was not used in Chapter 1 as well. There were also incomplete literature citations (EEA p.26, Bai p.41, Martin p.42, Gehin p.48, Nazaroff, Smolík p.50).

Concerning the Article 1, starting on p.62, a diagram depicting the thermodynamically favorable phases of ammonium nitrate under variable T and RH, i.e. phase diagram, will be very useful. Also, information on ultrapure water quality (conductivity?) and ammonium nitrate purity is missing. Such information could be crucial for experiments performed, since ultrapure water are frequently contaminated by organic complexes which may, positioning at the phase interface, may significantly influence inter-phase mass transfer. In Figure 2 showing the shrinking rate vs. residence time, information is missing on standard deviation of the shrinking rate measurements.

Questions:

1. What type of dryer was used in experimental apparatus, schematics on Fig 1?
2. Did you measure or estimate temperature gradient within the reactor?
3. What was the standard deviation and number of repetition of the shrinking rate measurements?
4. What is the leading process governing aerosol particle shrinkage and why?
5. CPC3775 measures down to 4nm particles. Why the number size distributions (Fig. 3) end at about 15 nm, and why there is linear scale at X axis while log scale will be better to view ~ 20nm peak?
6. Are the number size distributions (Fig 3) corrected for diffusional nanoparticles loses, which may arise due to long residence time?

7. How do you explain significant drop in dissociation constant between residence time 120 and 130s for 50nm particles at 30^{o}C?
8. Do you have an explanation for a large discrepancy between calculated and experimentally determined, for residence time 120s, temperature dependencies of dissociation constant as depicted on Fig 6? How would be those comparisons for aerosol particles of other sizes?

Correct answers to above question are indispensible part of the thesis defence.

Overall quality of the presented thesis is very good, and all the inaccuracies, omissions or typos in the text, except the author's specification contribution, are not of significant importance.

The presented thesis, corrected for upper-mentioned comments, fulfils the requirements for final work of PhD study in Environmental science at the Institute for Environmental Studies, Faculty of Science at Charles University in Prague and I do recommend thesis defence for the PhD title acquirement.

In Prague 26.5.2016

RNDr. Jan Hovorka, Ph.D.