9 of August 2014

Dear Editor Henrik Skov,

Thank you for your guidelines and comments on our manuscript! To meet the requests by the referees of straightening, streamline and trying not to be repetitious in the different Sections some changes has been made in the manuscript. Also, the Sections have been moved and the Figures/Tables with them in both the manuscript and the Supplement. The changes are highlighted with a yellow colour in the revised manuscript as well as listed in detail below. The answers to the referees (which are published in the interactive discussion) are included firstly in this document.

Best regards,

Cerina Wittbom
On behalf of all authors

NOTE! The first page no. refers to the published MS in ACPD, the second page no. (inside the brackets) refers to the revised MS that is sent to the Editor.

Response to Referee's (#1, acpd-14-c2874-2014) comments for
"Cloud droplet activity changes of soot aerosol upon smog chamber ageing"
doi:10.5194/acpd-14-8851-2014

Answer to General Comments:

In an attempt to meet the request from the referee of been sent back and forth the Experimental and Instrumentation sections (4 and 4.1) have been moved before the Theory and Modelling sections (2 and 3). Also, the figure order has been changed. Also some changes have been made in an attempt to streamline and straighten the MS, listed below.

The focus of the paper is the cloud droplet activity and how the hygroscopicity of the soot particles changes with organic coating or ageing. Therefore the introduction of the ADCHAM model has been kept short, although a small addition is made, not to mislead the reader of the focus. ADCHAM has been used (as stated in Sec. 3.2) "in order to better understand the mechanisms responsible for the observed changes in SOA coating, chemical composition and hygroscopic properties". Hence, the model is merely for support and to bring the discussion further.

Answer to Specific Comments:

Introduction:

This section have been changed somewhat in an attempt to meet the request of straightening and strengthen the introduction.

IVOCs

IVOCs have been changed to "Intermediate volatility organic compound " and a reference (Donahue et al., 2009) has been added to the MS at pg.8874, line 17 (MS: pg.22, lines 1-2) The division of VOCs according to Donahue et al. (2009) is generally accepted in the literature as described below:

"Low Volatility Organic Compounds (LVOCs): C* . {0.01, 0.1}μg m^{-3}. These compounds are mostly in the condensed phase in all but the most remote (and warm) parts of the atmosphere. Semi-Volatile Organic Compounds (SVOCs): C* .{1, 10, 100} μg m^{-3}. Significant fractions of these compounds will be found in both phases under typical conditions.

Intermediate Volatility Organic Compounds (IVOCs): C* . {10^3, 10^4, 10^5, 10^6} μg m^{-3}. These compounds are almost entirely in the gas phase, but they comprise an enormous number of difficult to measure compounds and (probably) a small but important fraction of the total atmospheric burden,…"

"Soot in the Arctic"

The paragraph on "soot in the Arctic" has been moved to an earlier and more general part of the introduction and somewhat changed to meet the request of the referee (#1).

Köhler theory

Köhler theory is introduced briefly in the Introduction (Sect.1) to get some background and motivation to why it is important to use the Köhler theory in this study. However, the Köhler equation is more thoroughly described in the Theory section (an addition has been made in the manuscript with reference to this section). In this paragraph the most common biogenic organic aerosol studies are listed to show the wider amount of studies performed in the field. The latter paragraph lists other kinds of studies as well as the studies regarding biogenic SOA above (gas phase and diesel soot particle SOA - anthropogenic, biogenic SOA, and ambient air) using the parameter κ. Hence, the two paragraphs have different foci.

Regarding the hygroscopicity of soot in general:

pg.8855, lines 4-5 (MS: pg.4, lines 12-14) in the Introduction states: "Freshly emitted soot particles are hydrophobic or limited in hygroscopicity and unlikely to contribute to the CCN population in the atmosphere (Weingartner et al., 1997; Meyer and Ristovski, 2007; Zhang et al., 2008; Tritscher et al., 2011).". Furthermore, it is stated that "non- or slightly hygroscopic compounds have a κ equal or close to zero" at pg.8857, line 13 (MS: pg.5, lines 29-30). Followed by the examples of κ for diesel exhaust. Due to the hydrophobicity of soot, κ is equal to zero for an uncoated soot particle (as defined at pg.8857, lines 12-13; MS: pg.5, lines 29-30). Many particles in the ambient air contain soot (e.g. Liu et al., 2013), but the soot fraction in the particle does not always reflect the obtained κ of these particles. Therefore, only the chamber experiment of Trischer et al. has been referenced. An addition on untreated soot particles has been made (pg.8857, line 14; MS: pg.5, line 30). Pure soot in the ambient air is hard to measure (if existing), therefore are κ values of ambient air listed (see answer above).

Rissler's κ

pg.8857, lines 4-9 (MS: pg.5-6, lines 31-32 and 1-3): The two different κ has been introduced to acknowledge that the κ was first suggested by Rissler. The definitions of the different κ are similar (as described in the paper) and easy to translate. It doesn't matter which one to use. However, the κ introduced by Petters and Kreidenweis is more commonly and broadly used and therefore we have decided to use that one. But to clarify to the readers, both κ are introduced.

Shape and morphology
The diameter of the agglomerated soot particles will be different depending on measurement techniques. For example, the non-spherical shape of a fresh soot aggregate will have a larger mobility diameter (measured by the DMA) than the volume equivalent diameter, as discussed in the paper. Knowledge of the structure is thus needed to relate mobility diameter to particle volume and mass. Additionally, the agglomerated shape of the freshly emitted soot particle might be able to hinder the water uptake (according to what is stated in the literature). The branches, made of soot primary particles, are unfavourably both changing the curvature of the surface area and there is also a possibility of unevenly distributed organic material, which "hide" in cavities and voids (discussed in the result section). An addition is made (pg.8855, line 6; MS: pg.4, line 14-16) to highlight the difficulties when using different measurement techniques.

Sect. 3.2 (now 4.2) ADCHAM:
The gas- and particle- phase chemistry is complex and hard to understand and the partitioning is not known. The kappa-Köhler theory deviates from measured results in the early ageing process (when the amount of organic material is small) and to support our assumptions and bring the discussions further the Aerosol Dynamics, gas- and particle- phase chemistry model for laboratory CHAMber studies (ADCHAM) was used. The results from the model are used for the molar mass of SOA (pg.8862, lines 19-21; MS: pg.15, lines 4-5). Otherwise, the general behaviour of the model results is used in the discussion. A clarifying addition is made in the text (pg.8864, lines 4 and 8; MS: pg.16, lines 11-13 and 15-21): "ADCHAM is a model primarily intended to be used to recreate laboratory chamber experiments on SOA. The model explicitly simulates the deposition and re-evaporation of organic compounds from the chamber walls, all fundamental aerosol dynamics processes, detailed gas- and particle- phase chemistry and the mass transfer limited mixing of compounds in the particle phase."

DEP2
pg.8864, line 6. After changing the order of the section the abbreviation is now introduced, as well as a clarification is made in the MS.

DMA-TD-APM
The same labelling is now used throughout the whole MS.

Sect. 5
Carbon mean oxidation state
pg.8869, line 1 (MS: pg. 17, line 8): The term "carbon mean oxidation state" is introduced here the first time in the manuscript, together with the equation used for the estimation of it and a reference. The carbon mean oxidation state is a useful parameter for understanding oxidation-reduction (redox) and combustion reactions. The term is commonly used to determine the changes in redox reactions. The oxidation state of a molecule describes the transfer of electrons. Using the AMS, we are able to measure this term. The oxidation state term is commonly used in the chemistry community (and other fields) and therefore it is of interest to present these results here. The reference used in the paper (Kroll et al., 2011) gives a good presentation of the carbon mean oxidation state, calculated from AMS measurements. An addition is made in the MS to clarify (MS:pg.17, lines 6-12).

CCN and primary particle diameter
pg.8870, line 1 (MS: pg.17, line 28 to pg.18, line 2)(same answer to both referees): As pointed out by the referee the volume equivalent diameter (d_{ve}) is used as input in the Köhler

theory. The d_{ve} in turn is affected by the structure and size of the primary soot spherules. The smaller primary spherules (FSP2) make up particles with higher agglomeration than when larger primary particles are present (FSP1 and DEP), according to previous studies (see pg.8876, lines 13 to 19; MS: pg.23, lines 24 to 29). Results from the DMA-TD-APM measurements show that the d_{ve} for FSP1 is closer to d_{ve} for DEP, than d_{ve} for FSP2 (Supplement, Fig. S 2). The primary particle diameters are calculated from the mass-mobility relationship. We assume that the particle diameters for FSP1 and all DEP experiments are more similar than FSP1 and FSP2.

To strengthen this assumption are the results from CCN measurements (a linguistic change is made in the MS). In Figure 10, the results show that the FPS1 and DEP particles need a higher critical supersaturation (s_c) for activation than the FSP2 particles, i.e. the FSP2 particles have more organic material for a given d_{ve} than FSP1 and DEP and are therefore easier to activate into cloud droplets. While in Figure 12, it is the opposite. The FSP2 particles are harder to activate for a certain $\mathrm{mf}_{\mathrm{SOA}}$, due to smaller d_{ve}. This is summarised in a conceptual way in Fig.A below.

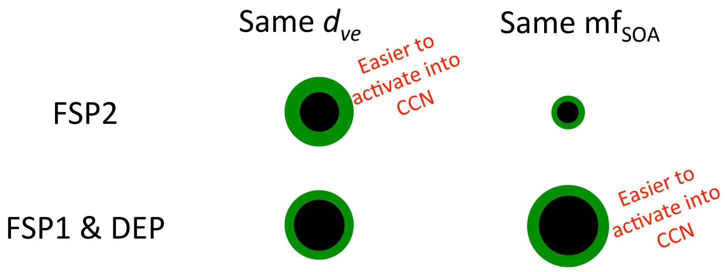

Figure A. Conceptual picture of how particles with the same volume equivalent diameter or the same mass fraction SOA activate at different critical supersaturation, depending on primary particle diameter. NOTE, the illustration is only conceptual and therefore shows spherical particles instead of the agglomerated soot structure.

This issue is discussed in Sect. 5.2 (pg.8877, lines 2-16 MS: pg.24, lines 9 to 20).

The 4 factors
The 4 factors are the outcome of our investigation. A linguistic change has been made in the MS to clarify this (pg.8872, line 8; MS: pg.19, line 30 to pg.20, line 4).

Sect. 6
The change in hygroscopicity and morphology of the ageing soot particles will affect the deposition of the particles in the human respiratory tract according to Londahl et al., 2009 (described in the introduction of the paper, pg.8854, line 11). Changes made in the manuscript (pg.8888, line 7, MS: pg.33, line 22).

Technical notes
These are corrected/added/changed if not commented below.

pg.8853, line 11 (MS: pg.2, line 22): "ERFari" is "...the effective radiative forcing from aerosol-cloud interactions (ERFaci..." as stated in the paper
pg. 8853, line 22-23 (MS: pg.3, line 2): "RFari" is "...radiative forcing from aerosol-radiation interactions (RFari..." as stated in the paper

---

Response to Referee's (#2, acpd-14-C2886-2014-supplement) comments for
"Cloud droplet activity changes of soot aerosol upon smog chamber ageing"
doi:10.5194/acpd-14-8851-2014

Answer to General Comments:

As requested, some attempts have been made to streamline the paper and make the results section less repetitious.

Regarding Submission to special issue
As pointed out by the referee BC (black carbon) is believed to affect the radiation balance in the Arctic region. However, it is not clear to what degree BC or aged BC influences this region as absorbing or reflecting particles, and the contribution to the cloud condensation nuclei (CCN) or ice nuclei (IN) populations. This study addresses the role of BC and of photochemically processed BC as CCN. The laboratory work performed here is part of the work accomplished within the CRAICC (Cryosphere-atmosphere interactions in a changing Arctic climate) community (Workpackage 5: *Cryosphere-aerosol-cloud-climate interactions*). Although, the results in this study are not directly of linked to the Cryosphere, it is of importance to understand the role of BC as CCN at micro scale in the Atmosphere. Part of the results in this paper has been presented at workshops and meetings within the CRAICC community.

In the overall aim it is stated that short-lived climate pollutants (soot being one of the most important) is a focus of CRAICC:
"*We will focus our attention on the role played by short-lived pollutants (recently also called short-lived climate forcers,* ***SLCF****) and clouds and their linkages with cryospheric changes".*

In the program for CRAICC WP5 *Cryosphere-aerosol-cloud-climate interactions* it is stated:

"The aim is to gain detailed understanding of the relation between the cryosphere, aerosols and clouds, as well as their climate feedbacks in the Arctic, with a combination of field and lab studies and modelling. The objectives are to analyze:

- *the contributions of natural and anthropogenic aerosol types in the North (primary, secondary, BC and their combinations) to different cloud types*

This is done by a combination of 1……….3) laboratory studies on ice nucleating properties of aerosols and cloud droplet formation ability"

"In particular it will provide information of the processes that control concentrations of soot, other inorganic and organic particles and ozone in the atmosphere and how these species interact with climate and cryosphere."
(For more information please see the CRAICC homepage:
http://www.atm.helsinki.fi/craicc/index.php/wp-5)

The paragraph regarding the Arctic in the introduction have been moved and changed somewhat to better fit the context in the introduction.

Answers to Specific comments:

The Experimental and Instrumentation sections (4 and 4.1) have been moved, as suggested by the referee (#2), before the Theory and Modelling sections (2 and 3). Hence, **Experimental** is now Sect.2, **Instrumentation** is Sect.2.1, **Theory** is Sect.3, **Modelling** is 4, **CCN activation modelling** is Sect.4.1 and **Modelling of gas-phase chemistry, organic aerosol formation and composition: ADCHAM** is Sect.4.2. Also, the figure order has been changed.

Distinguishing between POA and BC
pg.8861, Line 22 (MS: pg.14, line 11): States how POA has been approximated. The APM measures a small mass fraction of organics before the photochemical processes have started. This organic mass has been considered as POA. The AMS measurements support this assumption.

pg.8867, Line 1 (MS: pg.8, lines 23 and 24): increase/decrease: 120 s, constant max/min: 20 s. This has been included in the manuscript.

SP-AMS
pg.8868, lines 9-15 (MS: pg.9, line 30 to pg.10, line 7): That is correct, the conventional Tungsten vaporizer was retained during these experiments, and kept at the standard 600 °C (initially, we tried turning off the Tungsten vaporizer, however, the heat from the filament was enough to heat the vaporizer sufficiently for vaporization of the organic PM we sampled). The manuscript has been updated to indicate this.
The filament was also continuously engaged (the ions we studied here all origin from 70 eV electron ionization: hence, there is no signal without the filament current). The reason why we chose to operate the laser in hourly five minute intervals was that at the time of these experiments (late 2010, early 2011) we did not understand how the SP-AMS works well enough to sacrifice more of the (non-SP) AMS sampling, which we employed in an overlapping campaign (Nordin et al., 2013) and had previous experience with.

We used a HEPA filter to quantify the gas phase CO_2 interferences, and made sure the fragmentation table adequately described the gas phase contribution to CO_2^+. Interestingly the gas phase interferences were utterly negligible compared with the CO_2^+ signal originating from the rBC as illustrated in Fig. A below:

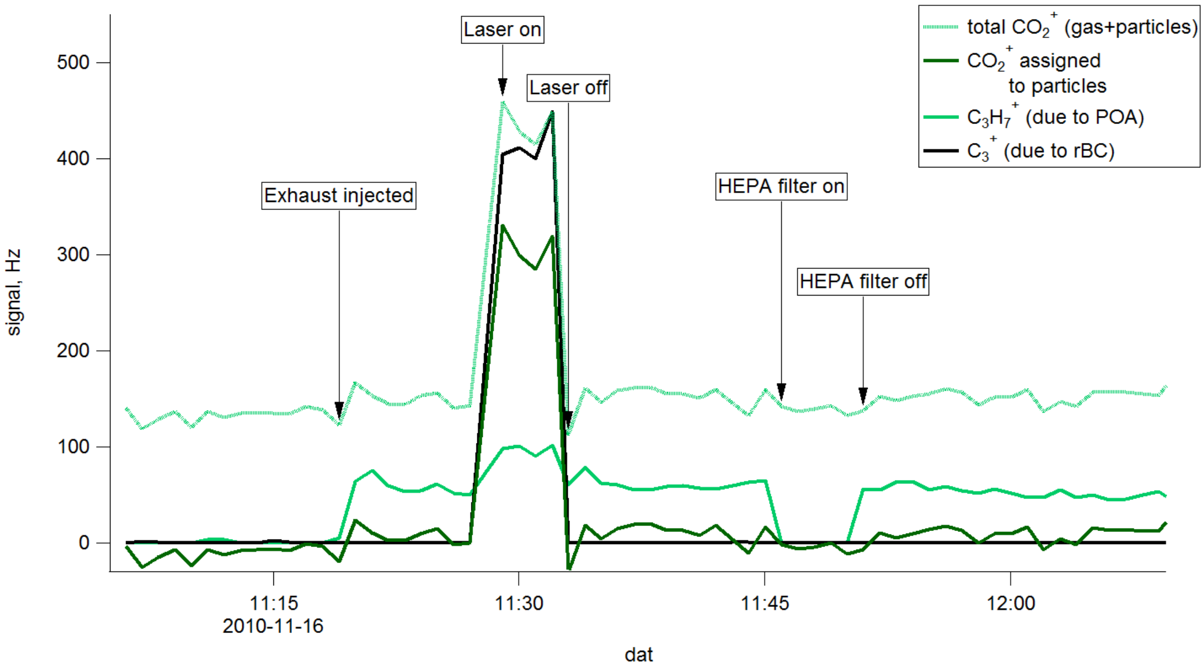

Figure A. Selected ions after exhaust injection in experiment DEP2.

The data with HEPA filter in line after 1145 was one of three sets used to determine the correct fragmentation table in this experiment (we used a constant value based on the average). For reference, the CO_2^+ signal due to aged organics sampled with the laser disengaged at the end of the experiment (which impacts the estimated mean carbon oxidation state) was about 400 Hz.

We used the sampling with laser engaged (which in our experiments caused vaporization but not ionization) to estimate the relative amount of organic coatings and BC. As detailed in the Supplement (because it is a technical point which mainly interests specialists) "Determining mf_{OA} from the SP-AMS data" we assumed a collection efficiency of 1 for the laser vaporizer for both organics and BC. Note however that our results are not sensitive to that number, as we report the relative amounts and not absolute concentrations. The relative ionization efficiencies used were 0.2 for BC and 1.4 for organic PM, default estimates due to lack of calibration experiments. For the conventional vaporizer we made no assumptions about the collection efficiencies: that data was used to investigate the chemical evolution of the organic PM we sampled. Given internal mixtures, collection efficiencies have no impact on that analysis.

DMA-TD-APM

A small addition regarding the volume equivalent diameter has been made as well as the operational details have been added in a Table to the Supplement (with a reference in the MS), MS: pg.9, lines 21-24 and Supplement Table S1.

CCN and primary particle diameter

pg.8870, line 1 (MS: pg.17, line 28 to pg.18, line 2)(same answer to both referees): As pointed out by the referee the volume equivalent diameter (d_{ve}) is used as input in the Köhler theory. The d_{ve} in turn is affected by the structure and size of the primary soot spherules. The smaller primary spherules (FSP2) make up particles with higher agglomeration than when larger primary particles are present (FSP1 and DEP), according to previous studies (see pg.8876, lines 13 to 19; MS: pg.23, lines 24 to 29). Results from the DMA-TD-APM measurements show that the d_{ve} for FSP1 is closer to d_{ve} for DEP, than d_{ve} for FSP2 (Supplement, Fig. S 2). The primary particle diameters are calculated from the mass-mobility relationship. We assume that the particle diameters for FSP1 and all DEP experiments are more similar than FSP1 and FSP2.

To strengthen this assumption are the results from CCN measurements (a linguistic change is made in the MS). In Figure 10, the results show that the FPS1 and DEP particles need a higher critical supersaturation (s_c) for activation than the FSP2 particles, i.e. the FSP2 particles have more organic material for a given d_{ve} than FSP1 and DEP and are therefore easier to activate into cloud droplets. While in Figure 12, it is the opposite. The FSP2 particles are harder to activate for a certain mf_{SOA}, due to smaller d_{ve}. This is summarised in a conceptual way in Fig. B below.

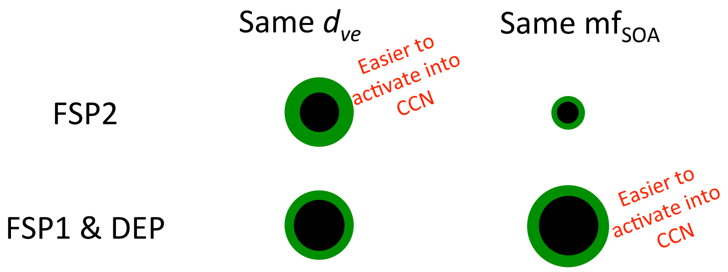

Figure B. Conceptual picture of how particles with the same volume equivalent diameter or the same mass fraction SOA activate at different critical supersaturation, depending on primary particle diameter. NOTE, the illustration is only conceptual and therefore shows spherical particles instead of the agglomerated soot structure.

This issue is discussed in Sect. 5.2 (pg.8877, lines 2-16, MS: pg.24, lines 9 to 20).

BC and organic masses from SP-AMS
pg.8871, lines 2-3 (MS: pg.18, lines 29-31): the text is still in the Supplement. We don't want to extend the paper further and change the focus of the paper. The text is still in the results section to make it easier for the reader to understand the context and don't mix up the instrument used (the SP-AMS mass fractions are only used for one experiment). However, a change/addition has been made in the Instrumentation section (pg.8868, line 13 (MS: pg.10, lines 3-7)) to give some information early on.

Figure 1. has been updated as suggested by the referee (#2).

Figure 9 and in the text:
The TEM pictures are not used to distinguish between hygroscopic and hydrophobic parts, only the soot is visible. The TEM pics are used for (a) showing the structure of the soot particles at different stages of coating/ageing (pg.8875, line 24; MS: pg.23, line 5), and (b) for visualising the theory of activation sites (pg.8875, lines 28 to pg.8876, line 6; MS: pg.23, lines 8-16). For the freshly emitted soot particles or the slightly coated soot particles an untypical behaviour is seen. From calculations using Köhler theory a lower critical supersaturation should be required then are actually measured. The deviation between modelled and measured results show that the overall soluble volume fraction is not the only important parameter for the CCN activation. Therefore, we suggest other possibilities and discuss them (activation sites is one possibility). In the conclusion (Sect. 6) we summarize what we believe is of importance. NOTE, this is only of importance for the freshly emitted/slightly coated soot particles.
Some linguistic changes have been made to clarify this. pg.8875, line 29 to pg.8876, line 4 (MS: 23, lines 10-23).

Minor Comments:
The minor comments are corrected and the yellow colour has been changed to pink or darkened when adequate.

Changes in the manuscript:

The Experimental and Instrumentation sections (4 and 4.1) have been moved, as suggested by the referee (#2), before the Theory and Modelling sections (2 and 3). Hence, **Experimental** is now Sect.2, **Instrumentation** is Sect.2.1, **Theory** is Sect.3, **Modelling** is 4, **CCN activation modelling** is Sect.4.1 and **Modelling of gas-phase chemistry, organic aerosol formation and composition: ADCHAM** is Sect.4.2.

To streamline the Results section and not making the reader go back and forward as much as before (as requested by the referee (#1) the Figure order has been changed as follows:

- Figure 7a and b are now Figure 2a and b.
- Figure 2-6 are now Figure 3-7.
- Figure 11 and 12 has swopped places.

Title Page, Author list:

"J.H. Pagels" and "A.C. Eriksson" have changed position in the author list from:

"C. Wittbom[1], J.H. Pagels[2], J. Rissler[2], A. C. Eriksson[1], J. E. Carlsson[2], P. Roldin[1,3], E. Z. Nordin[2], P. T. Nilsson[2], E. Swietlicki[1], and B. Svenningsson[1]"

to:

"C. Wittbom[1], A. C. Eriksson[1], J. Rissler[2], J. E. Carlsson[2], P. Roldin[1,3], E. Z. Nordin[2], P. T. Nilsson[2], E. Swietlicki[1], J.H. Pagels[2], and B. Svenningsson[1]"

Abstract:

pg.8852, line 17 (MS:pg.1 , line 28): "volatile" is exchanged for "volatility" → "intermediate volatility organic compound (IVOC), according to the definition by Donahue et al. (2009)
pg.8852, line 17 (MS:pg.2 , lines 4-5): "(POA or SOA)" is exchanged for "(e.g. primary or secondary organic aerosol)"
pg.8852, line 23 (MS:pg.2 , line 8): "(mf_{SOA}(APM))" is erased.
pg.8852, line 28 (MS:pg.2 , line 10): "(mf_{SOA}(APM))" is exchanged for "SOA mass fraction".

pg.8853, line 1 (MS:pg.2 , line 10): "with" is exchanged for "by"
pg.8853, line 1 (MS:pg.2 , line 11): "during the early stages of ageing" is added for clarification, whole sentence "The discrepancy between theory and experiments during the early stages of ageing might be due to solubility limitations, unevenly distributed organic material or hindering particle morphology."

1 Introduction

pg.8853, lines 15-16 (MS: pg.2, line 25-26): "increase in cloud albedo due to addition of cloud nuclei by pollution (Twomey, 1974)" has been moved within the sentence, whole sentence as follows:
"The indirect aerosol effect includes increase in cloud albedo due to addition of cloud nuclei by pollution (Twomey, 1974), reduction of drizzle and increased cloud lifetime (Albrecht,

1989) as well as an increase in cloud thickness (Pincus and Baker, 1994). Also, soot aerosol can contribute to daytime clearing of clouds (Ackerman et al., 2000).”
pg.8853, line 18 (MS: pg.2, line 28): “for” is erased. →“Soot particles make up a large fraction…”
pg.8853, line 26 (MS: pg.3, line 5):”BC is estimated to have” is exchanged for “, with”, making one sentence of two as follows: “After CO2, BC is estimated as the strongest anthropogenic climate-forcing agent in the present-day atmosphere (Bond et al., 2013), with a total warming effect of about +1.1 W m-2 (with 90% uncertainty bounds of +0.17 to 2.1 W m-2).”

pg.8854, line 5 (MS: pg.3, lines 25-26):”…linked with climate change and…” is erased and “…in addition to being linked with climate change…” is added, whole sentence: “According to the World Health Organization (WHO, 2012), emissions of diesel engine exhaust are classified as carcinogenic in humans in addition to being linked with climate change.”
pg.8854, line 6 (MS: pg.3, lines 26-27):”Exposure to diesel exhaust is associated with unfavourable health effects…” is exchanged for “The unfavourable health effects caused by exposure to diesel exhaust are described in several studies…”
pg.8854, line 9 (MS: pg.3, line 29): “acquire” is exchanged for “require”
pg.8854, lines 19-26 (MS: pg.4, line 5): The following has been erased in an attempt to straighten the introduction as requested by the referee (#1):
“Typically, the particle size distribution of diesel exhaust is bimodal. Most of the particle mass is found in the in the size range 0.05–1.0 μm (volume mean diameter of approximately 100 to 300 nm), composed of carbonaceous agglomerates and volatile matter absorbed onto their surface (Kittelson et al., 1998). Whilst most of the particle number is typically found in the diameter range of 0.001 to 0.05 μm (depending on running conditions) and consists of organic and sulphur compounds, and may also contain metal compounds. Emissions from diesel engines vary significantly with the engine load (Srivastava et al., 2011).”
pg.8854, line 28 (MS: pg.4, line 7): “by atoms” has been added to the sentence.

pg.8855, line 6 (MS: pg.4, line 14-16):” The size of these non-spherical particles will normally differ between measurement techniques (DeCarlo et al., 2004). For example, showing a larger mobility than volume equivalent diameter.” The two sentences are added to highlight the difficulties of measuring the freshly emitted soot particles.
pg.8855, line 27 (MS: pg.5, line 4):”, i.e.” is erased.

pg.8856, lines 8-24 (MS: pg.3, line 10): The paragraph regarding the Arctic is moved and changed/merged into the text for a better context.
Old paragraph: “It is believed that the radiative balance in the Arctic is influenced in numerous ways of aerosol particles. For example soot, when deposited on snow and ice, may enhance surface heating and ice melting by decreasing the surface albedo (e.g. Bond et al., 2013; Tunved et al., 2013 and references therein). As mentioned previously, photochemical ageing of the soot particles may enhance the absorption of light, which in turn then may influence and boost the warming of the surroundings even further. The particle number concentration is in general low (about a couple of hundreds cm^{-3}) in the Arctic (Tunved et al., 2013). Wet removal and photochemical ageing play central roles in controlling the aerosol size distribution properties during the Arctic summer time. If aged soot particles are transported to this remote site, an increase in hygroscopicity of these particles could result in an increased number of cloud droplets i.e. affect the cloud configuration of the Arctic. Changes in abundance and chemical properties of aerosol particles will indirectly affect cloud microphysical properties and lifetime, as mentioned above. As pointed out by Tunved et al.

(2013), it is highly relevant to obtain a better understanding of the microphysical properties of the different aerosol populations abundant in the Arctic environment, in order to improve the understanding of the direction and magnitude of a future climate change in the region."

New paragraph: " Soot is not only present close to sources, but can be transported long distances. The warming effect of soot in the arctic has recently gained increased interest, both due to transport of soot from urban areas and also due to possible future soot emissions in the arctic area from ship traffic which is made possible due to reduction in ice coverage. The enhanced light absorption by soot, due to condensing material during photochemical ageing, may influence and boost the warming of the surroundings even further. Also, soot deposited on snow and ice may enhance surface heating and ice melting by decreasing the surface albedo (e.g. Bond et al., 2013; Tunved et al., 2013 and references therein). The particle number concentration is in general low (about a couple of hundreds cm-3) in the Arctic (Tunved et al., 2013). Wet removal and photochemical ageing play central roles in controlling the aerosol size distribution properties during the Arctic summer time. As pointed out by Tunved et al. (2013), it is highly relevant to obtain a better understanding of the microphysical properties of the different aerosol populations abundant in the Arctic environment, in order to improve the understanding of the direction and magnitude of a future climate change in the region."
pg.8856, line 26 (MS: pg.5, line 14):" (further described in Sect. 3)"

pg.8857, line 1 (MS: pg.4, lines 17-18): "…SOA from…" are added and " compounds is exchanged for "precursors" for clarification, whole sentence: "Previous work regarding the activation properties and processes of organic aerosols in reaction chambers, often study SOA from biogenic precursors such as limonene, α-pinene and β-caryophyllene (Prenni et al., 2007 (α-pinene, β-pinene, Δ3-carene, and toluene); Duplissy et al., 2008 (α-pinene); Kundu et al., 2012 (limonene); Frosch et al., 2013 (β-caryophyllene)). "
pg.8857, line 7 and 9 (MS: pg.5, lines 24 and 26): "b" is erased from the reference: Petters and Kreidenweis (2007)
pg.8857, line 14 (MS: pg.5, line 30): "(such as untreated soot particles; e.g. Henning et al., 2012)" is added after "κ=0".
pg.8857, line 18 (MS: pg.6, line 3): "e" is erased: "for" instead of "fore"
pg.8857, line 24 (MS: pg.6, line 8): "s" is erased from the word "particles"; should be "particle"
pg.8857, line 25 (MS: pg.6, line 10): "was" is exchanged for "is"

pg.8858, lines 5-7 (MS: pg.6, line 19): "Comparing modelled (κ-Köhler) with empirical results show good agreement when taking the change in particle properties into account. " is erased

3 Theory (Sect.2 before)

pg.8858, line 24 (MS: pg.11, line 11): "…input values are listed in Table 2…" added/changed. → "(input values are listed in Table 1)."

pg.8859, line18 (MS: pg.12, line 8): a "4" is added to the AS dissociation number. → $(NH_4)_2SO_4$

4 Modelling (Sect. 3 before)

4.1 CCN activation modelling (Sect. 3.1 before)

pg.8861, line 8 (MS: pg.13, line 20): “similiar” is changed to “similar”.
pg.8861, line 12 (MS: pg.13, line 23): “TD” is added → “DMA-TD-APM”
pg.8861, line 12 (MS: pg.13, line 24): “Sect. 4.1” is changed to “Sect. 2.1”

pg.8862, line 2 (MS: pg.14, line 15): “SP-“ added: “SP-AMS” instead of “AMS”.
pg.8862, line 9 (MS: pg.14, line 21): “Fig. S1” is exchanged for “Fig. S 2”
pg.8862, line 21 (MS: pg.15, line 4): “Sect. 3.2” is changed to “Sect. 4.2”

pg.8863, line 16 (MS: pg.15, line 27): “(<0.2)” is moved and “…, not shown” is added to the sentence: “For fresh POA a κ value below 0.1 is expected, due to the low O:C ratio (<0.2, not shown) and high M_{POA} of the material, as pointed out by others (Tritscher et al., 2011; Kuwata et al., 2013).”
pg.8863, line 24 (MS: pg.16, line 3): “a” is erased the sentence should be: “Due to the insolubility of black carbon, the soot particle as a whole is treated as water insoluble.”
pg.8863, lines 24-25 (MS: pg.16, line 3): “As described previously κsum varies with volume fractions of the organic coating material.” Is erased
pg.8863, line 26 (MS: pg.16, line 4): “thus” is added

4.2 Modelling of gas-phase chemistry, organic aerosol formation and composition: ADCHAM (Sect. 3.2 before)

pg.8864, line 4 (MS: pg.16, line 10 and 11): “and to bring the discussion further” is added. ”we performed” is exchanged for ”was performed” and moved
pg.8864, line 6 (MS: p.16, line 12): “DEP2” is exchanged for “one of the experiments (DEP2, see Table 1)”. → Full sentence: “In order to better understand the mechanisms responsible for the observed changes in SOA coating, chemical composition and hygroscopic properties and to bring the discussion further detailed modelling were performed of the gas-phase chemistry, SOA formation and composition during one of the experiments (DEP2, see Table 1).”

pg.8864, line 8 (MS: pg.16, lines 14-15): the definition of ADCHAM is added: “…Aerosol Dynamics, gas- and particle- phase chemistry model for laboratory CHAMber studies; (ADCHAM; Roldin et al., 2014).”
pg.8864, line 8 (MS: pg.16, lines 16-20): One sentence has been added to meet the request from the referee: “ADCHAM is a model primarily intended to be used to recreate laboratory chamber experiments on SOA. The model explicitly simulates the deposition and re-evaporation of organic compounds from the chamber walls, all fundamental aerosol dynamics processes, detailed gas- and particle- phase chemistry and the mass transfer limited mixing of compounds in the particle phase.” And “ADCHAM…” has been changed to “It…” in the following sentence.

2.1 Instrumentation (Sect. 4.1 before)

pg.8866, line 11 (MS: pg.8, lines 5-6):”Sample air passes along this axis surrounded by sheath air.” Is added for clarification.
pg.8866, line 13 (MS: pg.8, line 8): “.., Table 1” is added within the parenthesis
pg.8866, line 15 (MS: pg.8, line 10): “(DEP1, Table 2)” is erased and “. Also, the mobility diameter (dm) was kept constant.” is added
pg.8866, lines 19-20 (MS: pg.8, line 14): “The supersaturation was changed in the stepwise way while keeping the mobility diameter (d_m) constant.” is erased
pg.8866, lines 24 (MS: pg.8, line 19): “Table 2” is changed to “Table 1”
pg.8866, lines 25 (MS: pg.8, line 18): “super saturation” is changed to “supersaturation”

pg.8867, Line 1(MS: pg.8, line 23): “(for 120 s)” is added.
pg.8867, Line 1(MS: pg.8, line 24): “(for 20 s)” is added.
pg.8867, line 9 (MS: pg.8, line 30): “Fig. S2” is changed “Fig. S 1”
pg.8867, lines 26-27 (MS: pg.9, line 14):”, described in detail elsewhere (Rissler et al., 2013)” has been moved to the end of the paragraph.
pg.8867, line 14 (MS: pg.9, line 4):”(Moore and Nenes, 2009)” is added for clarification

pg.8868, line 1 (MS: pg.9, lines 17): “...hence a DMA-TD-APM. The thermodenuder…” is added
pg.8868, line 3 (MS: pg.9, lines 20-23): “For calculations of the volume equivalent diameter (d_{ve}), the peak of the mass distribution was used. An example of operational details is found in the Supplement (Table 1) and a more detailed description of the operational procedure and calculations are found elsewhere (Rissler et al., 2013; Rissler et al., 2014). “ added to the manuscript for more details of the DMA-TD-APM, as requested.
pg.8868, line 11 (MS: pg.9, line 31): “r-BC” is changed to “rBC”,
pg.8868, line 12 (MS: pg.9, line 32):”further” is erased.
pg.8868, line 15 (MS: pg.10, lines 2-6): Adding/changing text according to referee suggestion “, while the Tungsten vaporizer (used in an (non-SP) AMS) was engaged continuously. The 5 minute intervals were used to estimate the organic aerosol (OA) mass fraction (mf_{OA}) further described in the Supplement. The Tungsten vaporizer data was only used to study the chemical composition of the OA. Both instrument configurations are described elsewhere (DeCarlo et al., 2006; Onasch et al., 2012). “
Whole paragraph as follows:
“The chemical composition of the particles (soot core and organic coating) was determined using an online Aerodyne High-Resolution Time of Flight Mass Spectrometer (HR-ToF-AMS, Aerodyne research). For detection of refractory black carbon (rBC) the instrument was equipped with a laser vaporizer, which is referred to as a Soot Particle Aerosol Mass Spectrometer (SP-AMS, Aerodyne research). The laser was operated in 5 min periods every hour, while the Tungsten vaporizer (used in an (non-SP) AMS) was engaged continuously. The 5 minute intervals were used to estimate the organic aerosol (OA) mass fraction (mfOA) further described in the Supplement. The Tungsten vaporizer data was only used to study the chemical composition of the OA. Both instrument configurations are described elsewhere (DeCarlo et al., 2006; Onasch et al., 2012).”
pg.8868, line 25 (MS: pg.10, line 16): an “a” is missing, should be “Analytik”.
pg.8868, line 28 (MS: pg.10, line 18): “in” is exchanged for “by”.

5 Results and discussion

5.1 The overall picture

Trying to meet the request from the referees we have tried to streamline the explanations and put the Figures in a new order to better fit the text, not making the reader go back and forward as much as before.

pg.8869, line 4 (MS:pg.17, line 4): “flame” is added
pg.8869, line 5 (MS:pg.17, line 5): “(Figs. 2, 4, 5 and 6)” is changed to “(further described in Sect.5.2)”
pg.8869, lines 6-11 (MS:pg.17, lines 6-12): trying to clarify the “carbon mean oxidation state”, the sentence is changed to sentences; “The newly emitted diesel exhaust aerosol show a carbon mean oxidation state in the range -1.9 to -1.6 (Fig. 2a), obtained from AMS results (where the carbon mean oxidation state ≈ 2 O/C-H/C; Kroll et al., 2011). The results are similar with previous studies (Kroll et al., 2011). Furthermore, the mass spectrum of the

newly emitted diesel exhaust aerosol show an organic (POA) content of mostly hydrocarbon species (Fig. 2b), consistent with diesel exhaust measurements by Canagaratna et al. (2007). "
pg.8869, lines 7, 10 and 12 (MS:pg.17, lines 7,11 and 13): "Fig. 7…" is changed to "Fig. 2…"
pg.8869, lines 22 (MS: pg.17, line 22): "s" has been erased from "particles"

pg.8870, lines 1-4 (MS: pg.17, lines 28-31 to pg.18, lines 1-2): "CCN measurements support that the d_{pp} in experiment FSP1 is close to the d_{pp} of the diesel exhaust particles (Supplementary material, Fig. S1, FSP1 – circles and DEP1-3 - triangles falls on the same line), while the d_{pp} in FSP2 are considerably smaller."
is exchanged for
"Results from these measurements show that the volume equivalent diameters (d_{ve}) of FSP1 are closer to the d_{ve} of the DEP (Supplement, Fig. S 2, FSP1 – circles and DEP1-3 - triangles falls on the same line), implying that the primary particle diameter (d_{pp}) also is similar for FSP1 and DEP, but different for FSP2. The differences between FSP1 and FSP2 are reflected in the CCN measurements, were FSP1 and DEP require a more similar supersaturation for activation, further discussed in Sect.5.2"

pg.8870, line 4 (MS: pg.18, lines 8-10): "The d_{pp} from calculations as well as TEM picture analysis are listed in Table 2." The sentence is moved and merged into the last sentence in the paragraph and Table number is changed, whole sentence: "Primary particle diameters (d_{pp}) from calculations and TEM picture analysis as well as selected sizes (d_m) for the CCN measurements for the different experiments are listed in Table 1. "

pg.8870, line 5 (MS: pg.18, line 3): "mobility" has been added for clarification
pg.8870, line 15 (MS: pg.18, line 14): "(Fig. 4-6)" is exchanged with "(see Sect. 5.2 for details)"
pg.8870, line 20 (MS: pg.18, line 18): "…, Fig. 3)" has been added within the brackets
pg.8870, line 20 (MS: pg.18, lines 19-20): "…, without any size selection upstream the CCNC." Has been added for clarification.

pg.8870, line 25 (MS: pg.18, line 24): "Fig. 2" is changed to "Fig. 3"
pg.8870, line 28 (MS: pg.18, line 27): "Fig. 3" is changed to "Fig. 4"

pg.8871, lines 7-8 (MS: pg.19, line 3): "Fig. 2 and 3" is changed to "Fig. 3 and 4"
pg.8871, line 17 (MS: pg.19, line 11): "Fig. 4-6" is changed to "Fig. 5-7"
pg.8871, line 22 (MS: pg.19, line 15): "Fig. 5" is changed to "Fig. 6"
pg.8871, line 26 (MS: pg.19, line 20): "compare Fig. 5 and 6" is changed to "compare Fig. 6 and 7"

pg.8872, line 5 (MS: pg.19, line 27): "life time" is changed to "lifetime"

5.2 Detailed picture

pg.8872, lines 8-9 (MS: pg.19, lines 30-31 to pg.20, lines 1-4): clarifying that the 4 factors are our way of summarising the properties that matters. Sentence is changed to: "In general, cloud droplet formation ability of aerosol particles can be described as a function of the number of dissolved molecules and ions in the activating droplet, parameterized by for example the κ-value. In the case of particles dominated by soot, the volume occupied by insoluble material influences the Kelvin effect and therefore has to be taken into account. In

present experiments, the time-dependent changes of activation properties of the coated soot cores are summarised to four factors:…”
pg.8872, lines 13-14 (MS: pg.20, line 8): “Fig. 5 and 6” is changed to “Fig. 6 and 7”
pg.8872, line 18 (MS: pg.20, line 13): “(Fig. 12)” is erased.
pg.8872, line 20, 25 and 26 (MS: pg.20, line 15, 20 and 22): “(Fig. 7…” is changed to “(Fig. 2…”
pg.8872, line 28 (MS: pg.20, line 23): “at” is changed to “below” and supersaturation is erased.

pg.8873, line 1 (MS: pg.20, line 24): “Sect. 3.2” is changed to “Sect. 4.2”
pg.8873, line 20 (MS: pg.21 , line 9): “initial” is added
pg.8873, line 24 (MS: pg.21 , line 13): “OA” is changed to “total organic aerosol (OA)”
pg.8873, line 25 (MS: pg.21 , line 14): “…, see e.g. Fig. 2” is erased
pg.8873, line 28 to pg.8874, line 1 (MS: pg.21, lines 17-18): “Just after the onset of UV exposure (within 5 min), the first activation of the exhaust aerosol particles is observed. Hence, when the exhaust aerosol and precursors are subjected to UV radiation, an enhancement in hygroscopicity occurs.” is changed to “As mentioned in Sect. 5.1, the first enhancement in hygroscopicity of the particles is observed when the exhaust aerosol and precursors are subjected to UV.”, to streamline the manuscript.

pg.8874, line 8 (MS: pg.21, line 24): “(Fig. 7…” is changed to “(Fig. 2…”
pg.8874, line 17 (MS: pg.22, lines 1-2): “intermediate volatile organic compounds” is exchanged for “intermediate volatility organic compounds” and a reference is added within the brackets: “(IVOCs; Donahue et al., 2009)”.
pg.8874, line 24 (MS: pg.22, line 8):”under-” is added, the word was missing in “underestimates”

pg.8875, line 1, 2 and 10 (MS: pg.21, line 12, 13 and 21): “(Fig. 7…” is changed to “(Fig. 2…”
pg.8875, line 14 (MS: pg.22, lines 24-26): The sentence have been changed from: “This is partly explained by the Kelvin effect, which is more pronounced for particles with higher curvature and partly by the fewer amounts of water-soluble ions or molecules.”
To:
” This is mainly explained by the fewer amounts of water-soluble ions or molecules and thus larger Kelvin effect, which is more pronounced for particles with higher curvature.”

pg.8875, lines 17 and 19 (MS: pg.22, lines 27 and 30): “Fig. 6” is changed to “Fig. 7”
pg.8875, line 24 (MS: pg.23, lines 3-4): “Fresh soot and particles dominated by soot have an agglomerate structure (Fig. 8), implying that the particle volume is smaller than expected for spherical particles.” Has been added for clarification.
pg.8875, line 25 (MS: pg.23, line 5): “during ageing” has been added
pg.8875, line 29 to pg.8876, line 11 (MS: pg.23, lines 10-23): “The activation could then be further hindered by hydrophobic material surrounding this site and thereby only part of the soot core would act as CCN (Fig.9a, solid, blue lines). Or, partly hygroscopic material is surrounding the activation site, leading to activation sites including more of the soot core (Fig.9a, dashed, blue lines). As the aerosol age the particles restructure and in the end of the experiments they have become more spherical like (Fig. 9c). The restructuring is due to condensation of organic (SOA) material onto the soot particles in the smog chamber. Additionally, condensing water in the CCNC may enhance the effect of restructuring. The

particles become more compact and spherical like with increasing amount of organic (SOA) coating material. This is due to a combination of two effects: SOA material filling the void spaces (Nakao et al., 2011) and restructuring of the agglomerates. The changed morphology due to for example SOA condensation is illustrated in Fig. 5…"

is changed to:

"If most of the particles' soluble material is excluded from the droplet at activation (Fig. 9a, solid, blue lines), due to formation of several smaller separate droplets, the critical supersaturation will be higher than expected assuming that all soluble material is in the same droplet and involved in the activation. In contrast, if droplet growth start at different sites successively incorporating more and more of the particle and growing to form one single droplet (one of these sites is illustrated in Fig. 9a, dashed, blue lines) all soluble material in the particle is involved in the activation.

The particles become more compact and spherical like with increasing amount of organic (SOA) coating material (Fig. 9c). This is due to a combination of two effects: SOA material filling the void spaces (Nakao et al., 2011) and restructuring of the agglomerates. The restructuring is due to condensation of organic (SOA) material onto the soot particles in the smog chamber. Additionally, condensing water in the CCNC may enhance the effect of restructuring. The changed morphology due to for example SOA condensation is illustrated in Fig. 6…"

pg.8876, line 29 (MS: pg.24, line 7): "collapse in" is changed to "restructure to", for clarification

pg.8877, lines 6-16 (MS: pg.24, lines 12-20): Trying to streamline the results section the following sentences: "However, for a given $mf_{SOA(APM)}$ these particles (FSP2) need a slightly higher supersaturation for activation than the soot agglomerates with larger primary particles (compare diamonds with circles and/or triangles, d_m = 150 nm, in Fig. 12). The indicative results of a morphology dependency of the activation properties could possibly be explained by a slightly more expansive collapse of the FSP into smaller sizes. However, a particle with smaller d_{pp} (FSP2) is probably activating at a lower supersaturation due to a larger $mf_{SOA(APM)}$ and hence a smaller $mf_{BC(APM)}$, for a certain d_{ve}, than a particle with larger d_{pp} (DEP and FSP1) (Fig. 10). For the same $mf_{SOA(APM)}$ on the other hand, a particle with larger d_{pp} will activate at a lower supersaturation due to a larger size of the agglomerate (d_{ve}) and thereby also containing more water-soluble molecules than a particle with smaller dpp (Fig. 12)."

are changed to:

" These particles (FSP2) are probably activating at a lower supersaturation due to a smaller mass of soot for a given dm and hence a larger SOA mass, than a particle with larger d_{pp} (DEP and FSP1) for a certain d_{ve}. However, for a given $mf_{SOA(APM)}$ the same particles (FSP2) need a slightly higher supersaturation for activation than the soot agglomerates with larger primary particles (compare diamonds with circles and/or triangles, d_m=150 nm, in Fig. 11). For the same $mf_{SOA(APM)}$, a particle with larger d_{pp} will activate at a lower supersaturation due to a larger size of the agglomerate (d_{ve}) and thereby also containing more water-soluble molecules than a particle with smaller d_{pp}."

pg.8877, line 19 (MS: pg.24, line 23): "Sect. 2" is changed to "Sect. 3"
pg.8877, line 21 (MS: pg.24, line 25): "(Fig. 11)" is changed to "(Fig. 12)"

pg.8878, line 2 (MS:pg.25, line 2): "(DMA-TD-APM)" is added

pg.8878, line 4 (MS:pg.25, line 3): "…-function…" is added
pg.8878, line 6 (MS:pg.25, line 5): "(13) and" has been added to the equation reference within the brackets: "Eq. (13) and (14)"
pg.8878, line 6 and 8 (MS:pg.25, line 5 and 8): "Fig. S1" is changed to "Fig. S 2"
pg.8878, lines 12-18 (MS:pg.25, lines 11-14):"To account for the change in morphology in the model, the volume equivalent diameter (d_{ve}) has been used. The volume equivalent diameter ($d_{ve,measured}$) has been derived from measurements of the mass-mobility relationship (DMA-APM). To improve the κ-Köhler model a fit-function has been derived to calculate d_{ve} from the mf_{SOA}(APM) and d_m. d_{ve} (in nm) for a diesel exhaust particle (DEP) with a primary particle diameter (d_{pp}) of ~28nm, from a Euro II vehicle can be approximated by (Supplement, Fig.S 2): "
is changed, due to recurrence in the previous paragraph, to:
"The volume equivalent diameter (d_{ve}) accounts for the change in morphology in the model, and can for a diesel exhaust particle (DEP) with a primary particle diameter (d_{pp}) of ~28 nm, from a Euro II vehicle be approximated by (in nm) (Supplement, Fig. S 2):"

pg.8878, line 29 to pg.8879, line 1 (MS: pg.25, line 27): "(i.e. a smaller d_{pp} require a slightly higher s_c)." Is erased.
pg.8879, line 1 (MS: pg.25, line 27): "Fig. 12" is changed to "Fig. 11"

pg.8879, lines 5, 7, 9 and 10 (MS:pg.26, lines 2, 5, 6 and 8): "Fig. 11" is changed to "Fig. 12"
pg.8879, lines 7 and 9 (MS:pg.26, lines 5 and 6): "yellow" is changed to "pink", due to changed colour in the figure as requested by referee #2.
pg.8879, line 22 (MS: pg.26, line 19): "surface" is changed to "sites"
pg.8879, line 29 (MS: pg.26, line 24): "Also, the condensing organic material may not be evenly distributed, and therefore only acting as activation spots." Has been erased due to repetition.

pg.8880, line 19 (MS: pg.27, line 10): "Fig.11" is changed to "Fig.12"
pg.8880, line 24 (MS: pg.27, line 14): "due to" is changed to "consistent with"

pg.8881, lines 1-6 (MS: pg.27, line 20):" In summary, the slightly processed soot particles need a higher supersaturation than predicted with all models. The slightly processed soot particles are probably hindered by e.g. shape effects and/or hydrophobicity of the SOA content, or only part of the particle contributes to the activation, as discussed before. As just mentioned, POA is treated as hydrophobic and SOA as hygroscopic, in the model, which might be too much of a simplification." Has been erased due to repetition.
pg.8881, lines 14-15 (MS: pg.27, line 28):"This should be studied more thoroughly in the future." Has been erased.
pg.8881, lines 19 (MS: pg.27, lines 31-32):"…assumed in calculations…" have been added and "…and calculations…" has been erased, new sentence as follows: "κ values for ambient particles show high variation depending on content assumed in calculations and instrument used for observations, …"

pg.8882, lines 15 (MS: pg.28, line 23):"0" is added to the M_{SOA} value, should be "±0.020 kg mol^{-1}"
pg.8882, lines 21 (MS: pg.28, line 29):"for" has been erased
pg.8882, lines 22 (MS: pg.28, line 30):"during activation" has been added

5.4 Uncertainties

pg.8883, line 14 (MS: pg.29, line 18): “Fig. 12” is changed to “Fig. 11”
pg.8883, line 22 (MS: pg.29, line 25): “the newly developed” are added and “are” is changed to “is”: “During measurements using the newly developed SFCA the aerosol is subjected to the same temperature…”
pg.8883, line 25 (MS: pg.29, line 28): “original” is added

pg.8884, lines 3 and 10 (MS: pg.30, lines 3 and 10): “…-TD-…” is added, hence: DMA-TD-APM
pg.8884, line 7 (MS: pg.30, line 7): “Sect. 3.1 is changed to “Sect. 4.1”
pg.8884, line 10 (MS: pg.30, line 10):”deviates” is changed to “differs slightly”
pg.8884, line 16 (MS: pg.30, line 15):”s” is erased from “Collections”

pg.8885, lines 6-7 (MS: pg.31, line 2):” Hence, the error and/or dislocation are the same for both empirical and modelled results.” Is erased due to repetition.

6 Conclusion

pg.8886, line 8 (MS: pg.31, line 28): “…size (III) and…” is erased.
pg.8886, line 12 (MS: pg.32, line 1): “relative” is added
pg.8886, line 13 (MS: pg.32, line 2): “of the primary particles” is added and “primary” is changed to ”fresh”, sentence as follows: “The results also indicate that the size of the primary particles and morphology of the fresh soot core might be of importance.”
pg.8886, line 17 (MS: pg.32, line 6): “…with regard to the…” is changed to “…compared to particles with larger d_{pp} at certain…”, whole sentence as follows: “On the other hand, aggregates with smaller d_{pp} activate at lower supersaturation compared to particles with larger d_{pp} at certain volume equivalent diameter (d_{ve}). “

pg.8887, line 9 (MS: pg.32, line 28): “The κ_{SOA}-value…” is changed to “Modelling of κ_{sum} (Eq. (7)) based on $\kappa_{\mathrm{SOA}}=0.13$…”
pg.8887, line 19 (MS: pg.33, lines 4-5):”…only parts of the coated soot particle are surface active…” is changed to “…unevenly distributed material…” for clarification

pg.8888, line 6 (MS: pg.33, line 21):”also” is added
pg.8888, line 7 (MS: pg.33, line 22): “previous studies” is exchanged for the reference “according to Londahl et al. (2009).” as requested by the referee.
pg.8888, line 8 (MS: pg.33, line 22): “This has not been studied here but should be subjected for further investigation.” Has been erased.

References

Added:

Donahue, N. M., Robinson, A. L., and Pandis, S. N.: Atmospheric organic particulate matter: From smoke to secondary organic aerosol, Atmos. Environ., 43, 94–106, doi:10.1016/j.atmosenv.2008.09.055, 2009

D. Liu, J. Allan, J. Whitehead, D. Young, M. Flynn, H. Coe, G. McFiggans, Z. L. Fleming, and B. Bandy. ”Ambient black carbon particle hygroscopic properties controlled by mixin state and composition”. Atmos. Chem. Phys., 13, 2015-2029, 2013, doi:10.5194/acp-13-2015-2013

Rissler, J., Nordin, E.Z., Eriksson, A.C., Nilsson, P.T., Frosch, M., Sporre, M.K., Wierzbicka, A., Svenningsson, B., Löndahl, J., Messing, M.E., Sjogren, S., Hemmingsen, J.G., Loft, S., Pagels, J.H., Swietlicki, E. "Effective density and mixing state of aerosol particles in a near-traffic urban environment", Environ Sci Technol. 2014 Jun 3;48(11):6300-8. doi: 10.1021/es5000353. Epub 2014 May 14.

Erased:
Petters, M. and Kreidenweis, S. "A single parameter representation of hygroscopic growth and cloud condensation nucleus activity." Atmospheric Chemistry and Physics 7, no. 8 (2007b): 1961-1971. – This reference is occuring twice, the ”a” should be erased from the one still in the list.

Tables
pg.8899: Table 1 and Table 2 switch place, due to new order of the sections in the manuscript.

Table 2 (changed to Table 1)
pg.8900, lines 1 (MS: pg.52, lines 1-4): "…of the dry particle mobility diameter ($d_{m,dry}$), the primary particle diameter (d_{pp}), the critical supersaturation measured by the two different CCNC instruments (s_c (#52) and s_c (#53)), and CCNC operational mode: conventional (ΔT-stepwise) or scanning flow (SFCA)." Has been added to the table caption to clarify the abbreviations as requested by the referee (#1). Also, the abbreviations are found in the Nomenclature (Table A1).

pg.8901, Table A1. Nomenclature
(MS: pg.47, line 13): "$d_{m,dry}$ … Mobility diameter of the dry particle" is added.
(MS: pg.47, line 14): "DMA-TD-APM … Differential mobility analyser-Aerosol particle mass analyzer with a thermodenuder" → adding "TD" and "with a thermodenuder"
pg.8902, Table A1. Continued (MS: pg.49, line 3): "s_c (#52) … Critical supersaturation measured by instrument no. 52" is added.

Figures
The Figure order has been changed upon request from the referees to streamline and strengthen the results section, and not making the reader go back and forth as much as before, as follows:

- Figure 2-6 are now Figure 3-7
- Figure 7a and b are now Figure 2a and b
- Figure 11 and 12 have been swopped

Figure 1, pg. 8903, line 1 (MS: pg.53)
A small change in the figure is made, adding a DMA and an additional CCNC to illustrate the SFCA operational mode better.
Figure caption, pg. 8903, line 5 (MS: pg.53, lines 6-8): "A Cloud Condensation Nucleus Counter (CCNC, DMT 100) measured the cloud-activation properties, while the Aerosol Particle Mass Analyzer (APM, Kanomax Japan 3600) characterized the particle mass-mobility relationship."
is exchanged for:
"Two Cloud Condensation Nucleus Counters (CCNC, DMT 100), running in parallel in

SFCA operational mode, measured the cloud-activation properties (for all experiments except DEP1). The Aerosol Particle Mass Analyzer (APM, Kanomax Japan 3600) characterized the particle mass-mobility relationship.”

Figure 4 (before Figure 3), pg. 8905 (MS: pg.57, line 1)
The yellow colour has been darkened.

Figure 5 (before Figure 4), pg. 8906 (MS: pg.58, line 11): “Sect. 4.1” is changed to “Sect. 2.1”

Figure 9 pg.8911, line 3 (MS: pg.62 , lines 4-6)
Figure caption is changed to clarify about the activation sites: “Possible activation sites in hydrophobic surrounding (solid, blue circles) and more hygroscopic activation sites (dashed, blue circles) are illustrated in (a) with diameters of…”
is exchanged for
“In (a) an activation site excluding most of the particles’ soluble material in the droplet at activation is illustrated (solid, blue circles), as well as a site with more material included (dashed, blue circles), for the diameters…”

Figure 10, pg.8912 (MS: pg.63 , line 1)
The yellow colour has been exchanged for pink.

Figure 11 (before Figure 12), pg.8914 (MS: pg.64, line 1)
The yellow colour has been exchanged for pink.
Figure caption, line 8: “yellow” is exchanged for “pink”

Figure 12 (before Figure 11), pg.8913 (MS: pg.65, line 1)
The yellow colour has been exchanged for pink.
Figure caption, line 10: “(13) and” is added to the equation reference within the brackets “(Eq. (13) and (14))”

Supplement
pg.1 and pg.3: The figures are swopped, due to changed placing of the Sections in the paper. (Figure S 1 and S 2 have swopped places).

pg.2, line 1: Table S 1 with caption is added, with information of the DMA-APM operational details:
Table S 1. An example of the DMA-APM settings (without the thermodenuder) used during DEP2. Shown here is the particle mobility diameter (d_m), the peak of the mass distribution for the fresh ($V_{\text{fresh soot}}$) and processed soot ($V_{\text{processed soot}}$), the rotational speed and voltage scan-range of the APM, the voltage steps during scanning and the resolution parameter for the fresh ($\lambda_{\text{fresh soot}}$) and the processed soot ($\lambda_{\text{processed soot}}$).

d_m (nm)	$V_{\text{fresh soot}}$ (V)	$V_{\text{processed soot}}$ (V)	APM Rotational speed (rpm)	APM Voltage scan-range	$\lambda_{\text{fresh soot}}$	$\lambda_{\text{processed soot}}$
90	50	100	5500	20-160	0.062	0.125

150	140	420	5000	40-400	0.074	0.223
200	180	620	3000	50-500	0.061	0.210
300	260	900	3000	500-2000	0.049	0.170

Table numbers are changed, due to the added table. Both in Table caption and in text, marked with yellow.
pg.4, line 8:”S1” is exchanged for “S 2”
pg.4, line 14 (Table caption):”S1” is exchanged for “S 2”
pg.5, line 4 and 14:”S1” is exchanged for “S 2”
pg.6, line 5:”S2” is exchanged for “S 3”
pg.6, line 12 (Table caption):”S2” is exchanged for “S 3”
pg.8, line 20:”S3” is exchanged for “S 4”
pg.8, line 24 (Table caption):”S3” is exchanged for “S 4”
pg.9, line 14:”S4” is exchanged for “S 5”
pg.9, line 16 (Table caption):”S4” is exchanged for “S 5”
pg.11, line 13:”S2” is exchanged for “S 3”

pg.9 line 14: “…(listed in Table S 5).” is added to the sentence. → whole sentence after Eq.S 2: “where α_i is the mass-based stoichiometric yield of the oxidation product i, K_i is its gas-particle partitioning equilibrium constant, and Mo is the total particulate organic mass concentration (listed in Table S 5).

pg.15, line 1: The yellow colour is changed to dark pink in Fig. S 7.
pg.15, line 4: An addition has been made to the figure caption: “Colour code with respect to particle mobility diameter (d_m) and different markers with respect to type of soot generator (experiment FSP1, 2 or DEP2). “