We thank the reviewer for the comprehensive feedback on our work. With the help of the reviewers' comments we greatly improved the understandability of our work and made it more accessible to a broader audience. Detailed answers to the individual comments are given below. For clarity, the reviewers' comments are written in black, and our response in red. Texts from the old version of the manuscript are typed in green and texts from the revised manuscript in blue.

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General comments:

This manuscript presented an improved experimental approach to perform atmospheric oxidation of soot particles using a Continuous Flow Stirred Tank Reactor (CSTR), which enables extended sampling time within a small-size conventional aerosol chamber. A new metric of activation time (*tact*) was developed to characterize the change of activated fraction (AF) in different regimes (i.e., filling, steady state, and flushing) for soot particles following heterogeneous ozone oxidation. Good agreements between theoretical calculations and parameterized CCN activities using *tact* were achieved for their experimental data. The *tact* concept was also applied into some previous studies with continuous flow chambers. Discrepancies in the CCN activity of BES particles can be better explained with considering *tact* and residence time distribution, in comparison to those initially interpreted by the bulk H/C and O/C ratios, which couldn't fully characterize the detailed change in particle chemical compositions. This work is worth further application in atmospheric sciences, yet some details and interpretations could be clarified, reorganized, and improved accordingly. I would recommend for the final publication in AMT upon major revisions, as detailed below.

Major comments:

1. In the motivation section (Page 2, Line 19): The "non-gradual transition" case of CCN activation suddenly appeared, with no prior introduction or definition of this new concept (instead, which was included in Sect.3). This content seemed to be disconnected with the information detailed in the last sentence, and I didn't catch the importance/necessity of developing a mathematical analysis for the non-gradual transitions in the following statements.

We understood that the wording we used does not precisely describe to what kind of changes we refer to. Our approach to improve the understandability is to add synonyms commonly used in the atmospheric science community. Additionally we added examples to illustrate this concepts. For the examples we chose common processes investigated by the atmospheric science community. Nevertheless, these concepts are not limited to atmospheric science and can be applied in different fields as well.

In case of the term "non-gradual" we refer to changes like phase-transitions where a property changes step-wise. This is the opposite of a gradual or continuous change of a property. An example would be the freezing of water. Below or above 0°C the density of liquid water/ice changes gradually with the temperature. At 0°C the density does not change gradually but changes step-wise by jumping between 0.92 g/cm^3 and 1.00 g/cm^3 . To clarify what is meant by "non-gradual" we extended the introduction of this phrase and added "step-wise change" and "transition between binary states" as alternative explanations. "Transition between binary states" hereby means that a system/particle can be described by two distinct states. Either a droplet is liquid or frozen. A transition from one state to another can be described as "non-gradual" as well.

We added a list of possible transition that can be described as "non-gradual", "step-wise change" and "transition between binary states" (P2 L26-39)

> Such transitions in binary systems are step-wise, also referred to as non-gradual changes in a particle property, such as:
>
> 1) Freezing of a water droplet: Step-wise and therefore non-gradual change in the particle density; the water is either in liquid or solid state.
> 2) Deliquescence of soluble aerosol particles: The particles show a step-wise i.e. non-gradual increase in diameter.
>
> Binary particle properties are not necessarily intrinsic particle properties, but can also be defined by the measurement protocol.
>
> 3) CCN-activity: The chemical and physical properties of an aerosol particle can vary, but the particle is either CCN-inactive or CCN-active at a defined super saturation (SS).
> 4) Growth beyond a threshold: Condensational growth of an aerosol particle leads to a continuous and gradual increase of the particle diameter. A binary system can be defined by introducing a threshold diameter that can be arbitrarily chosen. The aerosol particle is either smaller or larger than this defined threshold diameter. The same holds true when particles are separated e.g. in aerosol impactors.
>
> Therefore, the concept of non-gradual transitions/transitions within binary systems can be used to describe a multitude of changes in particle properties.

Such transitions between binary states are a necessary requirement for the analysis of data from experiment conducted in a CSTR aerosol chamber. Some of the above mentioned transitions are not triggered by a change in the ambient conditions but by a change of the particle itself e.g. due to aging. Aging processes show typically a time-dependency. We focus on the time needed to modify an aerosol particle to such a degree that it changes its state in a binary system. The example on which we focus is the CCN-activation of soot particles due to oxidation with ozone. Initially, a single soot particle is CCN-inactive at a given super saturation. After a certain aging time it becomes able to accumulate water under super saturated conditions. The chemical modification of the soot particle is a continuous process. The changing CCN-activity is not a continuous process. A particle is either CCN-inactive or CCN-active. The transformation from CCN-inactive to CCN-active is hereby a step-wise change in the particle properties.

The authors have introduced the concept of CSTR and suggested that *"The steady state in the CSTR is characterized by constant concentration of all compounds and constant reaction rates."*. It is a bit confusing that how the assumed "perfectly internal mixing" is achieved, even if without considering the influences of particle wall loss and coagulation during different experimental regimes.

The steady state in CSTR is indeed characterized by constant concentrations. All processes and chemical reactions proceed, but new compounds are constantly introduced to the CSTR as well as old products are continuously removed from the CSTR. After a certain time span the CSTR reaches an dynamic equilibrium and therefore the concentration do not change anymore. The same is valid for oxidation flow reactors like the Potential Aerosol Mass chamber. These kind of aging chamber also operate in steady state mode. In contrast to that, environmental aging chamber operated in batch mode and are not continuously filled with new compounds. These type of chambers have constantly changing conditions.

Particle losses to the chamber wall do occur in CSTR-experiment as well, but due to the constant feed-in of fresh aerosol the Particle-wall-loss-rate reaches a constant speed once the CSTR is in its dynamic equilibrium/ steady state.

The "perfectly mixed" refers to the active mixing inside the CSTR. We used a fan to mix the freshly introduced aerosol with the aerosol that entered the CSTR before. A real CSTR is theoretically never perfectly mixed. However, a perfect internal mixing can be extremely well approximated by actively stirring the aerosol inside the chamber.

Section changed from: (old P2 L6-8)

> The continuous flow stirred tank reactor (CSTR) describes an aerosol chamber, which is continuously filled with an aerosol flow constant in composition over time. The aerosol inside the CSTR is perfectly mixed, therefore a mix of aged and unaged aerosols is continuously extracted from the CSTR for analysis.

To: (new P2 L13-19)

> The CSTR approach describes an aerosol chamber, which is continuously filled with an aerosol flow constant in composition over time. The volume of the CSTR is actively stirred in order to achieve a homogenous aerosol mixture. Due to the mixing, the aerosol that is continuously extracted for analysis consists of a well-defined mixture of aerosols at different aging stages.

How should readers understand the "constant concentration of all compounds" during aging reactions in the CSTR, where the corresponding compositions/concentration of reactants/products are supposed to vary with such processes?

As described above the CSTR can reach a dynamic equilibrium. In this state, particles do react, get lost to the chamber walls, coagulate and so on. Due to the constant feed-in of new aerosol and pumping out old aerosol the overall concentration stays constant over time. We greatly expanded the description of the CSTR and the ongoing processes. (new: P4 L7 – P5 L15)

Filling regime

As the CSTR volume is initially sample free, the aerosol particle concentration in the CSTR increases continuously during the filling regime until it reaches a stable concentration. The aerosol particle concentration ($[A_{CSTR}(t)]$) at any point in time can be calculated as a function of the experimental duration (t) by eq. (2), where ($[A_{feed\text{-}in}]$) is the aerosol concentration in the feed-in flow.

$$[A_{CSTR}(t)]=[A_{feed\text{-}in}]\cdot\left(1-e^{\left(\frac{-t}{\tau_{CSTR}}\right)}\right) \tag{1}$$

We define that the CSTR reaches steady state conditions when the difference between $[A_{CSTR}(t)]$ and $[A_{feed\text{-}in}]$ is smaller than the resolution of the analytical instruments deployed. To standardize the time period, we chose the fourfold mean residence time (4τ criterion) as reference point for the start of the steady state in this publication. At this point the difference between $[A_{CSTR}(t)]$ and $[A_{feed\text{-}in}]$ is less than 2 % which is lower than the resolution of most aerosol particle counters (Mordas et al., 2008).

Steady state

The steady state is in fact the part of the filling regime where the CSTR is in a dynamic equilibrium. All processes and reactions continue but the concentrations of all compounds remain constant over time. In theory, this operation point can be maintained for an infinite experimental duration. Be aware that this does not mean that an infinite degree of aerosol aging can be achieved. In steady state the experimental duration is decoupled from the particle age, which is in contrast to experiments in batch-chambers but similar to OFR experiments. As a result of the continuous feed-in and flush-out flow, different aerosol fractions that enter the CSTR at different times are present simultaneously, resulting in a residence time distribution (RTD). In CSTRs the RTD can be described by eq. (3) and is plotted in Fig. 1 (solid black line – labeled with "steady state"). With an increasing individual residence time the fraction of aerosol particles declines exponentially. The individual residence time of a specific particle fraction is indicated by the color-coding in Fig. 1. The actual number of particles within an individual particle fraction at a specific residence time can be calculated by integrating RTD over time (eq. (4)). This leads to the residence time sum distribution RTD_{sum} represented by the colored area under the curve. Note, while we choose $RTD(t)$ and $RTD_{sum}(t)$ for a more intuitive denotation, generally E(t) and F(t), respectively, are the official formula symbols especially in the engineering community (Levenspiel, 1999).

$$RTD(t)= e^{\frac{-t}{\tau_{CSTR}}} \tag{2}$$

$$RTD_{sum}(t)= \int_0^t RTD(t)\,dt =1-e^{\frac{-t}{\tau_{CSTR}}} \tag{3}$$

Flushing regime

> From the point in time that no fresh aerosol but only particle free air is added the CSTR is operated in the flushing regime. This operation mode can be considered similar to the operation of batch-aerosol chambers as in both cases the aerosol is flushed out continuously.
>
> The initial RTD at this switching point (t_{switch}) and therefore the ratio of young to old aerosol fractions is preserved throughout the entire flushing duration. Nevertheless, the individual residence time of every single aerosol fraction rises with flushing duration. In other words: All particles age simultaneously. Figure 1 illustrates how the RTD changes in the flushing regime. Note, the time on the x-axis is plotted as dimensionless time in multiples of τ. Each color in the area represents an individual aerosol fraction with a corresponding residence time. Blue stands for the lowest and red for the highest residence times. The dashed black curve labeled "steady-state" represents the RTD in steady state while the other curves show the RTDs for additional time increments after the flushing regime has been initiated (t_{switch}). For example, the area und the grey curve labeled "+1 τ" represents the RTD 1 τ after initiation of the flushing regime. The grey dashed line stands for the activation time t_{act}, a threshold time that will be introduced later. Here it marks a threshold time. With increasing flushing time, the fraction of aerosol particles that have an individual residence time higher than this threshold time increases. From some point in time on all particles have crossed this threshold time as is the case for the particles under the light grey curve at "+2 τ" after t_{switch}.

Another question is about the configuration of the CSTR in this study: did the authors use a real CSTR device for their experiments or not? what kinds of equipment (and how) were actually coupled with the CSTR, in addition to a CCN counter which enables the CCN activation measurements (i.e., the AF results) of aged soot particles?

The CSTR describes a reactor concept that is continuously filled with educts while products are constantly removed. The compounds inside the CSTR are ideally perfectly mixed, which is realized by actively stirring the compound with e.g. a fan.

In the context of atmospheric science, every environmental aging chamber can be operated as a CSTR. The only technical requirement is the presence of e.g. a fan to stir the aerosol. In our experiments we operated a 2.78m^3 steel chamber in CSTR-mode .

The description of the aerosol chamber used was extended from: (old P14 L20 - 21)

> In the laboratories at ETH Zurich we performed aging experiments in a 2.78 m^3 stainless steel aerosol chamber run in CSTR mode.

To: (new P15 L20-29)

> In the laboratories at ETH Zurich we performed aging experiments in a 2.78 m^3 stainless steel aerosol chamber operated in CSTR mode. A detailed description of the chamber can be found in Kanji et al., (2013). The chamber was actively mixed with a fan, but had no further features to enhance mixing e.g. baffles. All instruments were

> connected to the chamber with stainless steel tubing with 4 mm inner diameter. Since the maximal tubing length from the CSTR chamber to the analysis instruments was 3 m the impact on the overall residence time is negligible.
>
> We investigated the change in CCN-activity of soot particles rich in organic carbon due to heterogeneous ozone oxidation. The soot particles were generated with the miniature Combustion Aerosol STandard (miniCAST, Model 4200, Jing Ltd., Zollikofen, Switzerland) which is propelled with propane and operates with a laminar diffusion flame. The miniCAST was operated under fuel-rich conditions (set point 6 according to the manual) in order to generate a soot which was rich in organic compounds (fuel-to-air ration: 1.03).

The aerosol chamber used is a "real CSTR" which and is very close to an "ideal CSTR". We added a section where we explicitly point out that in terms of changes in the particle number concentration no deviation between our "real CSTR" and an "ideal CSTR" could be detected. (new P16 L10-20)

> In the flushing regime the particle number concentration declines exponentially in both experiments. Eq. (5) describes the ideal/theoretical evolution of the particle number concentration in the flushing regime when taking the hydrodynamic residence time τ_{CSTR}according to eq. 1 into account. In the ideal case the decay is solely caused by the flushing process. In reality, the decay is a combination of flushing as well as additional particle losses e.g. wall losses or coagulation. Therefore, the real residence time can be obtained by fitting equation 5 to the experimental data after rearrangement for τ, to which we refer to as τ_{flush} from now on (Kulkarni et al., 2011). In both experiments τ_{flush} coincides at 104 min, which is lower than the hydrodynamic residence time τ_{CSTR} of 111 min. In other words, the particle concentration declines faster than expected. This difference is caused by particle losses to the chamber wall, which acts as an additional particle sink parallel to flushing and reduces the particle lifetime. Nevertheless, statistical analysis of the experimental data results in purely statistical noise centered on the fitting curve used to determine τ_{flush}. This indicates that in terms of mixing no difference between an ideal CSTR and the aerosol chamber used here can be detected with the applied instrumentation.

We conducted a broad range of experiments we were investigated the impact of different aging conditions onto different soot particles. We investigate parameters like single particle mass, hygroscopicity, chemical composition, Ice nucleation activity as well. Results from these experiments will be published shortly. We refrain from mentioning these measurements in order to not distract the reader, to keep the focus on the CSTR-approach and on the applicability of this approach.

Corresponding details are suggested to be provided especially
for those who are unfamiliar with such systems. From my perspective, the
organization of this section could be improved for better delivery of the key points.

We expanded and rewrote the introduction section and made a clearer distinction between the application of the CSTR approach for atmospheric experiments, the development of a CSTR-specific mathematical framework, the newly developed t_{act}-concept, as well as the application of the t_{act}-concept to other continuous flow steady state chambers, namely OFRs. (new: P2 L40- P4 L7)

> In the following, we discuss a theoretical basis for the analysis of time-dependent changes in binary systems within well-mixed continuous flow aerosol aging chambers (CSTR-approach). We developed a mathematical framework which allows the retrieval of characteristic parameters from the system of interest (e.g. CCN activity) and which allows for the calculation of the parameter of interest throughout the entire duration. Key element in this framework is the activation time (t_{act}) which marks the time after which the individual aerosol particle undergoes a transition within a binary system. We start by introducing an idealized system in which t_{act} can be described by a single number and proceed to a more realistic setting in which we incorporate a distribution of particles with different individual t_{act}'s (activation time distribution, P(t_{act})). Further, we test the t_{act}-concept on real experimental data and finally apply it to other types of continuous flow aging chambers such as OFRs. We show that application of the t_{act}-concept is capable of giving new insights to ORF data and further significantly improves the understanding of discrepancies in experimental results obtained in intercomparison studies Lambe et al., (2011) with different reactors such as the Potential Aerosol Mass Chamber (PAM) chamber and the Toronto Photo-Oxidation Tube (TPOT).

Furthermore, we expanded the section where we introduce the batch-mode aerosol chambers, the OFRs and compare both types with the CSTR. (new P3 L17-39)

> In an aerosol chamber operated in batch mode, the reaction volume is first filled with the sample aerosol as fast as possible to achieve high homogeneity of the sample. After the desired start concentration is reached further addition of the sample aerosol is stopped and the aging is initiated e.g. by addition of the oxidant. This point in time is generally defined as the start of the experiment and referred to as $t = 0$. Data acquisition of the ageing sample takes place while the reaction volume is flushed with sample-free gas. The composition throughout the chamber is homogeneous but evolving in time, therefore no steady state conditions are ever achieved. This concept is used to operate many large scale environmental chambers (Cocker et al., 2001; Leskinen et al., 2015; Nordin et al., 2013; Paulsen et al., 2005; Platt et al., 2013; Presto et al., 2005; Rohrer et al., 2005).
>
> A PFR is a steady state reactor in which no mixing along the flow path (axial mixing) but perfect mixing perpendicular to the flow (radial mixing) takes place. Further, a continuous feed-in of reactants and withdrawal of sample take place at equal flow rates simultaneously. This results in a constant composition of the output solely depending on the residence time within the reactor. This ideal system is approximated by many Oxidation Flow Reactors (OFR) e.g. PAM chamber (George et al., 2007), TPOT Chamber (Kang et al., 2007), Micro Smog Chamber (MSC; Keller and Burtscher, 2012), or the TUT Secondary Aerosol Reactor (TSAR ; Simonen et al., 2017). The main difference between an ideal PFR and real OFRs is that in OFRs significant but unintentional mixing of the aerosol along the flow path takes place (Mitroo et al., 2018). Therefore, OFRs show a significant residence distribution.

> The CSTR is a steady state reactor with a constant reactant feed in and sample withdrawal as well but opposite to OFRs, the volume is actively stirred to achieve a homogeneous composition throughout the reactor volume. Due to the active mixing, sample stream composition and conditions are the same as within the entire chamber volume. The concept of the CSTR requires perfect internal mixing, which cannot be achieved in real systems. However, due to the good miscibility and low viscosity of gases and the aerosol particles being homogenously dispersed, it is possible to achieve a degree of mixing which is very close to a perfectly mixed system. Especially in the case of mimicking atmospheric processes, residence times of several hours are achieved. Compared to that, the time needed for dissipating all gradients, which is in the order of seconds to minutes, can be considered small.

2. The Section 6, especially the last paragraph of which, is quite confusing. It is good to see the application of the activation time concept (*tact*) into data interpretation of previous chamber studies, with improved agreements among different datasets. Nevertheless, there are several concerns need to be addressed. First of all, the previously used chambers such as PAM, they are actually not CSTR or far from the ideal mixing condition during oxidation. As a result, how can you simply apply the *tact* or RTD concept for CSTR system into the data interpretation of OFR/PAM reactors? Necessary information is needed to clarify this point.

The PAM and TPOT chambers are indeed no CSTRs, are not internally well-mixed and therefore cannot be described with the here introduced CSTR-specific mathematical framework. However, they show a significant residence time distribution due the mixing along the flow path, therefore it is possible to apply the t_{act} concept. An RTD means that the aerosol particles that leave the OFR stayed inside the chamber for different individual times. If the measured AF behind the OFR is 0.3, we raise the question which particles of the whole aerosol particle distribution are the CCN-active particles. The t_{act} concept implies that only the oldest 30% of the particles are CCN-active and the youngest 70% are CCN-inactive. The time that separates the youngest and CCN-inactive 70% from oldest and CCN-active 30% is the t_{act} (necessary aging time).

In principle it is also possible that young and old particle activate equally well, however this seems to be unlikely for the here discussed BES-particles. Furthermore even this behavior could be captured by the t_{act}-distribution. In this unlikely case the activation time distribution would be a horizontal line ($P(t_{\mathrm{act}}) = 0.3$) and neither a peak nor a Gaussian shaped distribution.

In this manuscript we define 2 scenarios that combine this t_{act}-concept with the RTD reported by Lambe et al. (Figure 7) In the first scenario (High-OH) we calculated the fraction of particles older than 40s in the PAM and TPOT chamber. In the second scenario we did the same with a time of 180s. As can be seen the fraction of particles older than this threshold time t_{act} varies between both chambers. Since only the oldest particle can be CCN-active this leads to different measured AF-values. IN The High-OH scenario, the AF in the TPOT is higher than in the PAM chamber. In the Low-OH scenario The AF in the TPOT is lower than in the PAM chamber. The same trend was reported by Lambe et al.

However this a qualitative application of the t_{act}-concept. We added following section that mentions what would be needed for quantitative application of t_{act} – concept. (new: P20 L23-36)

> Up to now, the discussion did not include many important processes that are relevant in aging chambers e.g. particle wall-interaction, gas-phase-partitioning, fluctuating input concentrations while field measurements, or inhomogeneities inside the OFR. These aspects are important for many processes such as the formation of SOA and can be incorporated to the t_{act}-concept by modifying eq. (13). As the actual calculation requires a multidimensional data array and detailed knowledge about the chamber of interest, this subject matter is beyond the scope of this publication and will not be discussed further. Nevertheless, the overall conclusion is that application of the original/non-adjusted t_{act}-concept can explain why measurements within different OFR chambers agree in parameters, which dependent on the bulk properties of the aerosol particle population (e.g. average O:C ratio) and at the same time disagree in parameters, which are dependent on the condition/status of the individual particle (e.g. CCN-activity). Therefore, we suggest to apply the concept of the activation time t_{act} or the activation time distribution $P(t_{act})$ as metric in addition to calculating average values, such as the global *AF* and OH-exposure if following conditions are met. One, the system or parameter of interest can be described as a binary system and undergoes step-wise / non-gradual transitions such as CCN-activity. Two, the OFR used has a RTD broad enough to influence the outcome. Three, the conditions inside the reactor are either homogeneous or a correction for inhomogeneities (e.g. different oxidants concentrations inside the reactor) is implemented.

Another issue is that discrepancies in CCN activity of SOA formed from chamber oxidation experiments could be influenced by various factors, such as gas-particle partitioning and particle-phase reactions during SOA production as well as liquidliquid phase separation during activation processes. Additionally, the variability in different operation parameters such as relative humidity, initial concentration of VOC precursors, and acidity in the OFR/PAM chamber can affect the SOA formation process even for a same average OH concentration condition, further influencing the subsequent CCN activation process. In this sense, how to evaluate or exclude the impacts of these factors on the agreement of CCN activity (or AF) measurements for different types of OFR or PAM experiments? Namely, how can we confirm that the discrepancies are predominantly introduced by the activation time (or RTD) rather than by the other influencing parameters, although the application of tact can better capture the deviation of CCN activity (likely due to change in chemical compositions) than what the bulk H/C and O/C ratios do? Further discussion is needed to clarify the abovementioned points.

We only refer to the aging of BES-particle in the PAM and TPOT-chamber. Therefore, we do not discuss the application of the t_{act}-concept onto the formation and aging of SOA. We also only apply the t_{act}-concept qualitatively to the results obtained by (Lambe et. al 2011). To identify if the RTD of the two chambers is the only reason for a different measured AF a quantitative analysis would be needed. However, this greatly exceeds the scope this manuscript. Nevertheless, the t_{act}-concept predicts the overall trend in the AF measured downstream both chambers well. This is described in the section above.

The formation and subsequent aging of SOA in OFRs is a complex process and cannot by fully described by the here introduced t_{act}-concept. The main factor that inhibits a straightforward application is that SOA-particles only form in the chamber. As long as the educts are gaseous and therefore fully miscible, no air parcel can be separated from another air parcel by a measurement of the CCN-activity. After particles formed, each particle can have its individual trajectory inside the OFR. Therefore, particles can have different individual aging times and individual degrees of chemical modification. This can be detected by e.g. a CCN-Counter. For gases in an OFR that's not possible. Furthermore, the t_{act}-concept relies on the fact that a single particle is a closed system. If one particle has a high concentration of substance A while another particle has a low concentration of substance A, no exchange processes can dissipate this gradient. In the case of SOA, this can be an invalid assumption. Volatile compounds can evaporate and can condense on other particles and dissipate concentration gradients. An extended framework can potentially capture these processes and the t_{act}-concept has to be part of this.

Other aspects that should be implemented in such extended framework as well, would be the internal inhomogeneities of the OFR. For example, the concentration of OH-radicals inside the OFR is inhomogenously distributed. The OH-concentration for example increases the closer the light sources get. Knowledge of the temperature gradient/profile would also be relevant since the speed of chemical reactions is typically temperature dependent. At this point, we want to mention that every additional variable increases the complexity of the math exponentially. This degree of complexity was a major reason why we favored an internally mixed aerosol chamber. This allowed us to assume homogeneous conditions throughout the entire experiment.

Specific comments:

1. **Abstract**: What does the "non-gradual transitions" refer to here (Line 12)?

We greatly expanded the explanation of the phrase "non-gradual transition" throughout the entire manuscript (see major comment). In the abstract we changed it from: (old: P1 L12)

> We show that this concept can be applied to other systems investigating non-gradual transitions.

To: (new: P1 L14-15)

> This experimental approach and data analysis concept can be applied for the investigation of any transition in aerosol particles properties that can be considered as a binary system

In the last sentence, what specific kinds of "discrepancies" are you suggesting? It is better to clarify these concepts precisely, as which are important points to show the significance and applicability of this study.

We show for the specific example of CCN-activation of photochemically aged BES-particles, that the activation time concept is beneficial for the data-analysis from OFRs. However it is not limited to this, since it is a general concept. These points are now intensively discuss in the expanded manuscript. We refrain from discussing specific discrepancies in the abstract since this would greatly expand the abstract.

Changed from: (old: P1 L13-15)

> Furthermore we show how t_{act} can be applied for the analysis of data originating from other oxidation flow reactors widely used in atmospheric sciences. This concept allows to explain discrepancies found in intercomparison of different chambers.

To: (new: P1 L15-19)

> Furthermore, we show how t_{act} can be applied for the analysis of data originating from other reactor types such as Oxidation Flow Reactors (OFR), which are widely used in atmospheric sciences. The new t_{act} concept significantly supports the understanding of data acquired in OFRs especially these of deviating experimental results in intercomparison campaigns.

2. Page 2, line 8: How is the *"perfectly mixed"* defined here? It is unclear especially to readers those are unfamiliar with the CSTR technique.

To avoid the phrase "perfectly mixed" we mention instead that a homogenous aerosol mixture can be achieved by actively stirring the aerosol inside the chamber. This is a necessary requirement for a CSTR operation. At this point "perfectly mixed" is accurate since we refer to an ideal system. However, this can be confusing as the reviewer pointed out.

changed from: (old: P2 L7-8)

> The aerosol inside the CSTR is perfectly mixed, therefore a mix of aged and unaged aerosols is continuously extracted from the CSTR for analysis.

To: (new: P2 L14-15)

> The volume of the CSTR is actively stirred in order to achieve a homogenous aerosol mixture. Due to the mixing, the aerosol that is continuously extracted for analysis consists of a well-defined mixture of aerosols at different aging stages.

Following which, what do you mean that *"real processes in the atmosphere where aerosols are constantly emitted, mixed and removed"*? Are you sure of the "constantly" condition in the ambient environment? Which specific atmospheric processes have you included in this statement, any references can be provided to support the idea?

As the reviewer pointed out earlier many readers are not familiar with CSTRs. This reference to atmospheric processes shall give the uniformed reader a better understanding what it means to operate an aerosol chamber in CSTR-mode. To the authors knowledge, all other experimental approaches that use flow tube or batch-chamber aim to generate a uniformly aged aerosol. This stands in a strong contrast to the here presented approach, where we are aiming for a non-uniformly aged aerosol output.

This loosely resembles the conditions in the atmosphere. Due to the persistent emission of aerosol into the atmosphere, the ongoing modification and the ongoing removal of aerosols from the atmosphere a mixture of young, medium-aged and old aerosols are present in the atmosphere. Similar to that a freshly produced aerosol is fed-in/emitted to the CSTR. Inside the CSTR, the aerosol is constantly chemically modified and mixed with the fresh aerosol. The aerosol that is removed from the chamber is a mixture of young, medium-aged and old aerosols. We acknowledge that the atmosphere is not a CSTR, however both are comparable in the mixing aspect.

We clarified that we compare the CSTR-approach and the atmosphere in terms of mixing aerosols and measuring a non-uniformly aged aerosol.

Changed from: (old: P2 L8-9)

> This approach is close to real processes in the atmosphere where aerosols are constantly emitted, mixed and removed as well.

To: (new: P2 L16-19)

> From this perspective, the CSTR approach is closer to atmospheric processes than other reactor types as in the real atmosphere except for individual plume emissions aerosols are rather continuously emitted, mixed, and removed. This results in a mixture of aerosols at different aging stages, but of course, the atmospheric mixture is less well defined compared to an aerosol in a CSTR.

3. **Equation 5**: Why is the exponential part not expressed as "$e^{\left(-\frac{t-t_{\mathrm{switch}}}{\tau_{\mathrm{CSTR}}}\right)}$" for the flushing regime? Please check the conversion carefully.

We thank the reviewer for taking the time and checking the equations as well. That was a mistake on our side and we implemented a correction.

Equation changed from

$$[A_{\mathrm{CSTR}}(t)]=[A(t=t_{\mathrm{switch}})]\cdot e^{\left(-\frac{-t}{\tau_{\mathrm{CSTR}}}\right)} \quad (4)$$

To

$$[A_{\mathrm{CSTR}}(t)]=[A(t=t_{\mathrm{switch}})]\cdot e^{\left(-\frac{t-t_{\mathrm{switch}}}{\tau_{\mathrm{CSTR}}}\right)} \quad (5)$$

4. Page 6, line 20: As a crucial parameter introduced in this study, the activation time (tact) for non-gradual transitions was developed. However, what do you mean “If all the other parameters stay constant” during non-gradual transitions, which specific parameters are you referring to?

“If all other parameters stay constant…” refers to the previously mentioned external parameters that can trigger non-gradual/step-wise changes in a particle. We added examples for relevant external parameters. We created this t_{act}-concept based on this assumption since we operated our experiment in a temperature controlled chamber, at a defined RH and at a ozone concentration that was actively kept constant.

Changed from: (old: P6 L20-21)

> If the all other parameters stay constant, while a particles undergoes changes that result in a non-gradual transitions, this transition can be described as a function of time.

To: (new: P6 L26-30)

> We may assume a system in which all external parameters stay constant but the particle itself undergoes a continuous transformation, e.g. due to oxidation. After a certain period of time, this continuous transformation, in this specific case oxidation, can lead to a change in a binary property, e.g. CCN-activity. Ultimately, the step-wise or non-gradual transition is a function of time. We define the required time span (e.g. necessary aging time) that leads to a change in a specific particle property, resulting in a transition in a binary system in another particle property as the activation time (t_{act}).

Is it easy to achieve in practical conditions of laboratory chamber experiments?

This section introduces a theoretical and idealized concept for which constant background concentrations are assumed. Experimental short-comings are therefore not discussed in this section.

Besides that, it is rather easy to keep certain parameters like temperature, relative humidty and Ozone concentration constant in our CSTR-experiments. The fan inside the chamber creates a homogenous atmosphere. The temperature can be actively controlled with a heater/chiller. The ozone concentration was constantly measured and kept stable with a feedback loop that regulated the ozone source. This is an experimental advantage towards many OFRs. Results from these experiments will be published within the next months.

5. Equation 7: I think it should be “$e^{\frac{-t_{switch}}{-\tau_{CSTR}}}$ ” for the flushing regime? Please check the conversion carefully

The initial version of equation 7 is correct. t_{switch} cannot have any influence in the filling regime since its not reached yet.

6. **Equation 8**: Why is the simplified equation not expressed as ‘***tact*** = -ln(AF(t))·τCSTR’? I’m wondering how will the value of AF(t→∞) be, could it be 0 as

suggested by the exponentially decreased curve in Fig.2, or probably approaching 1 like what AF responds when switching to the flushing regime as shown in Fig.3? How should the readers understand the corresponding physical meaning of $AF(t \rightarrow \infty)$ in this steady state condition? Corresponding details are necessary.

The equation 6a. 6b and 7 describes how the AF inside the CSTR evolves over time while filling the CSTR.

$$t \leq t_{act}: \quad AF(t) = 0 \tag{6a}$$

$$t > t_{act}: \quad \mathrm{AF(t)} = \frac{\text{activated particles}}{\text{all particles}} = \frac{\int_{\mathrm{t}=t_{act}}^{\mathrm{t}} \mathrm{RTD(t)}\, dt}{\int_{0}^{\mathrm{t}} \mathrm{RTD(t)}\, dt} = \frac{\mathrm{RTD_{sum}(t) - RTD_{sum}(t=t_{act})}}{\mathrm{RTD_{sum}(t)}} \tag{6b}$$

$$t_{act} = \ln\left(1 - \left((1-AF(t)) \cdot \left(1 - e^{\frac{-t}{\tau_{CSTR}}}\right)\right)\right) \cdot (-\tau_{CSTR}) \tag{7}$$

During this time the particle concentration inside the CSTR is increasing continuously. Parallel to that the *AF* is also changing. After a certain time the changes in the particle concentration become negligible. The CSTR is then in a dynamic equilibrium called “steady state”. In this dynamic equilibrium, aerosol particles still become CCN-active due to aging but already CCN-active particle are also constantly removed from the CSTR by flushing. This state can be maintained for an in theory infinite time span. This means that the experimental duration t grows continuously, but the conditions inside the CSTR are constant. Therefore, the distribution of young, medium-aged and old particle inside the CSTR is constant and decoupled from the experimental duration t. This also means that the AF and the activation time t_{act} calculated from it do not change anymore. The same applies to OFRs which are operated as steady state reactor as well.

In the old version of the manuscript the x-axis in Fig. 2 was labeled with “particle age / min” which was now changed to “residence time / min”. The individual residence time is equal to the particle age. However, this is only mentioned at a later point in the manuscript, which is indeed misleading. The curve in Fig. 2 does therefore not show how particle number concentration that declines with increasing experimental duration. Instead it shows the abundance of particles with a defined residence time (=individual particle age) in the total aerosol particle mixture.

Figure 2 shows the RTD during steady state. The area under the curve represents the total particle population. The colored area is the fraction of particles that is older than the defined $t_{act.}$ This “oldest fraction” is equal the CCN-activated fraction in our model. Since this curve refers to the steady state it does not change over time, but stays constant. Therefore the AF does neither decline exponentially nor does it approach 1, but stays constant with increasing experimental duration. Also, the correlation between the individual necessary aging time t_{act} with the measured AF is not linear but exponential as can be deduced from the different AF-values in Fig.2.

As a result of this, the mentioned equation 8 cannot contain any dependency on the experimental duration t. In fact, equation 8 is derived from eq 7 for (t→∞). This is now stated in the manuscript as: (new: P6 L20 –P7 L1)

> After the conditions in the aerosol chamber reached steady state, the measured *AF* does not change anymore. This is due to the fundamental concept of a CSTR which entails a continuous addition of fresh particles and simultaneous withdrawal of sample at equal flow rates resulting in a dynamic equilibrium and a constant RTD.
>
> To simplify matters, the reason for the constant *AF* within this dynamic equilibrium can be visualized when focusing on three distinct time periods within the continuum of the RTD and thereby on three specific particle fractions. Fraction one is within the right tail of the RTD and consists of particles with a residence time that is above t_{act}. They are only a few compared to the total number of particles and a fraction of these is constantly flushed out with the sample stream. This would lead to a hypothetical reduction of *AF* if not simultaneously the second particle fraction of interest was in the situation to have an individual residence time that is just about to exceed t_{act}. The particles within fraction two are thereby transitioning from the CCN inactive particle fraction within the aerosol chamber to the CCN active particle fraction. The hypothetical loss of CCN inactive particles would lead to an increase in *AF* if not again simultaneously the third particle fraction of interest consisting of fresh and CCN inactive particles was about to be added to the chamber volume.
>
> Due to this dynamic equilibrium, eq. (7) can be simplified to eq. (8) assuming that the experimental duration *t* approaches infinity $\left(\lim_{t\to\infty} eq.\ (7) = eq.\ (8)\right)$
>
> $$t_{act} = -\ln(AF) \cdot \tau_{CSTR} \tag{8}$$

A comparision to the flushing regime in Fig 3 is not useful since this a different operational setting, that is described with its own equations. In the flushing regime the particle concentration and the AF inside the CSTR are changing constantly which is similar to aerosol chamber operated in batch-mode. The equations to describe the changing AF while flushing are introduced in the section “Particle activation during flushing regime”

7. Page 8, line 7: It sounds a bit strange of “global” AF? Is the “global” trying to represent the specific exponentially increased AF inside CSTR or just to show a different AF case with other non-CSTR chamber experiments?

A consequence of the CSTR-approach is that aerosol particles with different individual aging times are present in the chamber at the same time. Each of these aerosol fractions is CCN-active to a different degree. The term “global AF” refers to the combination of all AF-values from different individual aerosol particle fractions combined (=global). This global *AF* is the *AF* that is measured downstream the chamber

The explanation of the global AF in the manuscript was changed from: (old: P8 L7)

> This leads to an exponential increase of the *AF* inside the CSTR (global *AF*) until *AF*=1.

To: (new: P11 L21-22)

> This single value, from now on referred to as global *AF*, represents the average *AF* over all *AF*s of the individual sub-fractions within the population as will be explained in more detail in the upcoming sections.

Further the introduction of the term "global AF" was moved to alater point in the manuscript.
Line 10: ***"… and therefore the global AF only if tact = tswitch."*** Some information was missed in this sentence.

The here present concept assumes that the *AF* inside the CSTR is equal to the fraction of particles older than the threshold time t_{act}. Therefore, the *AF* can be calculated by calculating the fraction of particles older than this time. However, this comes with some difficulties in the flushing regime.

In Figure 1 can be seen how the RTD changes after the particle feed-in is stopped. The whole RTD-curve from steady state is shifted towards longer times. As a result of this, the fraction of particles older than a threshold time increases exponentially. This increase of "old particle fraction" change is captured in equation 9.

$$AF(t)_{\mathrm{flushing}} = \int_{t_{\mathrm{switch}}/\tau_{CSTR}}^{t/\tau_{CSTR}} \mathrm{e}^{\left(\frac{\mathrm{t}-2\cdot t_{\mathrm{switch}}}{\tau_{CSTR}}\right)} \, \mathrm{d}\left(\frac{t}{\tau_{CSTR}}\right) \qquad (9)$$

This equation 9 is the result of centering of the RTD during steady state at t = t_{switch}. Therefore, equation 9 describes which fraction of particles is older than t_{switch}. Since the fraction of particles older a defined threshold time is equal the AF, this equation would describe the changing AF for the special case that t_{switch} and t_{act} are equal. Implementing all other case requires a shift of the starting point of equation 9. This is done by introducing the parameter t_{offset}.

We rewrote the entire section, in order to increase the understandability (new: P9 L7 – P10 L14)

> At the beginning of the flushing regime (t_{switch}), the afflux of fresh particles is stopped and replaced by a particle free gas stream. As the sample extraction is maintained the total particle number (concentration) within the aerosol chamber depletes with time. Nevertheless, the particles that entered the aerosol chamber before t_{switch} continue to age during their increasing individual residence time. This causes a transformation of the RTD along the x-axis/residence time but no transformation of the shape of the RTD as the ratio of particles of different residence time stays constant. This process is indicated by the multiple curves in Fig. 1. The RTD of the particles in steady state is represented by the solid black line. As no fresh particles are added anymore, but the aging of the present particles continues the RTD curve is shifted along the x-axis towards the right.

For example, after a time period equaling the hydrodynamic residence time of the CSTR (+1 τ) the particle RTD is represented by the dark grey solid line. As only the fraction of particles with an individual residence time above t_{act} is CCN active, AF at +1 τ is significantly higher than during steady state. This is indicated by a larger area under the curve that crossed t_{act} (grey dashed vertical line in Fig 1.). Throughout additional flushing time the RTD is shifted further towards longer residence times. At some point all particles have a residence time beyond t_{act}. This means that all particles are CCN-active resulting in an AF of 1 which is for example the case for the particles in the area underneath the light grey curve in Fig 2 (+2 τ. In reality this is not a stepwise process with time increments of 1 τ, but a continuous process that involves an exponential increase of AF inside the CSTR until $AF = 1$.

This change in AF can be mathematically captured. The first step is to derive an equation that describes what fraction of the RTD has crossed the point t_{switch} after flushing has been initiated. Since this not equal to the AF, a second step is needed where an offset-parameter is introduced that converts the "fraction of particles older than t_{switch}" into the AF (=fraction of particles older than t_{act}).

The first step can be achieved by integrating the RTD backwards starting from $t = t_{switch}$. This is an unfavorable approach since it is not compatible with a constantly increasing experimental duration t. This can be avoided, by flipping the RTD horizontally at $t = t_{switch}$ and integrating forward in time from $t = t_{switch}$ to t, which is done in eq. (9). For a simpler integration the experimental duration t, was normalized by dividing it by the hydrodynamic residence time τ_{CSTR}

$$AF(t)_{flushing} = \int_{t_{switch}/\tau_{CSTR}}^{t/\tau_{CSTR}} e^{\left(\frac{t-2\cdot t_{switch}}{\tau_{CSTR}}\right)} \, d\left(\frac{t}{\tau_{CSTR}}\right) \qquad (10)$$

As mentioned before, eq. (9) only describes the fraction of particles that are older than t_{switch}. Since we defined AF as the fraction of particles with an age above the threshold time t_{act}, eq. (9) describes AF only if $t_{act} = t_{switch}$ holds true. To determine AF for conditions when $t_{act} < t_{switch}$ ($AF(t=t_{switch}) > 0$) or for a delayed activation, $t_{act} > t_{switch}$ ($AF(t=t_{switch}) = 0$), an additional parameter has to be introduced. This parameter is an offset of the AF-curve along the time-axis and is therefore called t_{offset}. Taking t_{offset} into account, eq. (10) can be obtained after integrating eq. (9).

$$AF(t)_{flushing} = e^{\left(\frac{t + t_{offset} - 2\cdot t_{switch}}{\tau_{CSTR}}\right)} - e^{\frac{-t_{switch}}{\tau_{CSTR}}} \qquad (11)$$

The parameter t_{offset} is initially unknown and has to be calculated. For this we need to differentiate between two cases. First, if t_{act} is larger than t_{switch} and therefore AF is 0 at the switching point, t_{offset} can be obtained by subtracting t_{act} from t_{switch} (eq. 11a). Second, if AF at $t = t_{switch}$ is above 0, t_{offset} has to be calculated by solving eq. 10 for t_{offset}. For this $AF(t)_{flushing}$ has to be set to $AF(t=t_{switch})$ and eq. 10 has to be rearranged as shown in eq. (11b).

$$AF(t{=}t_{\text{switch}}) = 0 \qquad t_{\text{offset}} = t_{\text{switch}} - t_{\text{act}} \qquad (12a)$$

$$AF(t{=}t_{\text{switch}}) > 0 \qquad t_{\text{offset}} = \ln\left(AF(t{=}t_{\text{switch}}) + e^{\frac{-t_{\text{switch}}}{\tau_{\text{CSTR}}}}\right)\cdot\tau_{\text{CSTR}} + t_{\text{switch}} \qquad (11b)$$

8. **Title of Sect.4**: What does the "first experiments" mean? Try to update the message into a more informative one.

The caption "Application in first experiments" was removed. The following captions were rephrased.

9. Page 12, line 10: What does the "uniform" mean: "activate uniformly" here and "a uniform aerosol population" in the caption of Fig.5? Are you trying to say the initial particles with the same particle size and chemical composition? If so, how to understand the Gaussian distribution scenario (i.e., ***"This is because there are some particles in the population, that activate earlier than the mean activation time."***), as all the uniform particles are supposed to activate at a same activation time? More straightforward/concise descriptions would be useful to explain the scenario clearly.

The reviewer understood the meaning of "activate uniformly" correctly. However, we do not consider this scenario as the most like scenario. It is more likely that some particles activate after a shorter/longer aging time than other particles. A reason for this can be that even in a size-selected aerosol particle flow some particles are slightly small/larger than the average diameter. Since the CCN-activity of particle shows a size-dependency this can lead to a non-uniform activation of the aerosol particles.

In the in previous sections "Introducing the activation time distribution $P(t_{\text{act}})$" and "Impact of the activation time distribution on the individual *AF*" we created a scenario where particles do not activate uniformly, but show a distribution of different t_{act}'s. We use a Gaussian distribution to capture the fraction of particles that activate after a certain necessary aging time t_{act}.

In section "Calculation of the total activated fraction (global *AF*)" we pick up the previously introduced t_{act} and $P(t_{\text{act}})$-concept and calculate the changing global *AF* throughout a full CSTR-experiment. This is done by integration of the contribution of individual aerosol fractions to the global AF over the whole range of possible t_{act}'s (equation 13)

$$AF(t) = \int_{t_{\text{act}}=0}^{t_{\text{act}}=t} AF(t_{\text{act}},t)\cdot P(t_{\text{act}})\ \mathrm{d}t_{\text{act}} \qquad (13)$$

10. Page 15, line 10: How was the particle wall loss rate of ***k*** = 0.000625 min-1 estimated? Where can the readers find the corresponding clues/data for calculation?

The paper the wall loss rate was obtained from the difference between theoretical and expected particle loss during flushing under the assumption of a first order loss kinetic.

We extended the explanation of how we calculated the particle wall loss numbers and refer to publications were the same calculation regarding particle losses was applied.

The section was changed from: (old: P15 L8-11)

> The measured particle flush rate τ_{flush} obtained during the flushing regime is 104 min in both experiments, which differs slightly from the theoretical flush rate τ_{CSTR} = 111 min. This difference is caused by particle losses to the chamber wall. From this difference the particle wall loss rate of k = 0.000625 min-1 and a mean particle life time upon wall loss of 1600 min was determined assuming first order loss rates

To: (new: P16 L10 –L24)

> In the flushing regime the particle number concentration declines exponentially in both experiments. Eq. (5) describes the ideal/theoretical evolution of the particle number concentration in the flushing regime when taking the hydrodynamic residence time τ_{CSTR}according to eq. 1 into account. In the ideal case the decay is solely caused by the flushing process. In reality, the decay is a combination of flushing as well as additional particle losses e.g. wall losses or coagulation. Therefore, the real residence time can be obtained by fitting equation 5 to the experimental data after rearrangement for τ, to which we refer to as τ_{flush} from now on (Kulkarni et al., 2011). In both experiments τ_{flush} coincides at 104 min, which is lower than the hydrodynamic residence time τ_{CSTR} of 111 min. In other words, the particle concentration declines faster than expected. This difference is caused by particle losses to the chamber wall, which acts as an additional particle sink parallel to flushing and reduces the particle lifetime. Nevertheless, statistical analysis of the experimental data results in purely statistical noise centered on the fitting curve used to determine τ_{flush}. This indicates that in terms of mixing no difference between an ideal CSTR and the aerosol chamber used here can be detected with the applied instrumentation.
>
> When dividing the real particle life time (τ_{flush}) into its individual components, a particle life time upon wall losses ($\tau_{wall\text{-}loss}$) of 1600 min can be determined in accordance with first order wall loss kinetic (Crump et al., 1982; Wang et al., 2018). The influence of particle coagulation can be considered negligible due to the low coagulation rate of 100 nm particle at concentrations of maximum 1500 cm^{-3} (Kulkarni et al., 2011).

11. Page 15, line 15: What is the meaning of the last sentence? What does the “otherSS” refer to? Where can readers find the corresponding details? Necessary information is needed.

This refers to scans through different SS. We only discuss the applicability of the CSTR approach in this paper, therefore we refrain from discussing the impact of ozone-aging onto CCN-activity at different SS. This will be done in a separate publication.

Sentence changed from: (old: P16 L14-15)

> The gaps in the curves during steady state are due performing measurements at other SS.

To: (new: P16 L27-28)

The gaps in the curves during steady state are due to changes in the operation of the CCNC form running on a constant SS (1.0% and 1.4%, respectively) to scanning over a range of SS

12. **Figure** 2: Is the "particle age" of x-axis with the same meaning of the "residence time" in Fig.1? If not, please specify accordingly in the corresponding places.

In this context "particle age" and "residence time" are identical. However until this point in the manuscript, this is not explicitly stated.

Therefore we changed the x-axis description from:

> particle age / min

To:

> residence time / min

13. **Figure** 6: Why is the unit of particle concentration in Fig.6(A1) and (B1) different from those in Figure 3? In Fig.6 (B1), why are the data after 800 min missing? As assumed early in this study that all compounds in CSTR have perfectly mixed thus with constant concentrations during steady state, how to explain the increasing trend in observed particle concentration in the duration of 400-600 min, i.e., AF almost reached a stable level around 0.2 at 1.4% SS conditions)? More detailed discussion should be provided in the corresponding data interpretation sections.

The particle number concentrations are not meant to have different units. It is always #/cm^3. The dot in the unit of the y-axises in Fig.6(A1) and (B1) was a graphical error and was removed.

The deviations from the theoretical changes in the particle concentration are due to a slightly changing particle input concentration. From an experimental point of view it is challenging to keep the particle input concentration constant over a period of 12 h. However, this small change affects the outcome only to a small degree.

The section was changed from: (old: P15 L 6-8)

> The particle concentration curves follow the theoretical filling and flushing curves in a CSTR. The slight decline in the concentration observed in the region where steady state is expected in graph A1 is due to a slight but undesired reduction in the feed-in flow that was experienced during the experiment.

To: (new: P15 L4 – 9)

> The graphs A1 and B1 in Fig. 6 show the particle concentration (black crosses; left axis), the measured global *AF* (red crosses) and the fitted global *AF* (blue dashed line, both right axis). The particle number concentration curves (black crosses) follow the theoretical filling and flushing curves as expected in a CSTR (Fig. 3). The slight decline in the concentration in steady state in graph A1 is due to a slight reduction in the particle input concentration that was experienced during the experiment. Visa versa the slight increase in the number concentration in graph B1 is due to a slight increase in the particle input concentration over time.

14. Page 19, line 6: The last sentence is a bit confusing. It is better to clarify the "metric" here, e.g. metric of what specific aspects.

We clarified under which circumstances the activation time concept can be benefical for the data analysis

Changed from: (old: P19 L6)

> Depending on the parameter of interest we suggest using t_{act} or $P(t_{\mathrm{act}})$ as metric.

To: (new: P20 L31-36)

> Therefore, we suggest to apply the concept of the activation time t_{act} or the activation time distribution $P(t_{\mathrm{act}})$ as metric in addition to calculating average values, such as the global *AF* and OH-exposure if following conditions are met. One, the system or parameter of interest can be described as a binary system and undergoes step-wise / non-gradual transitions such as CCN-activity. Two, the OFR used has a RTD broad enough to influence the outcome. Three, the conditions inside the reactor are either homogeneous or a correction for inhomogeneities (e.g. different oxidants concentrations inside the reactor) is implemented

Technical corrections:

1. **Abstract**, Page 1, line 10: *"… the newly introduced metric: activation time"*

done

2. Page 3, line 27: *"… can be calculated as a function of …"*. A similar issue exists in Line 16, Page 6.

done

3. Page 6, line 13: *"… to describe continues continuous changes"*?

done

4. Page 6, line 19: *"… can be considered as a non-gradual change."*

done

5. Page 6, line 20: *"If the all the other parameters stay constant, while a particles undergoes changes that result in a non-gradual transitions…"*

done

6. **Equation 9**: Why do you use different multiplication signs in these equations, e.g., "*" and "·"? It makes more sense to keep consistent within the same manuscript.

done

7. **Table 1**: Why is the layout of this table so different from other two tables in this manuscript? The corresponding details could be better organized.

We harmonized the table layouts

8. Title of **Sect.4.3**: *"Calculation of the total activated fraction"*

done

9. Page 12, line 12: *"While the uniform scenario shows no activity be for reaching tact …"* Do you mean 'before'?

changed to "before"

10. Page 14, line 8-9: *"As there is a significant share of particles activating significantly earlier than the nominal activation time (μ = 180) in the case of a Gaussian distribution a fraction of 1 % of the entire particle population within the CSTR is already activated after 87 min."* A comma is needed to clarify the point.

Changed from:

> As there is a significant share of particles activating significantly earlier than the nominal activation time (μ = 180) in the case of a Gaussian distribution a fraction of 1 % of the entire particle population within the CSTR is already activated after 87 min

To: (P15 L5-7)

> In the case of t_{act}-onset, there is a significant share of particles activating significantly earlier than the nominal activation time (μ = 180) in the case of a Gaussian distribution. Therefore, a fraction of 0.01 of CCN active particles within the entire particle population is already present after 87 min

11. Page 14, line 12: *"The difference in tact of 10 min between the two P(tact)-approaches is due to the application …"*
It is very common to see that tact was written as tact. Similar issues also exist in some other expressions, e.g., Pstep, which should be Pstep. Please check through the manuscript carefully and make necessary updates accordingly.
In the same paragraph, there are many long sentences without proper splits or connections, which might make the readers difficult or even confused to catch the meaning effectively. For instance:

We harmonized the subscripts

12. Page 14, line 15-16: *"As can be seen in Graph C of Fig. 4, 50 % of the particles with a residence time equal to the nominal activation time are activated in the case of a Gaussian distribution corresponding to tact0.5."*

Comma added.

13. Page 14, line 23: *"… were diluted with particle-free and VOC-filtered air…"*

Changed from:

particle free

To:

particle-free

14. Page 14, line 25: *"The aerosol flow was fed into the aerosol chamber, where a constant ~~Ozone~~ ozone concentration of 200 ppb was …"*

done

15. Page 14, line 27: *"The size distribution data was acquired by a … (SMPS) system from which the ~~the~~ total particle concentration could be derived."*

done

16. Page 15, line 5: The *"(blue solid line)"* is not needed, since there is only one curve in the corresponding subplots.

We thank the reviewer for mentioning this point, but we keep the "(blue solid line)" to avoid any confusion with the curves in the graphs Fig.6 A1 and B1.

17. Page 15, line 18: *"… µ as well as σ ~~are~~ is larger for P(tact) at a 1.0 % SS of compared to the results obtained for 1.4 % SS. The mean activation time being larger for 1.0 % SS indicates that the longer the chemical aging proceeds, the initially inactive soot particles activate ~~a~~ at a lower SS."*

done

18. Page 15, Line 23: The comma between "*P(tact)*" and "requires" is unnecessary.

comma removed

19. Page 17, line 1 and 3: *"Within these types of chambers …"*

done

20. Page 17, line 10: *"secondary organic aerosol (SOA)"*, and the "VOCs" should be defined before when it appeared for the first time.

done

21. Page 17, line 24: *"…to be directly proportional to the AFs…"*

done

22. Page 18, line 3: *"…we ~~presente~~ present two scenarios."*

changed

23. Page 18, line 9: *"…other parameters can agree very well."*

done

24. Page 19, line 22: *"…soot particles transitioning ~~form~~ from initial CCN-inactivity to CCN-activity over the course of …*

done