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.

Referee for Friebel and Mensah 2018
Journal: Atmos. Meas. Tech.

Summary:
The authors provide a theoretical framework that introduces the concept of activation time, which is the reaction time necessary to produce an observable chance in an aerosol population's property (CCN ability in this case). This is done in an effort to interpret data from a reactor operated in continuous mode rather than batch mode, the case being made because of the increased use of OFRs and OFR-like reactors compared to environmental or smog chambers. They describe the entire reactor operation from fill-up to shut-down. They provide a mathematical description which, to my understanding, is a piecewise solution in time for activation fraction of aerosols in question (soot aerosols subject to heterogeneous ozonolysis). This manuscript is front-heavy with concepts of chemical reactor engineering (ideal reactors and their residence time distributions), and how these are used to develop analytical expressions for the time profiles. Then their expressions are overlaid to data from two experiments to observe model agreement. Finally, they depict a theoretical example based on preexisting data in the literature to show how their parameter tact can be used to compare data from independent experiments, or even better, how future users chose to operate their reactors. This is very important work that fits the scope of AMT; particularly the illustrative example in Fig.7.

However, I think the manuscript be improved. An in-depth revision of Sections 1-4 is necessary, mostly for emphasis on technical details and wording to reach a broader audience (that is, one unfamiliar with reactor design or operation). For example, the authors need to clarify what they mean by 'non-gradual' as soon as it is mentioned. Are they referring to fast reactions, e.g., heterogeneous nucleation? Or are they simply referring to non-steady state?

We understand that many readers are not familiar with the wording we used. We thank the reviewer for pointing this out. 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 this we chose processes commonly investigated in 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 neither refer to fast reactions nor non-steady state. "Non-gradual" describes 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.

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 one 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.

There is a bit of a disconnect between the theory and application. Probably because the nature of the subject is challenging. The authors are encouraged to make it clear in the Introduction that they are looking at CCN. Also, because tact is yet to be explained, words like 'parameters' have no meaning thus far; they do eventually by the end, but I think not to confuse the reader a revision is necessary (I offer suggestions in the Major and Minor comments sections for the authors' considerations).

This manuscript introduces two new concepts that were developed side by side and support each other. However these concepts are not limited to one particular application. The first concept is the use of aerosol chambers in CSTR-mode. The second concept is the idea of analyzing data with the activation time concept, which relies on "non-gradual" transitions. We try to make a clear distinction between the experimental approach rather new in atmospheric sciences and the t_{act}-concept. We hope that the revision of the sections where non-gradual" transitions (section 1 "Motivation") and the t_{act}-concept (section 3

From what I can tell, their data is centered on reactors operated in continuous mode, yet the word 'OFR' is mentioned only in Section 5, when the introduction is focused on the large batch reactors.

(P3 L15-L39)

From a technical perspective, generally three types of reactors are distinguished: the batch-reactor, the plug flow (PFR) or flow tube reactor, and the Continuous flow Stirred Tank Reactor (CSTR).

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.

For that matter, comparison of the mathematical framework to that of a PFR is not present. If so, I think some mention as to why should be made. It seems to me that PFR-like reactors (e.g., flow tubes) work well. Why are CSTRs preferred by the authors? Mathematically, it would appear to me you need an RTD, and the PFR has one (Delta function), so why not compare?

The mission of this publication is to present a reaction chamber operation mode that is not that prominent within the atmospheric community yet but comprises important benefits for the investigation of atmospheric processes. These benefits include extended reaction times and low reactant concentrations. The limited popularity of the CSTR concept within the atmospheric community might be partly due to limited availability of analysis procedures of data resulting from such experiments. We developed the activation time concept to allow for the analysis of reactions and processes relevant to the atmospheric community. Therefore, this publication intentionally focusses on the activation time concept and the CSTR-mode operation of aerosol chambers. We refer to OFRs and large batch aerosol chambers to discuss the differences, benefits, and disadvantages of the CSTR concept in comparison to these reaction chamber concepts established in the atmospheric community. The mathematical framework for the analysis of data from ORFs is not comparable to the t_{act} concept for CSTR data presented here. Instead the parameters of interest of OFR data are average residence time, average exposure to oxidants and critical super saturation/super saturation onset. We discuss the different metrics and their comparability in section 3.5 "Equivalent parameters t_{act} – onset and t_{act} vs $t_{act}0.5$" and section 5 "Application of t_{act} to other continuous flow aerosol chambers" now in greater detail.

Major Comments:
Abstract: Details can be improved; I offer suggestions for the authors to consider in the Minor Comments.
1. Introduction: I strongly encourage the authors to be more precise in their sentences. It appears that what the authors communicate is not what they mean, and to reach a broader audience, I think details should be made clear. While Sections 5 and 6 are very clear and logical, at least in my view, Sections 1-4 are not. I encourage OFRs like the PAM (e.g., TPOT, CPOT, etc.) to be addressed early on. The authors can read more in Lambe et al. and Mitroo et al., already cited by them.

We expanded 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 chamber, namely OFRs. (P2 L40- P3 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).

Also what is not clear is whether the authors have a new CSTR design (different from that of conventional OFRs or Teflon chambers) or if they just develop a mathematical approach for data coming from a CSTR. Or both.

We do not present a new CSTR design but the application of the concept, which is well established in chemical engineering but not that prominent in atmospheric sciences, yet. The authors would like to highlight the fact, that CSTRs are neither OFRs nor Teflon chambers operated in batch mode. Teflon chambers could be operated in CSTR mode but this demands the installation of a fan as the concept of the CSTR requires perfect internal mixing of the sample.

Mention of the SAPHIR, in my view, belongs here.

The SAPHIR-chamber is mentioned as an example of a large reaction chamber:

(P1 L28-29)

> Here aging durations of up to 16 hours and beyond at atmospherically relevant reactant concentrations can be achieved, which has been shown e.g. for the SAPHIR chamber of FZ Julich with a volume of 270 m3 (Rohrer et al., 2005; Rollins et al., 2009).

2. Introduction to CSTR: This section is of course important for readers who are not familiar with environmental reactor engineering, however, is not only available in any chemical engineering textbook, but also summarized by Mitroo et al. (Appendix A).

We agree to the fact that Mitroo et al. discusses concepts relevant to for describing residence time distributions in OFRs within their paper. The equations presented in section "3.2 Tank-in-series model for indirect deconvolution" within their publication allow to calculate RTD and RTD_{sum} – curves. This is a general description of multiples CSTRs that are connected in series. For the special case where N=1 the mentioned equations become the equations we used in our study.

Instead of using RTD and RTD_{sum} Mitroo et al. labeled the curves as E and F curves, which is common in the engineering community. We prefer the first option since it is a more intuitive notation.

In "Appendix A: The use of E and F curves" of their publication Mitroo et al. describe how their RTDs were determined and how to calculate moments of the RTD.

However, none of this contains a description of the CSTR and its RTD, therefore we do not refer to Mitroo et al. in this section.

In section "5 Application of t_{act} to other continuous flow aerosol chambers" of our manuscript we discuss how different RTDs in different OFR-designs lead to differing results of a parameter of interest. A quantitative application of t_{act}, requires precise knowledge of the chambers RTD. The concepts of Mitroo et al. can be used to obtain that. However, within this work we only focus on a qualitative application of t_{act} to continuous flow aerosol chambers that are characterized by none perfect mixing

If the authors see fit, I would suggest renaming this section as 'SAPHIR operation' or something similar, and then have Filling, SS, and Flushing sections.

We recognize the suggestion of renaming this section but remain at the initial naming. This is due to the fact, that this section is intended to introduce the CSTR concept in general as well as its specific aspects and characteristics. The SAPHIR chamber is a batch reactor and not a CSTR. While within the operation of a CSTR the three different regimes of filling, steady state, and flushing can be achieved, no steady state can be achieved throughout the operation of a batch reactor. This is one of the fundamental differences between these two reactor types.

It seems that from Sections 5-6, their math can be applied to non-CSTRs like the PAM and TPOT, so I wonder if when the authors say 'CSTR' they mean 'non-batch'. Reactors operated in continuous mode range from CSTRs to PFRs, from a mixing perspective.

We agree to the reviewer that both CSTR and PFR are continuous mode reactors and include a phase of steady state operation. Nevertheless, we want to highlight the critical difference between these two reactor types. While perfect/well-mixing is an additional prerequisite exclusive to the concept of the CSTR, OFR are characterized by a partial mixing of the sample. As this section is titled 2 "Introduction of the CSTR" we focus on the mathematical framework relevant for CSTRs exclusively. For a comprehensive introduction of the new

concept and to allow the reader to first get acquainted to it, we postpone the introduction of the transferability of this new mathematical framework to other reactor types to a later section of this manuscript (section "Application of t_{act} to other continuous flow aerosol chambers"). Further, we attempt to clarify the distinction between well-mixed (CSTR) and partially-mixed (OFR) systems and mention this now in the abstract:

(P1 L15-18)

> 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.

The new mathematical framework presented in this publication is developed for the extraction of quantitative data from CSTR measurements. Nevertheless, equation 13 (P13 L11) represents a general expression that can be applied to any continuous flow but non-CSTR chambers as well. For the retrieval of quantitative date, additional information about the specific chamber is required. We show that despite the lack of access to such information the application of the t_{act}-concept allows not only for a qualitative comparison of the TPOT and PAM chambers but allows even for an explanation of the differences in the measurement results.

$$AF(t)= \int_{t_{act} = 0}^{t_{act} = t} AF(t_{act},t) \cdot P(t_{act}) \; \mathrm{d} \, t_{act} \tag{1}$$

The following section was added to discuss a quantitative application of t_{act} for data from OFR-experiments.

(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.

3. Introduction of the activation time (tact) for non-gradual transitions: A major comment I have here that I alluded to prior to this section is to be explicit when talking about parameters. E.g., P6 L20 "If all other parameters stay constant…" what does this mean? Flow parameters? Temp and RH? If so, what is the parameter that is changing? I don't think the reader thinks of AF by now.

"If all other parameters stay constant…" refers to the previously mentioned external parameters. These external parameters can trigger changes in single particles. For example a change in ambient temperature can lead to freezing or melting of a particle. In this section we discuss that such changes can also be the result of e.g. a chemical reaction. If the temperature stays constant but the particle becomes liquid due to a chemical modification we can use the time as parameter to describe this phase transitions.

Examples for the external parameters were added:

Changed from:

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 (P6 L29-32):

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.

Also, P6 L24-26 seem to me like the crux of the study (unless I'm mistaken).

This is not the crux of the study. The t_{act}-concept and the idea of describing changes in parameter on the particle level as transition within a binary system/a stepwise-change/a non-gradual transitions is the crux itself. The mentioned lines are an application of this general idea to the specific process of CCN-activation of soot particles due to aging. This application affects the experimental design but not the overall theoretical framework.

To (P6 L32-35):

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}). This concept is generally valid and can be applied to any kind of transition in a system defined as binary either by intrinsic or operational parameters.

Are the authors looking at a specific scenario where they keep RH constant but slowly react aerosol with (e.g, ozone for sake of argument) and there is a very small time window where enough reaction occurred to make the aerosols in the reactor cloud nuclei at that supersaturation? Is that time window what current reactors cannot accurately allow determination of, but this method does? Why can't a PFR be used to detect that? If so, this concept needs to go in the introduction, with specific application to CCN if helpful. Finally, for the authors' consideration, it appears they want to keep the x-axis uniform in their equations by introducing tswitch and toffset. Seems to me like these are just substitutes for a Heaviside function. Would the authors consider using a Heaviside function instead to make the math simpler?

The section 3 "Introduction of the activation time (t_{act}) for transitions in binary systems" is a general introduction of t_{act} and does not include any specific chamber design. This concept is also not limited to any specific scenario. We focus our introduction of the rather theoretical and abstract concept with the help of 2 processes (deliquescence and CCN-activation) that are well known in the atmospheric science community.

(P9 L19-28)

Binary systems can be considered as systems that show a step-wise change in a particle property as a function of an external parameter. Since this is opposite to a continuous/gradual change in a particle property, it can be also described as a non-gradual transition. As mentioned in the introduction, soluble aerosol particles such as ammonium nitrate exhibit a significant change in diameter with increasing relative humidity (RH) due to deliquescence. Similarly, the change from cloud condensation nuclei (CCN) to activated droplets due to exposure to a super-critical super saturation (SS) results in a fast increase of the particle diameter from the nanometer to the micrometer scale that is hard to be continuously tracked by standard measurement instrumentation. A defined diameter threshold is hereby used to distinguish between an aerosol particle and a solute droplet in the case of deliquescence. This is the same between non-activated CCN and cloud droplets. In both examples the relative humidity (RH) in the surrounding gas phase can be considered the external parameter that controls if an aerosol particle is in either of the two states of the binary system (effloresced vs. deliquesced/CCN vs. cloud droplet).

Up to this point, the t_{act}-concept and the equations are independent of the overall time-scale. It doesn't matter if the processes need seconds, minutes, or hours to proceed. Therefore we do not discuss applicability of PFRs or CSTRs in this section.

Nevertheless we developed the t_{act}-concept to retrieve data from aerosol aging experiments in a 3 m^3 CSTR. The aim of this approach was to achieve aging times of several hours. As can

be seen in Fig.6 (P17 L1) this long time spans are indeed needed to investigate the CCN-activation of soot particle due to ozone oxidation. So far, to the best of our knowledge, no OFR-design exists that can reach residence time of several hours which is applied in atmospheric science experiments on a general basis. Increasing the ozone concentration to mimic several hours of atmospheric aging in 2 min (which a typical residence time in OFRs) is a very different approach.

As assumed by the reviewer we want to keep the x-axis uniform. The x-axis is hereby the experimental duration t starting with the beginning of the experiment. Avoiding the parameters t_{switch} and t_{offset} would lead to equally complex equations while potentially increasing the confusion of the reader, since multiple time-axises/x-axises would be needed. Therefore we remain with the math presented so far.

The parameter t_{switch} was introduced since this time is defined be the experiment itself, namely the time after which the chamber is switched to flushing mode.

The explanation of parameter t_{switch} was extended from:

> However, eq. (9) only describes the fraction of particles that are older than $t$switch and therefore the global AF only if $t$act = $t$switch. To determine the AF for conditions when $t$act < $t$switch ($AF(t=t$switch) > 0) or for a delayed activation, $t$act > $t$switch, a new parameter
> $t$offset, is introduced. This parameter is an offset of the AF-curve along the time-axis. Taking $t$offset into account, eq. (10) can be
> obtained after integrating eq. (9)

To (P10 L5-9):

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).

4. Application in first experiments: I don't think this section header reflects the content. Maybe change to something else?

The caption "Application in first experiments" was indeed misleading and has been rephrased to:

> 4 "Application of the new tact to experimental data from CSTR-aging experiments"

Section 4.2 was described well. My only major comment here is why Figure 3 has a lag (noticed after seeing Table 2 and Figure 5) Why does the 'step' or 'non-gaussian' have a lag? Even in the filling regime a CSTR gives no lag. E.g., in P14 L10-13 I remain unconvinced that

the blue line in Fig. 5 should have a lag. I think that assumption (P7 L7; see comments for Fig. 5) is highly questionable. I think that leads to an artifact in the calculation, and that is reflected by the stark difference in tact-onset (Table 2). If the authors provide a counter, I'd be happy to know why.

First we want to highlight the distinction between the activation time t_{act} and the experimental duration t. t_{act} is the time needed to modify a particle to such a degree that a particle's property changes step-wise. The experimental duration t is the time that passed since the start of the experiment. We extended the respective section 3.1.2 "Particle activation during steady state" substantially in the attempt to clarify this distinction

Further, In Figure 3 only AF has a lag but not the particle number concentration. As mentioned by the reviewer there is no lag during filling of a CSTR. If we recall, that a chemical transformation of a certain degree is necessary before an individual particle can be CCN active, no CCN activity can be detected prior to this minimum time threshold which we refer to as t_{act}. The lag in AF up to t_{act} = 180 min is therefore an intrinsic behavior as the experimental duration t is shorter than this this minimum time t_{act}. This underlying concept defines the shape of the curves in Fig. 5 as well. We attempt to clarify this issue in section 3.1.1 "Particle activation during the filling regime":

(P7 L12-19)

Assuming that only aerosol particles are CCN-active which have an individual residence time in the aerosol chamber that is above t_{act}, the theoretical AF can be calculated according to eq. (6). Two different time ranges within the experimental durations need to be considered. If the experimental duration t is below t_{act}, AF is 0 as even the particles that entered the aerosol chamber at the very beginning have an individual residence time shorter than t_{act} and therefore cannot be CCN active yet (eq. 6a). If the experimental duration t is above t_{act}, AF is greater than 0 as a subset of the particles will have an individual residence time longer than t_{act} and therefore can be CCN active (eq. 6b). Application of eq. (3), which describes RTD and rearrangement of eq. 6 allows for the calculation of the activation time t_{act} based on an experimentally determined AF as shown in eq. (7). This equation is valid throughout the entire filling regime including steady state.

The AF-curve in a CSTR appears to be much smoother for the case of a Gaussian shape t_{act}-distribution. This is due to the fact that this scenario comprises particles with different individual t_{act}'s as introduced in section 3.3 "Impact of the activation time distribution on the individual AF". A small subset of the particle population activates much earlier than 180min, therefore a global AF = 0.01 is reached much earlier as well. At the same time an AF=1 is reached much later than in the case of the step-wise t_{act}-distribution because of particles with a significantly higher individual t_{act}. The difference in the tact-onsets from Table 2 is therefore no artifact from the calculations but a direct (and expected) result of applying the t_{act} – concept to the CSTR-approach. The text discussing Table 2 now reads:

(P15 L1-18)

As can be seen in Table 2, the individual values deviate with the biggest deviation in the case of t_{act} -onset. However, the presented deviations are solely caused by the underlying

distributions of the activation time. In addition, t_{act}-onset, t_{act}, and $t_{act}0.5$ are determined at different experimental times. While t_{act}-onset is directly determined by measuring the entire particle population within the CSTR (global AF), t_{act} is calculated from the global AF in steady state and $t_{act}0.5$ is obtained from the activation time distribution itself. In the case of t_{act}-onset, there is a significant share of particles activating significantly earlier than the nominal activation time ($\mu = 180$) 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. Opposite to this, the threshold value of 0.01 is crossed later than the nominal activation time in the case of the step distribution. This is because even though every single particle activates after exactly 180 min of individual aging time, it takes some additional time before a fraction of 0.01 of the entire particle population within the CSTR is older than 180 min leading to a t_{act}-onset of 185 min. The difference of 10 min in t_{act} between the two $P(t_{act})$-approaches is due to the application of eq. (8) which allows for the calculation of t_{act} from the global AF in steady state. Strictly speaking, this equation is defined for the ideal step function ($P_{step}(t_{act})$) only. Therefore the higher global AF value for $P_{gaussian}(t_{act})$ in steady state has to lead to a lower t_{act} value compared to $P_{step}(t_{act})$. Note, $t_{act}0.5$ is referring to the particle activation distribution $P(t_{act})$ only leading to a concordant value of 180 min in both cases. This can be seen in Graph C of Fig. 4, where 0.5 of the particles with a residence time equal to the nominal activation time are activated in the case of a Gaussian distribution corresponding to $t_{act}0.5$. In the case of a step function, all particles are activated once the respective particle population is older than t_{act}. In the following we will show how the actual activation time distribution $P(t_{act})$ can be retrieved from real CSTR experimental data.

5. Application to experimental data: No major comments here other than those that stem from the previous section.

6. Application of tact to other continuous flow chambers: I think this section would be very useful for OFR users on how to use OFRs for CCN meaurements! Still, neither the PAM nor the TPOT are CSTRs, so how have the authors applied tact to their RTDs? Also, what if aerosol content is not well known (e.g., field sampling)? How is their mathematical framework applied? I am still unclear as to what parameters are needed experimentally.

The PAM and TPOT chambers are indeed no CSTRs and cannot be described with the CSTR-specific mathematical framework introduced here. However, they show a significant residence time distribution due the mixing along the flow path as already discussed by e.g. Lambe et all. 2011 and Mitroo et al. 2018, 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 residence 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 necessary aging time t_{act}.

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

In this manuscript we present 2 scenarios that illustrate the application of the t_{act}-concept to the RTDs reported by Lambe et al. 2011 (Figure 7 P19). In the first scenario (High-OH) we calculated the fraction of particle older than 40 s in the PAM and TPOT chamber, respectively. In the second scenario we did the same with a t_{act} of 180 s. As can be seen, the fraction of particle older than these threshold times t_{act} varies between both chambers. Since in our definition only the oldest particle can be CCN-active this leads to different values of *AF*-values depending on the chamber. 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 results obtained based on the application of our t_{act} concept agree well with the trend reported by Lambe et al 2011. The respective text in section 5 "Application of t_{act} to other continuous flow aerosol chambers" now reads:

(P19 L7 – P20 L13)

In the following we show that the application of t_{act} can contribute significantly to the explanation of the aforementioned discrepancies in terms of CCN-activity of the BES particles.

In Fig. 7 we show the RTDs for 145 nm BES particles using the parameters for the bimodal Taylor-dispersion model given by Lambe et al., (2011) in Appendix A4 (Fig A3). We normalize the area under the curve to be one causing the area under each curve to be directly proportional to the *AF*s for a better visual comparison. Here, PAM chamber data is indicated by the dotted line/green area and TPOT chamber data is indicated by the dashed line/blue area. As can be seen, the two curves are not perfectly superimposed with the peak of the PAM chamber RTD being earlier than in the TPOT chamber RTD followed by a steep decline causing the two curves to cross at approximately 40 s. Overall the PAM chamber RTD (dotted line) shows a stronger dispersion causing the two lines to cross again at approximately 180 s.

Assuming a high OH-concentration leads to a higher reaction speed and therefore shorter t_{act} we present two scenarios. Scenario A representing a high OH-concentration is based on a t_{act} of 40 s (Fig. 7 A). Scenario B representing a low OH-concentration is based on a t_{act} of 180 s (Fig. 7 B). In both cases the BES-particles show CCN-activity, but the global *AF* differs significantly between both chamber types. In the high-OH scenario A, the TPOT chamber is more efficient in producing CCN-active BES-particle (AF_{TPOT} = 0.892; blue area) than the PAM chamber (AF_{PAM} = 0.655; green area) as can be perceived by the blue area being larger than the green area in the left panel. Opposite to this, the PAM chamber is more efficient (AF_{PAM} = 0.082) than the TPOT chamber (AF_{TPOT} = 0.047) in case of the low-OH scenario B, as can be seen in the right panel. These calculations indicate how the new t_{act} concept can contribute to the understanding and interpretation of experimental data that has been acquired in non-CSTR reaction chambers. At the same average OH-exposure, aging in different OFRs causes the same global *AF* only if the RTDs are the same. Since the RTDs of

the PAM chamber and the TPOT chamber are not the same, the same global AF can only be obtained if the t_{act}'s differ. Three examples of how t_{act} has to deviate between the PAM chamber and TPOT chamber to lead to the same global AF are given in the supplement.

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.

(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.

We want to point out that every additional variable increases the complexity of the math exponentially. Fluctuation input concentration during field sampling can be rather easily implemented. Fluctuation input concentrations distort the RTD in the OFR. This could be approach by normalizing the RTD not to 1 but to the particle input concentration and then to continuously integrate over all RTD for all time steps.

Having inhomogeneous conditions inside the OFR e.g. different concentration of OH-radical is rather difficult to implement, which was one motivation to use an internally mixed chamber.

7. Conclusion: No major comments here.

Minor Comments:

P1 L7: Arguably a small point, but I'd encourage a revision of "achieve extended observation times" to "obtain measurable reaction rates, due to long residence times" or words to that effect. I think owing to the small reactor design community there's often scant attention paid to the details of a reactor and how it operates by the average reader; and this work has potential for a broad audience, so ensuring the readers become educated about reactor design and meaningful parameters to evaluate its performance is important in my opinion.

P1 L8-9: Change "…in a CSTR mode." to "…as a CSTR." Also, if the authors wish to introduce the concept of a CSTR this early on, perhaps the opening sentence could mention the use of environmental chambers as batch or semi-batch reactors.

We understand and appreciate the suggestion of the reviewer to implement batch and semi-batch reactor early on in manuscript. However we present a different experimental approach and refrain from mentioning batch-mode operation of aerosol chambers in the abstract. In section "1 Motivation" we discuss environmental chambers as well as PFR/OFR and compare them with CSTRs (P1 L27 – P2 L11). The respective text now reads:

(P1 L8-10)

We present an experimental approach that enables long observation times at atmospherically relevant reactant concentrations in small chamber volumes by operating the aerosol chamber as a Continuous flow Stirred Tank Reactor (CSTR).

P1 L9-10: Mean values of what? Perhaps 'its mean value', referring to the data.

In OFR and environmental chamber experiments an average exposure to certain oxidants (e.g. photochemical age) is typically reported. Since this does not always represent the aging conditions well we introduced the t_{act} as an alternative metric.

P1 L10-13

We developed a mathematical framework that allows the retrieval of data beyond calculating mean values such as O_3-exposure or equivalent atmospheric aging time, using the new metric: activation time (t_{act}).

P1 L14: Are the authors referring to the PAM? Perhaps give an example.
P1 L14-15: Rephrase sentence. tact explains or tact helps explain? What are the different chambers?
Are they smog chambers vs. OFRs? Are they aerosol flow tubes vs. OFRs?

As the reviewer encouraged as to give OFRs more space, we now mention the intercomparion of the PAM and TPOT in the abstract and mention that the t_{act}-concept can be applied to different OFRs in general and that it can explain discrepancies found between the TPOT and PAM chamber.

P1 L15-18:

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.

P1 L17: This may apply throughout but "Aerosol particles" should be "Aerosols".

In atmospheric sciences the term "aerosols" is often used when exclusively the condensed phase suspended in a gaseous phase is meant. However general definition of the term "aerosol" actually includes the condensed phase as well as gaseous phase. In the attempt to reach a broad audience even beyond the atmospheric science community, we remain with the wording "aerosol particle".

P1 L17-18: Small detail, but stating aerosols are 'emitted' into the atmosphere implies they may not be generated by nucleation. The authors can consider the following rephrase: "Atmospheric aerosols undergo various reactions; the timescale for which depends on their lifetime."

Please note, in accordance with the discussion above "Atmospheric aerosols" refers to the gaseous components as well. The sentence now reads:

P1 L21-22

Atmospheric aerosols undergo various chemical reactions and physical modification processes once they are emitted into the atmosphere.

P1 L19: Citation for sea salt aerosol lifespan (textbooks are appropriate as well) please.

Both numbers can be found in Textor et al.

Also, replace comma with '"whereas".

done

P1 L21: Please check 'aerosol particles', as per my previous comment.

We refer to aerosol particle only when excluding the surrounding gas phase, therefore we keep the original wording

P1 L23: "task" should be plural; also, I'd change 'the investigation of aerosol' with 'understanding atmospheric aging'.

task changed to tasks

We present a general experimental approach to study aerosols. This is not limited to "atmospheric aging" even though this is the main focus.

P1 L23-27: I'd encourage the authors to reword this section and not to gloss over how large

reactor are 'technically' unfeasible, but instead be more explicit (e.g., wall losses, time dependencies, etc.). Also, I don't think the SAPHIR has a set 16 h operation time, so perhaps the authors can say '12-24h' to indicate a range.

We appreciate the comment of the reviewer and rephrase the appropriate sentence. As Rollins et al., 2009 explicitly mentioned an aging time of 16h we continue to include this number:

P1 L28-30

Here aging durations of up to 16 hours and beyond at atmospherically relevant reactant concentrations can be achieved, which has been shown e.g. for the SAPHIR chamber of FZ Julich with a volume of 270 m^3 (Rohrer et al., 2005; Rollins et al., 2009).

P1 L28-29: This is a technical detail I would like not to be overlooked. "…in order to reduce the reaction time…" is not the objective; it is a consequence (advantageous, admittedly, for investigating physiochemical properties of SOA or LVOCs). The objective is to artificially augment the reaction rate. I believe the authors know this, but a reader may not, and I encourage the idea of having these details be clear. This is important work and should be presented as such!

We appreciate the comment of the reviewer and the revised sentence now reads:

P1 L31-34

The second option is to increase the concentration of the reactive compounds such as oxidants and aerosol particles, in order to trigger higher reaction rates and thereby reduce the reaction time (George et al., 2007; Huang et al., 2017; Kang et al., 2007; Keller and Burtscher, 2012; Simonen et al., 2017). This allows to significantly reduce the volume of the aerosol chamber.

P2 L4: Please consider adding a citation of Renbaum and Smith, doi: 10.5194/acp-11-6881-2011

done

P2 L6: In the engineering literature, CSTRs are well described, but the acronym is use as a general term for any well-mixed vessel. The authors choose to apply (or design?) a CSTR for their work in addition to the mathematical expression for tact. Have I understood this correctly? Might I suggest them to give their reactor a more personalized name?

We agree to the reviewer. We operate an aerosol chamber in CSTR mode. In the previous of the manuscript the distinction between the different concepts was not clear. We changed the wording throughout the manuscript to "aerosol chamber operated in CSTR mode". This does not include a new chamber design, but is a different experimental approach.

Since CSTRs are known for quite a while we also cannot give it a personalized name. There is also no new reactor design involved like in the most OFRs. Furthermore the tank used here

was so well mixed that no deviation from a perfect mixing could be detected. The respective sentence now reads.

P2 L3-5

Here we present an experimental approach that can be used to achieve long aerosol aging times with neither need for large chamber volumes nor high reactant concentrations by operating an aerosol chamber in the Continuous flow Stirred Tank Reactor (CSTR) mode.

P2 L8: An ideal CSTR is perfectly mixed. A real CSTR is well-mixed.

We agree with the reviewer's statement. We adjusted the wording throughout the manuscript to avoid any confusion. We only use "perfectly mixed" in context of an ideal CSTR. The respective sentence now reads:

P2 L14

The volume of the CSTR is actively stirred in order to achieve a homogenous aerosol mixture.

P2 L9: Perhaps the authors can rephrase "…close to real processes in the atmosphere…" to "…mimics mixing in the free troposphere more accurately than [other reactors]". But more importantly, is the mixing state in this reactor important because it mimics atmospheric dynamics or because it allows more accurate data retrieval from laboratory experiments?

This reference to atmospheric processes shall give the reader a better understanding what it means to operate an aerosol chamber in CSTR-mode. We clarified that we compare the CSTR-approach and the atmosphere in terms of mixing aerosols and measuring a non-uniformly aged aerosol. The respective text now reads:

P2 L14-19

Due to the mixing, the aerosol that is continuously extracted for analysis consists of a well-defined mixture of aerosols at different aging stages. 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.

P2 L11-12: That's absolutely the case for a CSTR! At steady state, the distribution of ages is fixed, and is dependent only on reactor volume and flowrate. I think this needs to be clearer.

We focus on this specific aspect in section 2 "Introduction of the CSTR" and the respective text now reads:

P3 L3-5

> The key parameter for the description of reactions within a CSTR is the hydrodynamic residence time (τ_{CSTR}) which is also the mean residence time. It can be obtained from the reactor volume (V_{CSTR}) and the volumetric flow through the CSTR ($\dot{V}$) as shown in eq. (1) (Levenspiel, 1999).

P2 L14: In addition to refining the sentence to make it sound less informal, I would encourage the authors to cite also Levenspiel's Omnibook (I think they cite it later, but it's missing in the Reference Section).

> done

P2 L19: I'm confused, what do the authors mean when they say 'non-gradual' (see Major Comments)? Do they mean time-dependent? Do they mean non-steady state? This is a key concept in their work, so I would ask them to define it explicitly for the reader.

"Non-gradual" describes changes like phase-transitions where a property shows a step-wise change. The opposite is 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. When excluding super-critical conditions, at 0°C the density does not change gradually but changes step-wise. 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 one can therefore be described as "non-gradual". Further, we added a list of possible transition that can be described as "non-gradual", "step-wise change" and "transition between binary states"

P2 L24-39

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.

P3 L3-4: More than a physico-chemical (physio-chemical?) perspective, I'd say from a flow or mixing state perspective. Also, PFR can be placed in acronym in brackets (although PFRs can be mentioned in the introduction), and CSTR has already been spelled out earlier, so just the acronym should suffice here.

Changed from

From the physico-chemical perspective

to

From a technical perspective.

PFR acronym added

P3 L9: Can the authors make the case that environmental / smog chambers are batch-type reactors?

Changed from:

In a batch-reactor the reactants are introduced at the beginning of the experiment aiming for homogeneity and then the reaction
is allowed to procede. The composition throughout the vessel is homogeneous but evolving in time, therefore no steady state
conditions are ever achieved. After a certain reaction time the sample is discharged or collected and subjected to further
analysis.

To (P3 L17-23):

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).

P3 L9: Again, I would urge the authors to be detailed. A PFR (which is the idealized reactor design on which flow tubes are built) allows no axial mixing (as the authors point out), but is perfectly mixed radially! The ADM (mentioned by Lambe et al.) allows for deviation from the

PFR and is closer to describing flow tubes, but that discussion can be briefly mentioned, if needed at all.

The ADM describes how the RTD in a real OFR deviates from the RTD in an ideal PFR. For the application of the t_{act}-concept it is sufficient that the RTD is known. Therefore we do include different methods to characterize the RTD. We mention the PFR as reactor concept and point out that it is approximated by OFRs

Changed from

> A flow tube is a steady state reactor in which no mixing along the flow path takes place resulting in a constant output of products depending on the residence time within the reactor.

To (P3 L24-31)

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.

P3 L17: Residence time of what? The large chambers?

> "in large chambers" added

P3 L19: Consider replacing "During a subsequent…" with "Following steady state, upon shut down, is the…"

We thank the reviewer for the suggestion but we want to highlight the fact that the purpose of the flushing regime is to shut down the chamber exclusively but it allows to investigate the parameter of interest in an addition regime.

P3 L20: To better illustrate their point, I think the authors can put an arbitrary schematic in the Supplement rather than alluding to a figure that has not yet been explained.

> We remove the reference to Fig. 3 for clarity.

P3 L21: I don't think 'hydrodynamic' is necessary, but I could be wrong.

We clarified that it is the hydrodynamic residence time as well as the mean residence time. In case the reactor would have dead zones this would not the case.

Changed from:

> The key parameter for the description of reactions within a CSTR is the hydrodynamic mean residence time (τ_{CSTR})

To (P4 L3-4):

> The key parameter for the description of reactions within a CSTR is the hydrodynamic residence time (τ_{CSTR}) which is also the mean residence time

Equation (2): A suggestion to simplify notation, perhaps the subscript 'CSTR' can be removed, seen as it is implied. Also, (t) can be placed outside of the square brackets, as can the subscript 'feed-in', which I would also suggest be replaced with a subscript zero.

Using A_0 is indeed a more common notification to describe a single-step process. Here we focus on a two-step process (filling and flushing) therefore it would be necessary to define two different A_0 ($A_{CSTR} = 0$ and $A(t=t_{switch})$) which can confuse the reader. Since the square brackets are intended to indicate "concentration", we remain in the terminology used so far.

P4 L1: This is a good point by the authors! I would encourage a citation of Lambe et al., seen as what the authors are describing here is essentially the result of a tracer study (A is a chemically inert tracer essentially).

Unfortunately, the authors are not aware of the appropriate publication by Lambe et al. that the reviewer refers to. We want to highlight the fact that the beginning of steady state has to be chosen depending on the experimental conditions (e.g. resolution of detection) and can be different in different experiments.

P4 L11: I would encourage a citation of Mitroo et al.
Equations (3-4): These are E and F-curves as described by Mitroo et al.; it may be worthwhile to mention.

We added a sentence to explain our reasoning for the differing wording.

P4 L28-30:

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).

P6 L16-19: This needs to go either at the end of the introduction, or at P2 L19 in my view.

We believe we have addressed this comment in the new version of the manuscript by significantly extending the introduction of the term "non-gradual".

P6 L20-21: This sentence needs to be rewritten as it is too handwavy and comes across as pseudoscience. "…a particle that undergoes changes that result in a non-gradual

transition…" made no sense in my mind until I finished reading the manuscript. Could the authors come up with a physical example to help convey what change has been 'undergone' that resulted in a 'transition'? Or is the 'change' itself rapid (e.g., heterogeneous nucleation)? Are the authors implying they can model a process this fast as a function of time, and decouple it from other timescales within the reactor? Is a CSTR the best approach?

We thank the reviewer for his comment and aim to improve the understanding by specifying what kind of transitions we refer to.

Changed from:

> 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 P6 L29-35:

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}). This concept is generally valid and can be applied to any kind of transition in a system defined as binary either by intrinsic or operational parameters.

P10 L4: "aerosol particles"

See discussion above

P10 L6: "aerosol particles", but more importantly, what properties are distributed around a mean value? If they are physical (e.g., dpg, sigmag, etc.) maybe. If they are chemical (e.g., nitrate content) then not really.

From a very fundamental point of view, even the chemical compositions of aerosol particles from rather pure source (maybe sea salt) varies slightly between the particles. Often this distribution is so narrow that it does not matter. That is why it is often ignored.

To avoid any confusion of the reader we change the wording from:

> However, this is rarely as in reality an aerosol population consists of aerosol particles, whose properties are typically distributed around a mean-value (e.g. the mode of a particle size distribution).

To (P11 L11-23):

However, this is not the case for many parameters. In case of the particle diameter, for example, every aerosol particle has its individual diameter and the total population can be

described by a distribution of particle diameters around a mean diameter. An eventual size-selection does impact the mean diameter and the width of the distribution. Still, the size selected particles will not have the identical diameter.

P10 L7-9: I don't follow the logic here. If I understand correctly, the authors are saying that, due to multiplicity of charges on some aerosols, an aerosol population that follows a lognormal distribution if plotted by mobility diameter doesn't follow a lognormal distribution by aerodynamic diameter? I don't see how an aerosol population that is unimodal in mobility diameter can be multimodal in aerodynamic (or geometric) diameter.

The mobility diameter is a function of size and shape of the particles. The aerodynamic diameter is a function of the density as well. If the effective density of aerosol particle changes with the diameter, both size distribution can be quite different. For soot particles this often the case. Additionally a DMA selects particles that are monodisperse with respect to their mobility in an electric field. Nevertheless the same particles have a multimodal distribution of their mobility diameters due to different charges.

We clarified this in the manuscript.

Changed from:

> While the aerosol population might be mono-modal and narrowly-distributed with respect to one parameter such as the aerosol particle's electrical mobility diameter, it can be multi-modal and broader distributed with respect to another parameter (e.g. aerodynamic diameter).

To:

> Furthermore the aerosol population might be mono-modal and narrowly-distributed with respect to one parameter such as the aerosol particles electrical mobility diameter, but it can be multi-modal or broader distributed with respect to another parameter e.g. the aerodynamic diameter. Therefore, it has to be expected that the activation time (t_{act}) is also characterized by a distribution.

P10 L15: Maybe "…has the potential to activate." instead of "…activates.", because after t=180 min, they don't all activate.

Based on our definition all particles older than t_{act}=180 min are CCN active, but not all particles are older than t_{act}. We thank the reviewer for the comment and the sentence now ready:

P11 L24-25:

This curve was calculated based on the assumption of uniformity, i.e. every aerosol particle that is older than t_{act} = 180 min is CCN active.

P10 L17: Why was 30 min chosen as standard deviation?

The numbers presented here are chosen to represent an exemplary Gaussian distribution. In addition, these values are close to our experimental value and we hope to improve the readability of the text by avoiding to discuss the same aspect with significantly different values depending on the section of the manuscript (3.3 vs. 4). The respective sentence now reads:

P11 L26-29:

> To discuss the impact of an activation time distribution $P(t_{act})$ on the evolution of AF in a CSTR we consider a model system with $P(t_{act})$ representing a Gaussian distribution with an exemplary mean (μ) of 180 min and an exemplary standard deviation (σ) of 30 min (eq. (12)).

P14 L1: Unless I'm mistaken, tacts don't really differ; only tact-onset for PGaussian differs.

This is a theoretical comparison of different ways to obtain t_{act}. The system is idealized and therefor no instrumental uncertainties affect the outcome. Therefore any deviation between the numbers is significant. We now have extended the discussions of the numbers presented in Table 2 in order to present the comparability of our parameters to the literature. The respective text now reads:

P14 L1-18

As can be seen in Table 2, the individual values deviate with the biggest deviation in the case of t_{act} -onset. However, the presented deviations are solely caused by the underlying distributions of the activation time. In addition, t_{act}-onset, t_{act}, and $t_{act}0.5$ are determined at different experimental times. While t_{act}-onset is directly determined by measuring the entire particle population within the CSTR (global AF), t_{act} is calculated from the global AF in steady state and $t_{act}0.5$ is obtained from the activation time distribution itself. In the case of t_{act}-onset, there is a significant share of particles activating significantly earlier than the nominal activation time ($\mu = 180$) 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. Opposite to this, the threshold value of 0.01 is crossed later than the nominal activation time in the case of the step distribution. This is because even though every single particle activates after exactly 180 min of individual aging time, it takes some additional time before a fraction of 0.01 of the entire particle population within the CSTR is older than 180 min leading to a t_{act}-onset of 185 min. The difference of 10 min in t_{act} between the two $P(t_{act})$-approaches is due to the application of eq. (8) which allows for the calculation of t_{act} from the global AF in steady state. Strictly speaking, this equation is defined for the ideal step function ($P_{step}(t_{act})$) only. Therefore the higher global AF value for $P_{gaussian}(t_{act})$ in steady state has to lead to a lower t_{act} value compared to $P_{step}(t_{act})$. Note, $t_{act}0.5$ is referring to the particle activation distribution $P(t_{act})$ only leading to a concordant value of 180 min in both cases. This can be seen in Graph C of Fig. 4, where 0.5 of the particles with a residence time equal to the nominal activation time are activated in the case of a Gaussian distribution corresponding to $t_{act}0.5$. In the case of a step function, all particles are activated once the respective particle population is older than t_{act}. In the following we will show how the actual activation time distribution $P(t_{act})$ can be retrieved from real CSTR experimental data.

P14 L12: Fix "tact", but more importantly, please address the Major Comment surrounding this sentence (the lag in Fig. 3 before tact).

done

P14 L13: Fix "P(tact)"

done

P14 L15: Fix "Pstep(tact)".

done

P13 L5-6: Please provide appropriate citations.

Changed from:

> In the case of CCN-activation it is either an SS-onset often characterized by a minimum threshold (e.g. 1 % **AF**) or a critical SS, when 50 % of the particles activate.

To (P14 L11-14):

> Results from batch chamber experiments as well as from oxidation flow reactor experiments are often presented in terms of SS-onset or critical SS. While the SS-onset is defined by a minimum threshold (e.g. 0.01 *AF*) the critical SS is reached when 0.5 of the particles activate (Friedman et al., 2011; Koehler et al., 2009; Rose et al., 2007).

P14 L21: I would appreciate either a description of the chamber or literature that describes it. I'd really like to know, as I think is important for the reader, if this chamber is indeed well mixed (does it have impellers, fans, baffles?) to where the equations can be applied to the data, or is this chamber not really well mixed? What about residence time in the tubing? The tracer data may require some convincing (see four comments down P15 L6).

We picked up this comments and implemented it in the manuscript.

Changed from:

> In the laboratories at ETH Zurich we performed aging experiments in a 2.78 m3 stainless steel aerosol chamber run in CSTR mode.

To (P15 L20-24):

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.

P14 L22: For those not familiar with soot generation, what is a miniCAST, set point 6?

Changed from:

> We investigated the change in CCN-activity of soot particles rich in organic carbon from propane combustion (miniCAST, set point 6) due to heterogeneous ozone oxidation.

To (P15 L25-29):

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).

P14 L30-35: Would the authors see fit to put these two points at the end of the Introduction Section?

We thank the reviewer for this suggestion, but in view of the complex nature of the concept discussed within this manuscript the authors are worried that moving this information to the beginning of the manuscript might confuse readers not that familiar with chemical reaction chamber designs as the reviewer.

P14 L31: Again, I'd encourage the authors to refrain from using the word 'perfectly mixed' when talking about a real reactor. Might I suggest 'well-mixed'. More to my point: no RTD is available until Fig. 6; can a description of the chamber, or literature on it be presented?

Changed from:

> 1) Can the aerosol chamber be operated in CSTR-mode for up to 12 h which requires a constant aerosol feed-in flow and a perfect internal mixing?

To (P15 L40-41):

1) Can the aerosol chamber be operated in CSTR-mode throughout an entire day, which requires a constant aerosol feed-in flow and a good internal mixing?

P15 L6: Following the comment above: How the particles depict a CSTR would be more believable if the authors provide some way of showing it. Maybe plot an E-curve for the data and overlay that of an ideal CSTR over it? If I calculated it right, 2.78 m3 / 25 LPM is ~111 min. Why is tact more than twice that? In P7 L7 the authors claim tact is one mean residence time for a CSTR. If their chamber is not as well mixed as believed that's OK, but it should be stated (and at least be better mixed than OFRs!).

The authors apologize for not being clear enough in the definition and introduction of the activation time t_{act}, the mean residence time τ, and the individual residence time (i.e. particle age) in the original manuscript. The respective text now reads:

P16 L4-32

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.

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 ($\tau_{wall-loss}$) 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).

Based on the discussion above, the measured AFs (red crosses) show the expected change throughout the entire experiment in Fig 6 A1 and B1. In the beginning of both experiments AF is 0. After a minimum aging time each AF starts to increase until it reaches a constant level (A1: AF = 0.091, 1.0 % SS; B1: AF = 0.233, 1.4 % SS). 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. In the flushing regime, each measured AF increases exponentially. CCN data could be acquired successfully throughout the entire experiment until the global AF reached ~1.0 (> 1000 min) in the first experiment presented in graph A1. In the second experiment presented in graph B1, instrumental issues

caused the acquisition of the global *AF* to end prematurely after approx. 800 min of experimental duration.

Tables and Figures:

Figure 1: Please indicate a unit for the x-axis (I think it's seconds). Also, this figure is confusing because it should just be one curve representative of SS, but the authors mention in the caption "…while flushing the CSTR." I understand what the authors mean, but maybe the reader won't so this figure or its citation in the text should be made clearer.

We thank the reviewer for pointing out this issue. We have redesigned the entire figure to increase its readability. Please note, the values of the x-Axis are multitudes of the hydrodynamic residence time. The caption has been revised appropriately.

Caption change from:

> Figure 1: RTD inside the CSTR for steady state and for different time steps (multiples of τ) while flushing the CSTR. The area below the curve is proportional to the fraction of aerosol particles at a specific residence time. The individual residence time of a specific particle fraction is indicated by the color coding.

To:

> Figure 1: RTD inside the CSTR within steady state (black line) and for different time steps after the CSTR operation was switched to the flushing regime. The area below the curve is proportional to the fraction of aerosol particles at a specific residence time. The individual residence time of a specific particle fraction is indicated by the color-coding. The time on the x-axis is plotted as dimensionless time in multiples of the hydrodynamic residence time τ_{CSTR}.

In addition, we now highlight in the text only the solid black line labeled with "steady state" represents SS while the other curves represent the RTD at increasing time increments after switching to the flushing regime. The respective text now reads:

P3 L7-15

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}.

Figure 2: No major comments.
Figure 3: No major comments here, other than the curiosity of how a graph like this would look like for a PFR.

The particle concentration / F-curve from Mitroo et.al. would be a Step function. The AF-curve for ideal PFRs with no activation time distribution is either 0 or as step function as well.

Figure 4: No major comments.

Figure 5: Upon seeing Fig. 5, I struggle to now understand Fig. 3 (or, the blue line in Fig. 5). I was under the impression tact is when reactants are introduced. If that is the case, why does the red line show AF > 0 at t < tact? Or am I missing something? A CSTR has no lag by design; only PFRs have lags. Even in the 'filling regime'. I think the root of my misunderstanding can be traced back to P7 L7. Why is AF = 0 when t < tact? Even for a system with no Gaussian spread, purely based on CSTR design, at t = 0+ AF (however small) is non-zero. If the authors can explain their assumption in P7 L7, I think it would clear this up (at least for me).

The authors hope to have improved the introduction of t_{act} chapter 3 in this revised version of the manuscript as t_{act} is the time needed to modify a single particle to such a degree that it is CCN active. t_{act} is **not** the time span since when reactants are introduced into the reaction chamber as this is defined as the experimental duration t.

Table 1: No major comments.
Table 2: No major comments on the table itself (maybe capitalize the subscript 'gaussian'?); but I have comments on how the authors choose to explain the difference in values of tact-onset for Step and Gaussian (see comment section).

Done in the comment section

Table 3: No major comments.
Figure 6: No major comments, but I do have a question: it's unclear how the authors' fit matches data well. Was it a fit? E.g., if instead of soot they used salt, what is needed experimentally to determine the blue dotted line in this Figure? Did I miss something in the text?

Changed from:

> The **P($t$act)** presented in A2 and B2 of Fig. 6, respectively, were obtained by performing a curve fitting operation assuming
> **P($t$act)** to be a mono-modal Gaussian distribution and with the parameters **μ** (= $t$act0.5) and **σ** to be optimized.

To:

The graphs A2 and B2 in Fig. 6 show the activation time distribution $P(t_{act})$ (blue solid line) retrieved from the measured global AFs. The $P(t_{act})$'s presented were obtained from curve fitting the measured AF-curves using eq. (13), which describes the evolution of AF taking the activation time distribution into account. For this, assumptions concerning the type of distribution had to be made. Here, we assumed that $P(t_{act})$ can be described by a mono-modal Gaussian distribution as presented in eq. (12). A brute-force algorithm was used that optimized the characteristic parameters μ (=mean) and σ (=standard deviation) in order to achieve the best fit to the measured global AF using the least-square method. The results of this fitting procedure are presented in Table 3 as well as in A2 and B2 of Fig. 6. In the first experiment with the experimental settings at 1.0 % SS and 100 ppb O_3 μ as well as σ of $P(t_{act})$ are larger (253.7 min and 35.5 min) compared to the results obtained for the second experiment at 1.4 % SS and 50 ppb O_3 (153.6 min and 24.6 min). From a theoretical perspective, there are two competing aspects. On the one hand, due to the higher ozone concentration the threshold of chemical transformation leading to CCN activity of the particles should be reached earlier. On the other hand, the threshold of chemical transformation should be lower at higher SS. Our results presented here could indicate that the difference in SS in this specific range might be more important than the difference in ozone background concentration within the considered range. At the current stage we cannot draw any final conclusions on how these two competing aspects actually interplay but additional experiments are planned to resolve this issue.

In addition, we list t_{act} obtained from AF during steady state following eq. (8) as described in section 3.1.2 in Table 3. Based on error propagation calculation, the instrumental uncertainty for obtaining t_{act} from steady state is ± 11.6 min. In our experimental setup the differences between t_{act} and μ are 3.9 min and 2.1 min, respectively, and therefore below the instrumental uncertainties. This is a very beneficial aspect when considering a broad application of the CSTR-concept in atmospheric science experiments. In general, an accurate determination of $P(t_{act})$ requires a sufficiently high time resolution throughout the whole experiment. This can be difficult to achieve depending on the general experimental conditions such as the type of instrument, since running SS-scans with a CCNC can be time consuming. However, if a characterization of the aged aerosol during steady state is sufficiently precise, a potentially time consuming acquisition of a large number of data points for the determination of $P(t_{act})$ does not provide additional benefits.

Figure 7: No major comments, but to be clear, is this illustrative? That AFTPOT > AFPAM at high [OH], and the reverse for low [OH], is subject to experimental data, right?

The specific values for t_{act} are chosen for illustrative purposes. The AF-values are theoretical calculations based on the framework presented here and match the trend seen in the experimental data as presented by Lambe et al. 2011.