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 (t_{act}) 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 t_{act} were achieved for their experimental data. The t_{act} 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 t_{act} 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.

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. 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?

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? 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.

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 (t_{act}) 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 t_{act} or RTD concept for CSTR system into the data interpretation of OFR/PAM reactors? Necessary information is needed to clarify this point.

   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 liquid-liquid 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 t_{act} 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.

Specific comments:

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

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. 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?

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

4. Page 6, line 20: As a crucial parameter introduced in this study, the activation time (t_{act}) 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? Is it easy to achieve in practical conditions of laboratory chamber experiments?

5. **Equation 7**: I think it should be "$e^{\frac{-t_{switch}}{\tau_{CSTR}}}$" in the exponent. Beside, it's better to add a pair of parentheses for "$-\tau_{CSRT}$", since it appeared after "·".

6. **Equation 8**: Why is the simplified equation not expressed as '$t_{act} = -\ln(AF(t))\cdot\tau_{CSTR}$'? I'm wondering how will the value of $AF(t\rightarrow\infty)$ 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.

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?

   Line 10: *"... and therefore the global AF only if t_{act} = t_{switch}."* Some information was missed in this sentence.

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

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.

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

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

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.

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.

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.

Technical corrections:

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

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

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

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

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

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.

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.

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

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

10. Page 14, line 8-9: *"As there is a significant share of particles activating significantly earlier than the nominal activation time (*$\mu = 180$*) 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.

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 t_{act} was written as tact. Similar issues also exist in some other expressions, e.g., Pstep, which should be P_{step}. 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:

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* $t_{act}0.5$*."*

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

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

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."*

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

17. Page 15, line 18: *"…* μ *as well as* σ *~~are~~ is larger for* $P(t_{act})$ *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."*

18. Page 15, Line 23: The comma between "$P(t_{act})$" and "requires" is unnecessary.

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

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

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

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

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

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