We thank the referee for the detailed review of our manuscript. Our responses are in blue.

1. (General comment): There are a large number of equations in the manuscript; I found some of these hard to follow. If it is important that the derivation of these equations (e.g., Eq. (3) or Eq. (7)) be understood by most readers, the authors should provide a more detailed derivation in the supplemental material. My feeling is that if the reader will accept the derived results without fully understanding the derivation (as I did) they will still be able to make use of the equations and understand the work as presented.

   The solution of equation (1) can be carried out by first solving the pure diffusion problem in the absence of reaction. This is the standard problem of diffusion (or heat conduction) in a sphere. Solutions can be found in the classic books: Crank, Mathematics of Diffusion (1975) and Carslaw and Jaeger, Conduction of Heat in Solids (1946). The pure diffusion solution can be extended to the case of first-order reaction using the method of Danckwerts, on page 330 of Crank. Eq. (7) is quite easily obtained by integrating Eq. (3) over the volume of the particle. Thus we feel it is not necessary to include the step-by-step solutions for these equations in the supplementary material.

2. Page 28636, line 3: It seems that the symbol kc refers only to the bulk reaction as it is defined as the pseudo-first order approximation. Do the authors differentiate surface reactions from bulk reactions? If not, the decision to disregard surface reactions should be justified (see e.g., the importance of the surface reaction of OH and Cl^- in Laskin et al. 2003 among many others).

   Bulk and surface reactions are not explicitly differentiated in the present framework. A reaction would essentially become a "surface reaction" if the reaction timescale ($\tau_c = 1/k_c$) is much smaller compared to the particle-phase diffusion time scale [$\tau_{da} = R_p^2/(\pi^2 D_b)$], such that there is sufficient time for the solute to be completely consumed by the reaction at or near the surface before it can diffuse into the bulk of the particle. These bulk vs. surface reaction scenarios are already discussed in Sect. 2 and illustrated in Fig 3. The reaction between OH and Cl- in Laskin et al. (2003) was treated as a very fast chemical reaction (a net probability for uptake and reaction of 0.85) and the transport of OH from the gas phase to the surface was essentially gas-phase diffusion limited. Such reactions are already handled in the present framework. Special cases such as adsorption on solid surfaces followed by chemical reaction are presently not implemented in the framework, but can be easily added as and when necessary.

3. Page 28638, line 8-10: The decision to invoke the pseudo-first order (PFO) approximation needs to be more fully justified. As discussed in Eq. 8 of Berkemeier et al. (2013), when the reacto-diffusive length is expressed in terms of a PFO rate constant it is implicit that the reactant in excess is well mixed depthwise throughout the particle. In my opinion this calls into question the validity of using a PFO rate constant throughout this work as the bulk reactant need not be homogeneously distributed depth-wise, e.g., when the bulk of the particle is solid or nearly-solid.

   The first-order approximation essentially means that we model the dynamics of chemical reaction in the particle with a time scale, the inverse of k_c. If the time scale for the reaction is long as compared to that for diffusion, then the reaction takes place essentially uniformly in the particle. If the time scale for the reaction is short as compared to that for diffusion, then the solution predicts that the reaction proceeds as a front that propagates slowly into the particle. This latter case would be appropriate for a semi-solid particle, for example. Thus, the solution itself automatically accommodates both extremes of time scale. The only issue of the validity of the first-order assumption is whether the actual chemistry itself can be modeled by a single time scale. In the case of a chemically complex SOA system, the PFO representation provides the type of order-of-magnitude estimate that is desired, especially when trying to parameterize the model for use in regional and global models. However, it may be possible to parameterize the PFO rate constant for a condensing solute in terms of its second order rate constant multiplied by the average concentration of the reactant solute in the particle phase even when the reaction time scale is shorter than that for diffusion. Additional work, beyond the scope of the present manuscript, is needed to test and validate this approach. We have acknowledged this issue at the end of Sect. 3.2 in the revised draft manuscript.

4. Page 28639, line 18: The statement here that the timescale of diffusion varies as the radius squared is true in the case of Fickian diffusion. However, in glassy particles, diffusion fronts may move linearly inward, leading to a linear dependence on particle radius (see Zobrist et al. 2011), if the material being taken into the particle is a plasticizing agent such as water.
   This point concerning the timescale of diffusion in glassy particles is well taken. We have added a clarifying statement in the text concerning this issue.

5. Page 28641, line 3-8: Were the times QSS calculated using an assumption of Fickian diffusion (see note 6)?
   Yes.

6. Page 28642, line 9-11: the assertion that all particles with $k_c > 10^{-2}$ s^{-1} are in quasi-steady state seems to be very important. The argumentation that supports it (lines 5-8) needs to be expanded for those that are less intimately familiar with the results of this study.
   This result is inferred directly from Fig. 6b, which shows that $\tau_{QSS} \leq 0.7$ min for solutes reacting with $k_c = 10^{-2}$ s^{-1} in particles of any size and any bulk diffusivity. The typical timescale for changes in the bulk gas phase concentrations due to transport and chemical reaction is of the order 10 min or more. Thus, from a practical standpoint, the particle-phase concentration profiles of solutes reacting with $k_c > 10^{-2}$ s^{-1} (which have $\tau_{QSS} < 0.7$ min) may be assumed to be at quasi-steady state in particles of any size and any D_b value. We have clarified this point in the second last paragraph of Sect. 2 of the revised manuscript.

7. Page 28644, line 14-15: Using Raoult's Law implies that the surface is in equilibrium with the gas phase. This assumption needs to be supported given that particles can often be out of equilibrium with the gas phase.

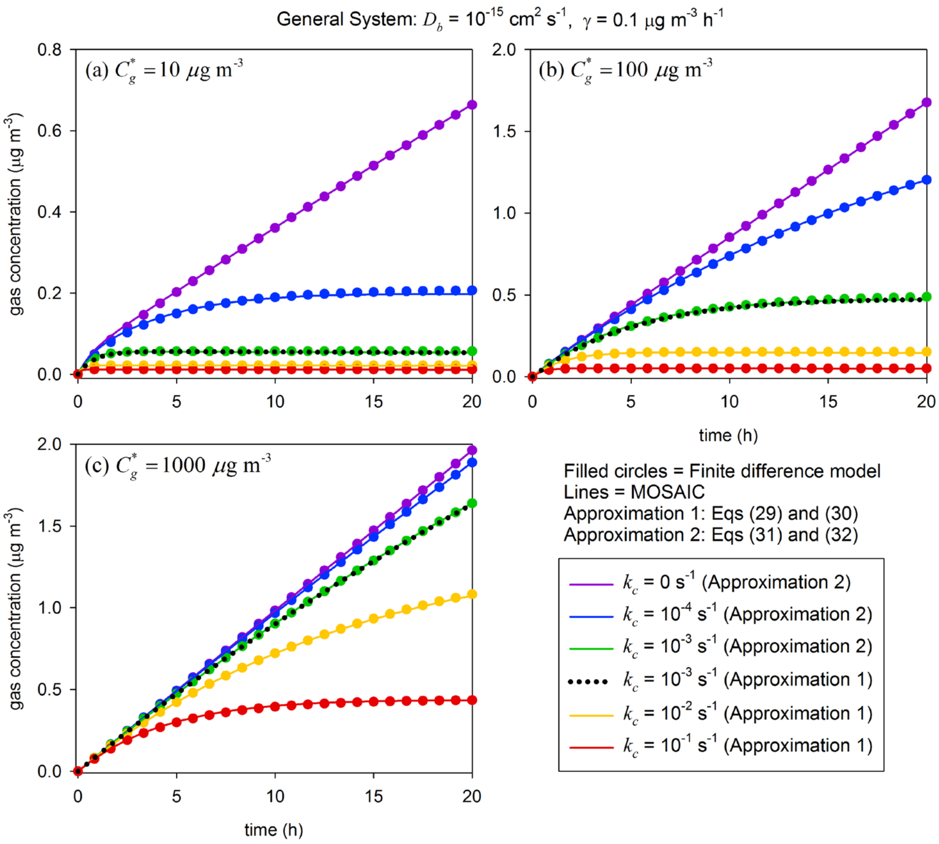
   The bulk of the particle can be out of equilibrium with the gas phase, but phase equilibrium at the interface is achieved rapidly. We have added the following text in the manuscript to support this assumption: "The timescale to achieve interfacial phase equilibrium between $C_{g,i}^{s}$ and the particle-phase concentration of i just inside the surface, A_i^s (mol cm^{-3}(particle)) is at least (Seinfeld and Pandis, 2006):

$$\tau_{p,i} = D_{b,i}\left(\frac{4}{\alpha_i \overline{v}_i}\right)^2, \qquad (15)$$

   where $\overline{v}_i$ is the average speed of solute molecules in the gas phase. From kinetic theory of gases $\overline{v}_i = (8\Re T/\pi M_i)^{1/2}$ where $\Re$ is the universal gas constant (8.314 X 10^{7} erg K^{-1} mol^{-1}), T (K) is temperature, and M_i is the molecular weight of the solute. For representative values of $D_{b,i} \le 10^{-5}$ cm^{2} s^{-1}, M_i = 300 g mol^{-1}, T = 298 K, and α_i ranging from 0.1 to 0.001, the value of $\tau_{p,i}$ is of the order 10^{-6} s or less, which means it can be safely assumed that the interfacial phase equilibrium is achieved virtually instantaneously."

8. Page 28646 – 28647: Is there a transition regime between the two approximations? How accurate is the assumption that behavior changes suddenly from one approximation to the other?

   This is a good point. We repeated the solutions in Fig. 10 for k_c = 10^{-3} s^{-1} with Approximation 1 and found that they are nearly identical to Approximation 2 for the same k_c value. Thus, the transition from one approximation to the other does not cause a sudden change in the behavior of the solution. We have added this result in the revised Fig. 10 (given below) and clarified it in Sect. 3.2.2.



Revised Fig. 10.

9. Page 28661, section 5: are there any systems that are not well-represented by this framework? Are there transition regimes that potential users of this framework must be aware of?
   We have added a new “Sect. 3.3 Future Considerations”, which outlines three issues that must be taken into consideration when specifying the actual physical and chemical details in the proposed framework. It is reproduced below for convenience:

3.3 Future Considerations

While the general system framework is amenable for eventual use in regional and global climate models, it currently awaits specification of the various gas and particle-phase chemistries important for SOA formation. The following issues must be must be taken into consideration when specifying the various physical and chemical details in the model and evaluating it using laboratory and field observations.

First, the present framework uses a pseudo-first order (PFO) reaction for a condensing solute as a proxy for second order chemical reactions that may occur within a particle. The assumption of PFO reaction for the condensing solute is valid when the pre-existing bulk reactant species is uniformly distributed with the depth of the particle, e.g., when the reaction timescale for the reactant species is much longer than that for diffusion. The issue arises when the reaction time scale is much shorter than that for diffusion such that the bulk reactant species is not homogeneously distributed depth-wise (Berkemeier et al., 2013). In such cases, it may be possible to parameterize the PFO reaction rate constant for the condensing solute in terms of its second order rate constant multiplied by the volume average concentration of the pre-existing reactant solutes in the particle phase. The detailed finite difference model using second order reactions can be used to provide guidance for improving and validating the parameterized reactions in the semi-numerical framework.

Second, while the present framework allows particles of different sizes and composition to have different bulk diffusivities, it cannot explicitly treat the potential variation of diffusivity within a given particle of complex morphology. Examples include black carbon or solid ammonium sulfate particles coated with organics as well as particles with non-ideal internal mixtures of hydrophobic and hydrophilic organics. The diffusion-reaction process inside such complex and potentially non-spherical particles will again have to be parameterized based on the average bulk properties, with possible guidance from more detailed finite difference models where applicable.

Third, as mentioned earlier, the new framework can be readily adapted to kinetically partition water soluble organic gases into the particulate aqueous phase if that is the only liquid phase in the particle. However, additional work is needed to extend the present framework to mixed inorganic-organic particles in which water and organics may form separate liquid phases (You et al., 2012).

Minor comments:

1. Page 28637, line 12 (equation 4): Is this really just the ratio of the particle radius to the reactodiffusive length? If so, it would be helpful to point this out for those readers that are not intimately familiar with this sort of framework.
   Yes, the dimensionless parameter q can be viewed as the ratio of particle radius to the reacto-diffusive length. We have added text to this effect (before Eq. 4) in the manuscript.

2. Page 28639, lines 16-17: If the phrases "surface-area controlled" and "volume controlled" have a specific definition, a reference would be appropriate here. These phrases imply to me that the behavior of the system scales with radius squared (surface area) or cubed (volume). However, Brekemeier et al. (2013) recently showed that several possible responses to changes in radius occur, ranging from no effect on the normalized time to react a well-mixed particle limited by chemical reaction in the bulk (Brx) to an inverse-square dependence on particle size for systems limited by bulk diffusion (Bbd). Note that these scenarios are all for systems in which the bulk loss dominates the reactive uptake, and that Table 4 of Berkemeier et al. is normalized so that the intuitive volume controlled cubic dependence on radius appears as zero.
   We have rephrased that sentence to: "Thus, the particle growth is volume reaction-controlled when the concentration profile is uniform and tends to be surface reaction-controlled at the other extreme (Seinfeld and Pandis, 2006)."

3. Page 28642, line 2-4: Instead of separating particles by D_b, it seems that q would be a better metric for which particles will be in steady state. Since these are non-reacting particles q is necessarily zero,

but perhaps an analog such as $R_p/\mathrm{sqrt}(D_b)$ would be more appropriate.

Unfortunately, q is not a unique indicator of which particles will be in steady state. For example, a particle with $R_p = 10^{-5}$ cm, $k_c = 10^{-1}$ s^{-1}, and $D_b = 10^{-11}$ cm^2 s^{-1} has q = 1 and τ_{QSS} = 0.008 min. Another particle with $R_p = 10^{-5}$ cm, $k_c = 10^{-4}$ s^{-1}, and $D_b = 10^{-14}$ cm^2 s^{-1} also has q = 1, but τ_{QSS} ~30 min. The first particle would reach quasi steady state much more rapidly compared to the second particle.

4. Page 28644, line 13: Are there any assumptions that go into Eq. (14)? If so, they should be re-stated here.
   We have briefly described the source of Eq. (14) and cited Seinfeld and Pandis (2006) for further details on the use of this correction factor.

5. Page 28648, line 2: The symbol for particle-side mass transfer k_b conflicts with the PRA framework definition (Poschl et al. 2007) and is potentially confusing as many people use this symbol for the PFO bulk chemical reaction. Could a different symbol be used for the quantity?
   The particle-side mass transfer symbol has been changed to k_p and the particle-side film thickness is now denoted by δ_p.

6. Page 28654, line 15-17: the authors note that the y-axis labels on figure 11 should be interpreted as the decadal logarithm. Why not include this directly on the y-axis label?
   "$\log_{10}$" is now directly included on the y-axis labels.

7. Page 28659, line 3-5: do all small particles grow much faster than large ones, or only semi-solids? The text as written implies that only semi-solid particles experience this difference in growth rate.
   In general all small particles grow faster than the large ones. But if all particles (small as well as large) are semi-solids (in the present case with $D_b = 10^{-15}$ cm^2 s^{-1}), there will be a steep gradient in Q across the size distribution (e.g., From Fig. 7 we see that $Q \approx 0.7$ for D_p = 0.02 µm and $Q \approx 0.1$ for D_p = 0.2 µm). In contrast, $Q \approx 1$ for liquid particles of all sizes. This steep gradient in Q for semi-solid particles is responsible for the much faster growth of small particles than the large ones when compared to the liquid particle case.

   We have therefore clarified the sentence in question as indicated by the red text: "However, significant narrowing is still seen for $D_b = 10^{-15}$ cm^2 s^{-1} due to the steep gradient in *Q* across the size distribution, which causes the small semi-solid particles to grow much faster than the large semi-solid ones when compared to the corresponding liquid aerosol case where $Q \approx 1$ across the entire size distribution."

8. Page 28666 Line 7: The reference to Eq. (S12) seems out of place here and might be a typo.
   This is indeed a typo and has been corrected to Eq. (B3).

9. Page 28681, Figure 6, panels b-d: what would happen if the lines were plotted using different values of q instead of different values of D_b? Might this simplify the figure by collapsing some of the

variation between panels?

For a given kc, the plot of τ_{QSS} vs. D_p will yield horizontal lines for different values q. However, we have chosen to plot the lines for different values of D_b so that the effects of all three independent variables (k_c, D_p, and D_b) can be understood at a glance rather than having to calculate D_b values for different combinations of k_c, D_p, and q. And because q by itself is not a unique metric for steady state (as exemplified in response to minor comment #3), it is not possible to collapse the different k_c panels into one.