

Interactive comment on “Impact of chamber wall loss of gaseous organic compounds on secondary organic aerosol formation: explicit modeling of SOA formation from alkane and alkene oxidation” by Y. S. La et al.

### Author Response

We thank the two anonymous referees for their comments and suggestions. We respond below to the points raised by each referee and summarize the changes made to the revised manuscript.

### Response to referee #1

#### General comments

**1. The representation of gas-wall partitioning is a bit confusing. If I understand correctly, the parameters on the right hand side of Equation (1) are all constants, which is reasonable since this equation gives the relationship between the deposition rate on the wall ( $k_{gw}$ , eq) and evaporation rate from the wall ( $k_{wg}$ , eq) at equilibrium. Then, the question is how to represent  $k_{gw}$  and  $k_{wg}$  at different time steps? Here,  $k_{gw}$  largely depends on the gas phase diffusion and wall accommodation, and generally follows a first-order decay trend. On the other hand,  $k_{wg}$  depends on the amount of total absorbing organic mass in the chamber wall. For example, an organic vapor ‘A’ is injected into the chamber. At  $t = 0$ , all of the vapor ‘A’ is present in the gas phase and there is zero amount of ‘A’ on the chamber wall and thus  $k_{wg,A}(t = 0)$  should be zero. As more and more vapor ‘A’ accumulates on the chamber wall due to deposition, the evaporation rate of ‘A’ continues increasing, and eventually, the deposition and evaporation rates of ‘A’ reach equilibrium. My point is the Teflon film can be treated as a giant absorbing organic mass, with its amount characterized by  $C_w$ , which is a constant. But for individual species, their deposition/evaporation rates depend not only on  $C_w$ , but also on the amount of their own masses on the wall. The authors need to clarify this.**

The gas to wall and wall to gas transfer rates of a given semi-volatile organic compound (SVOC) depends on (i) the concentrations of the SVOC in the gas phase and on the wall at a given time and (ii) the gas/wall and wall/gas rate constants. The temporal variation of the concentration of a SVOC in the gas phase due to the gas/wall transfers is given by the following expression:

$$\left[ \frac{d[SVOC(g)]}{dt} \right]_{gas/wall} = -k_{gw}[SVOC(g)] + k_{wg}[SVOC(w)] \quad (1)$$

where  $[SVOC(g)]$  and  $[SVOC(w)]$  are the concentration of the SVOC in the gas phase and on the wall at a given time, and  $k_{gw}$  and  $k_{wg}$  the first order rate constants (assumed to not depend on time) for the transfer of a SVOC from the gas to the wall and from the wall back to the gas phase respectively.

The expressions of the temporal variation of the concentration of a SVOC in the gas phase due to the gas/wall and also to the gas/particle transfers will be added in the paper for clarification. The term “rate” was a few times used in the paper instead of “rate constant”. This will be corrected in the new version of the manuscript.

**2. Could the authors consider adding more discussions regarding the impact of gas wall partitioning on SOA composition? Particularly:**

**2.1) Are the wall loss rates the same for all the products generated from one hydrocarbon? Or different wall loss rates are applied to individual compounds based on their chemical properties, such as vapor pressure?**

The time rate of change of the gas phase concentration due to the gas/wall and wall/gas mass transfer is given by equation (1) above. In the Matsunaga and Ziemann (2010) parameterization for gas/wall transfers, it is considered that:

- the gas transfers towards the walls are driven by the turbulence inside the chamber and interfacial mass transport onto the walls;  $k_{gw}$  is thus a constant, independent of the compound structure (e.g. Yeh and Ziemann, 2015).

- the gas/wall partitioning equilibrium follows the Raoult's law ;  $k_{wg}$  is calculated based on  $k_{gw}$  and the equilibrium constant (see Eq. 1 in the paper).  $k_{wg}$  depends on the temperature, the volatility of the species and the  $C_w/(M_w\gamma_w)$  value. Matsunaga and Ziemann (2010) optimized  $C_w/(M_w\gamma_w)$  values from chamber observations and reported values of 9, 20, 50 and 120  $\mu\text{mole m}^{-3}$  for n-alkanes, 1-alkenes, 2-alcohols and 2-ketones respectively (see p23897 I9).

The following sentence will be added to discuss the values of  $k_{wg}$  (p23898 I12) in the revised version: "In the Matsunaga and Ziemann (2010) parameterization for gas/wall transfers, it is considered that the gas transfers towards the walls are driven by the turbulence inside the chamber and interfacial mass transport onto the walls;  $k_{gw}$  is thus a constant independent of the compound structure (e.g. Yeh and Ziemann, 2015)."

**2.2) Why the first generation products exhibit the most significant gas-wall partitioning, see the dodecane case in Figure 8? Shouldn't be the case that the lower the volatility of compounds, the more deposition occurring on the wall? Is there any other process that could affect the gas-wall-particle distribution of oxidative products, e.g., the chemical reaction timescale vs. the total simulated SOA growth duration?**

The exploration of the distribution of a species (with no oxidation process) between the gas, the particle and the wall shows that for times lower than 1 h (i.e. duration of alkane experiments), wall loss affects mostly species having a volatility between  $10^{-8}$  and  $10^{-5}$  atm (see Figure 2). Compounds of lower volatility would be impacted on longer timescales, exceeding the duration of these chamber experiments.

When oxidation occurs, the impact of wall loss will be indeed dependent on the chemical lifetime of the species. The range of SVOC+OH rate constants is between  $10^{-12}$  and  $10^{-10}$   $\text{molec}^{-1} \text{cm}^3 \text{s}^{-1}$ . For a typical experiment, [OH] is typically  $5 \cdot 10^7$   $\text{molec cm}^{-3}$ . The chemical loss rates due to OH oxidation therefore lie between  $5 \cdot 10^{-5}$  to  $5 \cdot 10^{-3}$   $\text{s}^{-1}$ . For the experiments simulated here, gas to wall transfer rate of a given SVOC ( $k_{gw} = 1.7 \cdot 10^{-3} \text{ s}^{-1}$ , with  $\tau_{gw} = 10$  min) could be of the same order of magnitude than the OH loss rates; wall loss could thus be in competition with OH oxidation. The various mass transfers and oxidation processes were therefore represented in this study dynamically.

Comparison of the mass transfer and oxidation rate characteristic times will be added p23899 I4.

The simulated results show that most species formed during the alkane experiments are 1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup> generation products, in agreement with observations. Species that exhibit significant gas/wall partitioning have vapor pressures in the  $10^{-8}$ - $10^{-5}$  atm range, as discussed above and in the paper (see Fig. 2, Fig. 8 and section 4.2).

#### Minor comments

**3. Page 23895, Line 19: The '0.35 yield unit' is a bit confusing, since the SOA mass yield ( $d\text{elM}/d\text{elHC}$ ) is actually unitless. Would it be better to give a relative increase, e.g., one order of magnitude, in the SOA yield upon vapor wall loss correction?**

The quantification of wall loss impact on final SOA yields will be presented in the revised version of the paper using a factor of increase/decrease on the final yield for a better understanding.

**4. Page 23896, Line 22: The authors may want to also consider citing Zhang et al. (2014, PNAS) since this paper systematically evaluated the impact of vapor wall losses on SOA yields generated from a series of biogenic and anthropogenic precursors.**

Thanks for the reference. The citation to the Zhang et al. (2014) paper will be added in the introduction section p23896 l21 and to the references.

**5. Page 23896, Line 23: The authors need to clarify in what way the loss of organic vapors on chamber walls is not well characterized, like the deposition rates of structurally different species? Or the impact on the SOA yield and composition?**

Very little is known on this process yet. Only a few studies have been published on the wall loss of organic vapors (see introduction). The processes involved in this wall uptake are not clearly understood, gas to wall and wall to gas rate constants have only been estimated for a few species and in very few chambers. The parameterizations used to describe this process are therefore empirically derived and may not be appropriated for another chamber. Species impacted by this wall loss during oxidation or SOA formation experiments are unknown and the impact on oxidation processes and SOA formation have never been quantified yet.

**6. Page 23897, Line 5: The expression of either  $k_{gw}$  or  $k_{wg}$  should be given.**

The discussion on the values of  $k_{wg}$  p23898 l6 will be moved before for clarity in the corrected version of the manuscript, p23897 l5.

**7. Page 23897, Line 16: Are the authors comfortable with this assumption? The gas phase diffusion onto particles should be really fast if one plugs numbers into Equation (2). Could the authors give some comments on different diffusion processes, i.e., gas phase diffusion, gas-particle interface transfer, and particle phase diffusion, that ultimately control particle growth?**

The gas-to-particle partitioning was described according to Matsunaga and Ziemann (2010) model. The gas to particle mass transfer was assumed here as limited by gas phase diffusion. Studies have however shown that the gas to aerosol mass transfer could be limited by the interfacial mass transfer and/or particle phase diffusion (e.g. Shiraiwa and Seinfeld, 2012; Zhou et al., 2013). Accommodation coefficients and aerosol viscosities encompass order of magnitude. No constraints are available for these parameters for the experiments examined here and these processes were ignored in the simulations. The value used for  $k_{gp}$  is therefore an upper limit if the interfacial mass transfer and/or particle phase diffusion limit the gas-to-particle partitioning.

The following sentence will be added p23897 l21: "This value for  $k_{gp}$  is an upper limit if the gas to particle transfers are limited by interfacial mass transfer and/or particle phase diffusion (Shiraiwa and Seinfeld, 2012). The wall uptake could then be underestimated."

**8. Page 23905, Line 16: Add ( $\Delta M$ ) after 'the ratio of SOA mass produced'. Also, specify the reaction time at which the SOA yields are measured.**

The ( $\Delta M$ ) will be added and the reaction time specified in the revised version of the paper.

**9. Page 23906, Line 25: What is the timescale with respect to gas-particle partitioning in the simulations?**

For the exploration of the distribution of a given species as a function of its volatility, gas to particle mass transfer rate constants were of  $k_{gp} = 2.6 \times 10^{-1} \text{ s}^{-1}$  and  $k_{pg} = 1.1 \times 10^6 P^{\text{vap}} \text{ s}^{-1} \text{ atm}^{-1}$  and gas to wall mass transfer rate constants were of  $k_{gw} = 5.6 \times 10^{-4} \text{ s}^{-1}$  and  $k_{wg} = 1.9 \times 10^2 P^{\text{vap}} \text{ s}^{-1} \text{ atm}^{-1}$ .

These values will be added in the revised version p23898 l18.

**10. Page 23907, Line 9: Could the authors explain why the discrepancies between simulated vs. observed SOA yields rise with the size of precursors for the terminal alkene cases?**

The experimental data shows that for the 1-alkenes series, the final SOA yield increases with the precursor's carbon number up to a plateau for  $C_{>13}$  species. The simulated SOA yields grow with the carbon chain length of the precursor, without showing any plateau. The implementation of organic

vapor wall loss does not explain this behavior. SOA formation from alkene oxidation is largely controlled by the fate of the  $\beta$ -hydroxyalkoxy radicals resulting from OH addition on the C=C bond in the gas phase. As discussed in the paper, sensitivity tests were thus performed on the SAR used to estimate branching ratios for alkoxy radical evolution, using the VER instead of the ATK configuration. The simulated SOA yields strongly depend on the Structure Activity Relationship (SAR) used to estimate the branching ratio for alkoxy radical evolution, however none of the SAR is able to represent the observed plateau for  $C_{>13}$  species.

The inability of the model to capture the constant yield observed for  $C_{>13}$  species points out that some chemical pathways are not well represented or missing in the GECKO-A mechanism (e.g. autoxidation processes, dimers formation and/or heterogeneous oxidation processes in the particle phase). The measurement of final SOA yields does not provide enough insights to identify the source of the model/measure discrepancies; information on SOA composition is required to go further.

This following sentence will be added p23908 l7: “The implementation of organic vapor wall loss or the use of a different structure activity relationship to estimate the fate of the alkoxy radicals does not fully explain the discrepancies between the model and the observations for the alkene series. Some chemical pathways are clearly not well represented or missing in the GECKO-A mechanism (e.g. autoxidation processes, dimers formation and/or heterogeneous oxidation processes in the particle phase...). The measurement of final SOA yields does not provide enough insights to identify the source of the model/measure discrepancies; additional information on SOA composition would be needed.”

**11. Page 23910, Line 6: It is interesting to see that the volatility domain where significant partitioning to walls belong to the semi-volatile regime. Is it because the precursor concentrations are at ppm level?**

The partitioning of a species to the wall depends mainly on the value of  $C_w/(M_w\gamma_w)$  and on the vapor pressure of the species. For the typical conditions used in these experiments, Fig. 2 shows that the species that significantly partition between the gas and the wall after 1 h have vapor pressures in the  $10^{-8}$ - $10^{-5}$  atm range (see Fig. 2), i.e. the semi-volatile vapor pressure range. This result is independent on the amount of species.

The high concentration of precursors used in this experiment will (i) increase the total (in the gas, in the particle and on the wall) concentration of a species and (ii) indirectly change the distribution of a species between the different phases by increasing the amount of organic particulate matter onto which SOA can be formed.

**12. Page 23919, Table 1: Are the initial CH3ONO concentrations actual measurements? If not, how did the author estimate their values? As shown in Figure 6, the simulated delHC masses are in general lower than the measurements. Is this due to the underestimation of initial CH3ONO mixing ratio?**

The initial mixing ratio of CH3ONO was calculated from the known quantity of CH3ONO injected inside the chamber (see Lim and Ziemann, 2009, Matsunaga et al., 2009 and Matsunaga, 2009).

The legend of Table 1 will be changed into: “Measured injected concentrations and environmental conditions of the experiments”.

**13. Page 23922, Figure 2: Label the three panels with gas-particle/wall timescales, i.e., 6 min, 1h, and equilibrium.**

The panels will be labeled with the simulation time in the revised version of the paper.

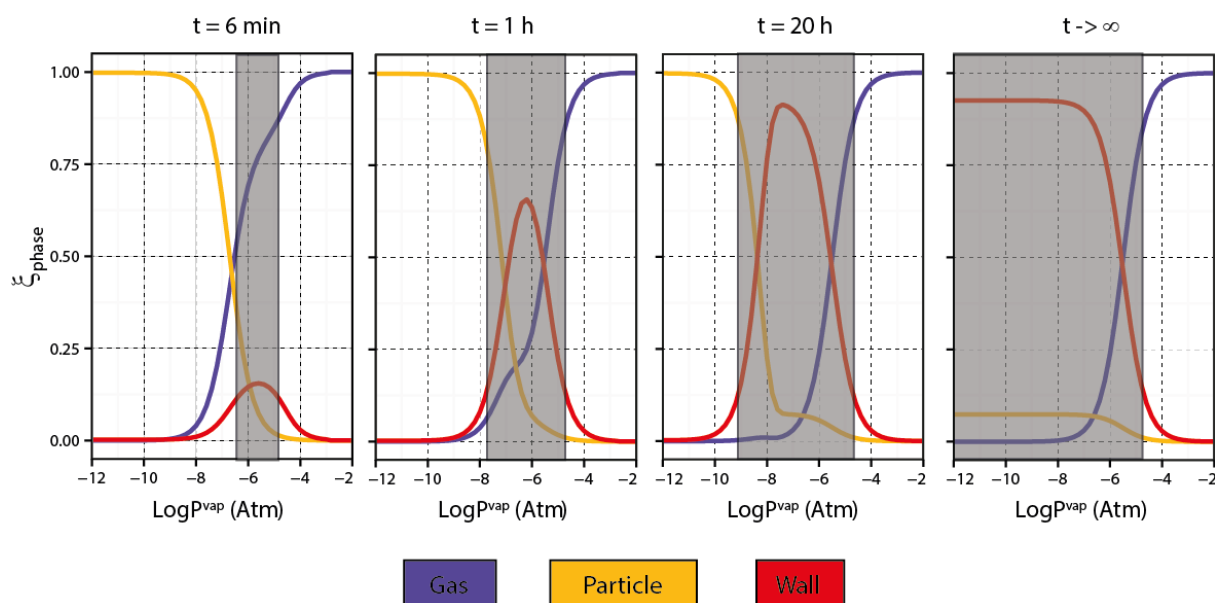


Figure 2. Time evolution of the fraction in the gas, particle and wall phases as a function of the vapor pressure for a continuous distribution of species considered to be initially only in the gas phase. Grey zones represent the volatility range impacted by wall losses at a molar fraction higher than 10%.

**14. Page 23924, Figure 4: Why the number of products from 2-methyl-1-alkanes photochemistry does not follow a nice trend like those generated from other alkane and alkene precursors?**

This was a mistake in the figure that will be corrected in the revised version of the paper.

### Response to referee #2

#### Major comments

**1. The Introduction section contains a thorough discussion of gas-particle and vapor-wall partitioning as equilibrium processes following Raoult's law. Figure 2 shows that the distribution of species at 1 h of photooxidation differs greatly from that at equilibrium (photooxidation duration not specified, but presumably longer than 1 h). It is also noted that at 1 h of photooxidation, partitioning is under kinetic, not thermodynamic control. The model is compared with photooxidation experiments of 1-h in duration, but equilibrium behavior is not discussed beyond the Introduction section. Discussing equilibrium behavior in the Introduction section is irrelevant if this behavior is not addressed later in the manuscript. If the authors choose to leave discussion of equilibrium behavior in the manuscript, then how would the presence of semisolid SOA affect equilibrium partitioning of modeled species? Also, what photooxidation duration is required to reach equilibrium, and how does this duration compare with the amount of time ambient particles remain in the atmosphere?**

The equilibrium for very low volatile species is indeed not reached under typical chamber experiment durations. An additional graph will be added to show the distribution of a species after 20h, a maximum duration for chamber experiments. However we will keep the figure at equilibrium which is useful to understand the gas/particle/walls distribution of a given species with vapor pressure and time.

**2. Simulation behavior is tested against experiment results using  $\Delta HC$  values which only represent data taken at the beginning and end of experiments. This neglects many processes occurring during photooxidation. How does time-dependent modeled hydrocarbon decay match with experimental results? Are  $NO_x$  and  $O_3$  experimental data available, and if so, how do they match with model predictions?  $NO_x$  and  $O_3$  data would be especially important when addressing the fate of DHF.**

Experimental data available from Lim and Ziemann (2009), Matsunaga et al., (2009) and Matsunaga (2009) are the amounts of precursor, NO, methyl nitrite and organic seeds injected in the chamber for each experiment (see Table 1).

The quantity of reacted precursor,  $\Delta HC$ , is equal to:

$$\Delta HC = [HC]_{tot}^i - [HC]_{tot}^f$$

where  $[HC]_{tot}^i$  and  $[HC]_{tot}^f$  are the total mass concentration of hydrocarbon in the chamber at the beginning (i.e. the injected amount) and the end (i.e. the sum of the amount in the gas phase, particle phase and on the wall) of the experiment respectively. The initial (before turning the light on,  $[HC]_{gas}^{on}$ ) and final (after turning the lamps off,  $[HC]_{gas}^{off}$ ) concentrations of the precursor in the gas phase were measured, once the gas/aerosol/wall equilibrium has been achieved, to calculate  $\Delta HC$  as:

$$\Delta HC = \left(1 - \frac{[HC]_{gas}^{off}}{[HC]_{gas}^{on}}\right) [HC]_{tot}^i$$

The temporal evolutions of the concentration of precursor, NO<sub>x</sub> or O<sub>3</sub> during the experiment are not available.

The sentence (p23905 l3) about  $\Delta HC$  was misleading and will be removed in the revised version. The following section will be added in the revised paper (p23900 l14): "The quantity of reacted precursor,  $\Delta HC$ , is equal to:

$$\Delta HC = [HC]_{tot}^i - [HC]_{tot}^f$$

where  $[HC]_{tot}^i$  and  $[HC]_{tot}^f$  are the total mass concentration of hydrocarbon in the chamber at the beginning (i.e. the injected amount) and the end (i.e. the sum of the amount in the gas phase, particle phase and on the wall) of the experiment respectively.  $\Delta HC$  was calculated experimentally using the initial (before turning the light on,  $[HC]_{gas}^{on}$ ) and final (after turning the lamps off,  $[HC]_{gas}^{off}$ ) measured concentrations of the precursor in the gas phase once the gas/aerosol/wall equilibrium has been achieved as:

$$\Delta HC = \left(1 - \frac{[HC]_{gas}^{off}}{[HC]_{gas}^{on}}\right) [HC]_{tot}^i$$

The experimental  $\Delta HC$  are reported Figure 6 for all the experiments."

**3. Use of the GECKO-A modeling framework allows the authors to study wall uptake by species category and oxidation generation. The discussion of wall uptake by species uptake in Section 4.2 is very nice. It would also be informative to discuss the effect on SOA yield of wall uptake for different generation photooxidation products by comparing the simulated wall and no wall cases. From Figure 8, it appears that during dodecane photooxidation there is substantial wall loss of first-generation photooxidation products; whereas, for hexadecane, first-generation photooxidation products are less affected by wall loss. For which compound is there a greater difference between modeled wall and no wall cases?**

Wall loss uptake of a given species depends on its vapor pressure (see Fig. 2 and Fig. 8) but does not directly depend on the number of generation (see response to question 2.2 of the major comments of Referee #1).

#### Additional comments

**p. 23897, line 9. Are values for  $C_w/(Mw\gamma_w)$  chamber-dependent? How do the values of  $\tau_{gw}$  used in this manuscript compare with those calculated using the wall deposition rates proposed by Zhang et al. (ACP, 2015)?**

The  $C_w/(M_w\gamma_w)$  and  $k_{gw}$  values optimized by Matsunaga and Ziemann (2010) are expected to be chamber dependent. Zhang et al., (2015) measured values for  $k_{gw}$  about twice orders of magnitude lower than the values reported by Matsunaga and Ziemann (2010). Such a large difference in  $k_{gw}$  between the two chambers is however not expected and the reason for this difference remains unknown (Yeh and Ziemann (2015), McVay et al. (2015)).

***p. 2390, line 11. How did the mass of DOS seed aerosol compare with the effective chamber wall mass?***

The initial DOS mass ranged between 200-400  $\mu\text{g m}^{-3}$ . The effective chamber wall mass optimized for 2-ketones is two orders of magnitudes higher ( $C_w$  (2-ketones)=24  $\text{mg m}^{-3}$ ). The equilibrium constant for the gas/wall partitioning ( $k_{gw}/k_{wg}=[\text{SVOC}(w)]_{\text{eq}}/[\text{SVOC}(g)]_{\text{eq}}$ ) is thus 2 orders of magnitudes higher than the equilibrium constant for the gas/particle partitioning ( $k_{gp}/k_{pg}=[\text{SVOC}(p)]_{\text{eq}}/[\text{SVOC}(g)]_{\text{eq}}$ ). As discussed in the paper for 1 h timescale, species having vapor pressure in the  $10^{-8}$ - $10^{-5}$  atm range will predominantly partition to the wall (thermodynamic control) while species having a vapor pressure lower than  $10^{-8}$  atm will mostly partition to the particles (DOS or SOA).

***p. 23903, line 2. Why was a SOA density of 1 used?***

Lim and Ziemann (2009) and Matsunaga et al. (2009) measured an aerosol density of 1.06  $\text{g cm}^{-3}$  and 1.13  $\text{g cm}^{-3}$  for SOA formed from alkane and alkene oxidation respectively on DOS seed aerosols. An aerosol density of 1  $\text{g cm}^{-3}$  was therefore used for the simulations.

The references of Lim and Ziemann (2009) and Matsunaga et al. (2009) will be added in the revised version to justify the density choice of 1  $\text{g cm}^{-3}$  p23903 l3.

***p. 23904, line 26. In the experiments studied, only a fraction of the initial hydrocarbon was reacted during the experiments. In this case, does the fact that the parent alkanes/alkenes partition to the walls before the start of experiments affect the SOA yields (i.e., is this hydrocarbon coming off the walls during the experiment leading to an incorrect  $\Delta\text{HC}$  value)?***

The  $\Delta\text{HC}$  used in the calculation of both experimental and modelled yields corresponds to the amount of hydrocarbon which has reacted (see response to question 2 above).

***p. 2390, Section 4.1. Do experimental and modeled yields take into account particle wall loss, and if so, how is gas-particle partitioning to deposited particles treated?***

The experimental SOA yields published in Lim and Ziemann (2009), Matsunaga et al. (2009) and Matsunaga (2009) are corrected from particle wall loss considering a first order loss process and no interaction between the gaseous compounds and the wall deposited particles. The wall loss of particles has therefore not been included in the model. Simulated results were directly compared to the measured SOA yields corrected from particle wall loss.

A sentence will be added in the corrected version of the manuscript for clarification p23905 l21.

***p. 23922, Figure 2. The time evolution component of this figure is unclear from the figure and caption. The plots should be labeled with the simulation duration, or this should be stated in the figure caption.***

Labels were missing in fig 2 - thanks for pointing this out. The panels will be labeled with the simulation time in the revised version of the paper.

## Bibliography

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