Review of “Microphysics regimes due to haze-cloud interactions: cloud oscillation and cloud collapse” by Fan Yang et al.

This manuscript could be considered for publication after a major revision.

The authors conducted a series of large-eddy simulations (LESs) of a cloud in a convection chamber using a haze-capable Eulerian-based bin microphysics scheme to explore haze-cloud interactions over a wide range of aerosol injection rates. They observed three microphysics regimes as they increased the aerosol injection rate: slow microphysics regime, fast microphysics regime, and cloud oscillation. The cloud oscillation is a new phenomenon being reported for the first time in this study. To understand the physical mechanism, they conducted a detailed analysis by introducing a box model. They also found that cloud collapse can occur if the side wall humidity is low. By solving a haze-only box model analytically, they also found the existence of a haze-only solution when aerosol injection rate is high.

As the LES using Twomey-type activation reproduced the slow and fast microphysics regimes but could not capture the cloud oscillation, they concluded that the haze-cloud interaction is critical in polluted conditions, but we could still use Twomey-type activation parameterizations for less polluted conditions.

The manuscript is relatively well written. However, the analysis is not comprehensive enough, and this makes the author’s main conclusions not fully convincing. In particular, I see two major issues in this study:

1. They concluded that we could still use Twomey-type activation parameterizations for less polluted conditions. But, in my opinion, the conditions they tested are rather limited and the conclusion could be misleading.
2. I was confused that the author’s interpretation regarding the cloud oscillation is sometimes not consistent with the plots they presented. A more careful analysis on supersaturation, supersaturation fluctuation, activation rate, deactivation rate, and sedimentation rate is desired. I believe it should foster a deeper understanding about the cloud oscillation phenomenon.

Please also see my more detailed comments provided below.

I believe the quality of the study will be significantly enhanced if these points are addressed. I look forward to reading the revision of this manuscript.

Major Comments

1) **[request] P. 10 ll. 207-208 “The apparent transition between slow and fast regimes as shown in Fig. 1 provides an opportunity to estimate τ_m”**
   From the discussion on p. 16, it seems the authors are thinking that the transition point is $n_{in} = 0.1\,\mathrm{cm}^{-3}\mathrm{s}^{-1}$ and $\tau_m \approx 70\,\mathrm{s}$, but it is indicated only implicitly on p. 10. Please clarify this point.

Well, it should be possible to calculate the turbulent mixing time from the flow field directly, e.g., by calculating the integral time scale of the turbulence.

2) **[request] Figure 4**

Please also show the time evolution of total particle number $(N_d + N_{a/h})$, activation rate, deactivation rate, sedimentation rate, s, and $\sigma(s)$. This should provide more insight into understanding the mechanism.

3) **[request] P. 11 l. 220 "Note that Na increases with time for nin ≥ 10 cm−3s−1."**

Do you mean $n_{in} \geq 40\,\mathrm{cm}^{-3}\mathrm{s}^{-1}$? Please clarify.

4) **[suggestion] P. 11 l. 224 "The oscillation period increases as nin increases, ..."**

It should be also informative to point out that the oscillation amplitude increases as n_{in} increases.

5) **[request] Figure 5i**

For a better comparison, please use the same color bar as in Figure 6.

6) **[question] P. 11 ll. 234–235 "The sharp increase in Nd (Fig. 5b) corresponds to a larger activation rate (Fig. 5c) due to the enhanced supersaturation (Fig. 5i), …"**

From Figs. 5c and 5f, it looks like the mean R_{deact} is always larger than mean R_{act}, though on average $R_{act} - R_{deact} = n_{in}$ has to be satisfied. Why is it?

7) **[question] P. 11 l. 235 "..., while the decrease in Nd corresponds to a larger deactivation rate and a smaller supersaturation."**

If we compare Figs. 5f and 5i, the deactivation rate is larger even when supersaturation is larger, which is counterintuitive. Is this due to supersaturation fluctuation?
On a related note, is removal by sedimentation much smaller than R_{deact} and R_{act}?

8) **[question] P. 11 l. 240 "When s > scrit (scrit ≈ 8‰in this study), ..."**

From Fig. 5i, it looks like $s < s_{crit}$ always holds. Why is it?

9) **[question] P. 12 ll. 243–245 "Shortly thereafter, Nd decreases because droplet activation is suppressed when s < scrit, and meanwhile, droplets are lost due to sedimentation and deactivation."**

Which is dominant, sedimentation or deactivation? From Figs. 5i and 6, mean supersaturation s is always positive. Then, does deactivation occur due to supersaturation fluctuation? How big is $\sigma(s)$?

10) **[question] P. 12 l. 249 "It is interesting to see that the oscillation evolves with time clockwise in qh −Nh diagram (Fig. 6c) …"**

If we compare the color and size of the circles in Figs. 6c and 6d, it looks like the sequence in Fig. 6c is in the opposite order, i.e., anticlockwise.

11) **[comment] Pp. 12–13 ll .236–250**

From the observation described in this section, it is not clear why cloud oscillation does not occur when using the CL_CCN scheme.

12) **[suggestion] Eqs. 1, 2, and 4**

The dq_l/dt here represents the change of q_l through diffusional growth, but droplet removal by sedimentation also changes q_l. To avoid confusion, the authors should clarify this point, e.g., by using a subscript: $dq_l/dt|_{diff}$.

13) **[question] Eq. 3**

If I understand correctly, the supersaturation fluctuation is not considered in this box model, right? Is it not important for the phenomenon?

14) **[question] P. 16 l. 299 "T0 and qv0 are set to be 290 K and 13.9 g kg−1, …"**

What is the corresponding supersaturation s_0?

15) **[request] P.16 l.302 "... the estimated τm for Da = 1 based on LES results …"**

Please clarify that the estimated τ_m was $70\,\mathrm{s}$.

16) **[request] Figure 8**

Please also show the time evolution of total particle number $(N_d + N_h)$, activation rate, deactivation rate, sedimentation rate, and s.

17) **[question] P.16 l.316 "In contrast, simulations using the CLCCN scheme do not show oscillations …"**

What will happen if we use Twomey activation for this box model? Fig. 4 revealed that LES with CL_CCN does not show oscillation, but it seems to me nothing prevents the Twomey box model from exhibiting oscillation?

18) **[question] Sec. 3.2.2 "Cloud oscillation in a box model"**

Additional questions about this section:

- Because supersaturation fluctuation is not considered in this model, $s > 0$ always holds, and deactivation of droplets does not occur in this model. Is this correct?
- Instead, the droplets are removed from the system only by sedimentation in the box model. Is this correct?
- The oscillation amplitude of N_d in Fig. 8 is smaller than that in Fig. 4. Is this because of the absence of deactivation in the box model?
- Because supersaturation fluctuation is not considered in this model, almost all haze particles should be activated when $s > s_{crit}$. This is the reason why N_h decreases to almost 0 in Fig. 8 (though this is not happening in Fig. 4). Is this correct?

19) **[question] P.18 l.344 "..., the oscillation frequency approaches zero)?**

Why do you think the frequency approaches zero when switching to the haze-only regime?

In dynamical systems theory, there are various types of bifurcations that are responsible for the onset of oscillation. If the frequency approaches zero, it suggests it is an "infinite period bifurcation". On the other hand, if the oscillatory solution arises when a fixed point (haze-only solution) is destabilized, it is a Hopf bifurcation. Then, the frequency is finite at the bifurcation point. See, e.g., Strogatz (2014).

20) **[suggestion] Eqs. 7–11**

Again, it should be clarified that only the contribution via diffusional growth is considered, e.g., by using a subscript: $dq_l/dt|_{diff}$.

In particular, Eq. 11 is confusing if dq_l/dt is used on the l.h.s.; if we take it literally, it indicates q_l grows exponentially! But, of course, the correct meaning is $dq_l/dt|_{diff} = q_l/\tau_{sed}$. I would suggest the expression $n_{in} = N_h/\tau_{sed}$ as an equivalent but more intuitive alternative.

21) **[question] Eq. 10**

I think the use of this formula is not appropriate for this analysis, because $r_{eq} \to \infty$ as $RH \to 1$. How about simply assuming $r_{eq} = \text{const.}$?

22) **[question] Figure 10**

How are the left ends of the two lines determined? Are they corresponding to $RH = 1$? If so, Eq. 10 should not be used because it is not accurate when RH approaches 1.

23) **[request] P.21 l.374 "..., so that s < scrit all the time."**

Because of the use of Eq. 10, the analysis is valid only for $s < 0 (< s_{crit})$. Please clarify this point.

24) **[comment] Sec. 3.2.4 "Haze-only regime"**

The existence of the haze-only solution is presented in this section, but it does not guarantee it is stable.

25) **[request] P.23 l.433 "deactivation (s < scrit)"**

Do you mean $s < 0$?

In both CL_haze and CL_CCN, deactivation occurs only when the supersaturation is locally smaller than zero. Hence, if there is no fluctuation, $0 < s < s_{crit}$ (s represents mean supersaturation) does not induce any deactivation. (More precisely speaking, if all the activated droplets are large enough.) If the supersaturation fluctuation is large, deactivation can occur locally even when $s_{crit} < s$.

26) **[request] P.24 ll.443–444 "Haze-cloud oscillation is more likely to occur under conditions of weak supersaturation forcing, …"**

Why do you think so? Please elaborate.

27) **[comment] P.24 l.455 "Our results suggest that haze-cloud interactions are very important especially in polluted conditions."**

The authors suggest that we could still use Twomey-type activation parameterizations for less polluted conditions, but I am not fully convinced. NaCl aerosol particles with a dry radius of 62.5 nm are considered in this study, but they are relatively small. I think the haze-cloud interaction should be more important for larger aerosol particles because the equilibrium wet radius gets larger and the activation/deactivation time scale gets longer. In addition, aerosols considered are monodisperse in this study, but I also think that haze-cloud interaction is more important for polydisperse aerosols. Please see, e.g., Fig.5 of Richter et al. (2021).
These limitations of this study should be discussed and emphasized more.

Minor Comments

28) **[request] P.2 ll.54–55 “Shaw et al. (2023)”**
Because polydisperse aerosol injection is discussed in the previous sentence, please clarify that monodisperse aerosol injection was assumed in Shaw et al. (2023).

29) **[request] P.7 Fig. 1**
Please also show the standard deviation of supersaturation $\sigma(s)$.
What is the definition of droplet mean radius r_d and droplet number concentration N_d for the CL_Haze scheme? Are the haze droplets included in the population? Please clarify.

30) **[comment] P.9 Table 2 and elsewhere “Na/Nh”**
This looks like “Na divided by Nh” and I believe it is confusing to the readers. How about simply writing “Na, Nh” or “Na or Nh”?

31) **[comment] P.9 Table 2**
Please consider including units in the headers to enhance readability.

32) **[question] P.10 ll.198–199 “Note that the net activation rate (Ract−Rdeact) is close to nin for each case suggesting that the cloud reaches a quasi-steady state.”**
In a quasi-steady state, the removal rate by sedimentation should be equal to the net activation rate and injection rate. Did you confirm this? To put it another way, how did you confirm that one hour is sufficient to reach a quasi-steady state?

33) **[comment] P.15 l.269 “Each particle represents numerous real particles per unit volume. We refer to this as multiplicity, …”**
Note that multiplicity is defined differently in Shima et al. (2009).

34) **[request] P.23 l.426 “But they do not capture the distribution properties …”**
Please clarify that “they” represents “analytical estimates”.

Typo

35) **P.3 l.59 response -> respond**

36) **Eq. 4**

G is not needed?

37) Eq. 5

If δn_i is representing the decreased amount of multiplicity, Eq. 5 has to be

$$\delta n_i = n_i \left(1 - \exp\left[-\frac{\delta t}{\tau_{sed}(r_i)}\right]\right) \approx n_i \frac{\delta t}{\tau_{sed}(r_i)}.$$

38) P.21 l.385 "following by Thomas et al." -> "following Thomas et al."

39) P.22 Eq. 12

This must be $\delta N_h = N_h \Delta t / t_{ql}$.

References

Richter, D.H., MacMillan, T. & Wainwright, C. A Lagrangian Cloud Model for the Study of Marine Fog. Boundary-Layer Meteorol 181, 523–542 (2021). https://doi.org/10.1007/s10546-020-00595-w

S. Shima, K. Kusano, A. Kawano, T. Sugiyama, and S. Kawahara, Q. J. R. Meteorol. Soc. 135, pp.1307-1320 (2009). DOI: http://dx.doi.org/10.1002/qj.441

Strogatz, S.: Nonlinear Dynamics and Chaos: With Applications to Physics, Biology, Chemistry, and Engineering, 2nd Edn., 513 pp., Westview, 2014.