Respond to Reviewer 1.

The authors presented relationship among particle collision radial distribution function (RDF) and mean relative velocity (MRV). They compared the results from Direct Numerical simulation (DNS) and theory proposed by them. The results show that collision is affects the RDF and MRV and imposes strong coupling between them.

The work done in the manuscript is novel. However, authors have to provide more details of the numerical simulations before it can be considered for publication in the journal. The major comments are provided below.

>> We thank the reviewer for appreciating the novelty of our work and for his/her comments and suggestions, we find these comments rather helpful in improving the quality of the manuscript.

Major Comments

1. More information about the DNS has to be provided in the introduction section. In particular, the reference should be given for DNS done by others using similar condition of turbulence and numerical schemes. See the references Kumar et al. (2012,2014) and Thomas et al. (2020).

>> We have added more information about the DNS and more citations to works on settings similar to our work. We should also mention that the original manuscript had already cited a number of early papers involving DNS under similar setting such as Chun et al. (2005), Sundaram & Collins (1997), Salazar et al. (2008), Saw et al. (2012b, 2014), Voßkuhle et al. (2013), Wang et al. (2008), Bec et al. (2007). We have now also cited some of the references mentioned above by the reviewer where suitable.

2. Section 2:
   (i) Some references are required for the DNS set up.
   (ii) More details about particle characteristics such as initial size distribution. The number density, temperature and humidity used for the simulation.

>> (i) More relevant citations are now included. We should also point out that a significant number of citation was already given for the set-up of our DNS, for instance: we have cite Rogallo (1981), Pope (2000) and Mortensen (2016) for the standard pseudospectra method and the general, standard, DNS of this type; we have cited Ireland (2013) for the novel time-stepping method used for advancing the particles; We have cite Rogallo (1981) for the standard 2/3-dealiasing rule employed. Now, we have also added citations for the particle advection equation and the collision-processing procedures among other things. (ii)These details are now described in the manuscript (in particular in Section 2). We have also added a dedicated section in the Supplement with further details on the DNS.

3. Section 4 (or 2): Simulation details:

   (i) Is there and phase change considered for the particles?

>> No, this is not considered. We have now made this point clear in Section 2 and discuss the limitations due to this neglect.

4. The article starts with “considering turbulent clouds containing small heavy particles”, however, gravitational acceleration is not considered in particles velocity equation (Page- 3, line 72, equation number is not given). Justification for this, given by the authors, is not strong enough. The entire study was done to investigate the effect of particle collision on particle relative motion. Hence in this scenario, effect of gravity should be considered in droplets velocity equation.

>> As we have now explain in the introductory section of the manuscript: “the focus of our work is on the fundamental relationship between collision, RDF and MRV, and to highlight differences from the case with non-colliding particles (Chun et al., 2005). To that end, we have designed the DNS to have an idealized setup similar what was done in (Chun et al., 2005), which would allow us to identify without doubt the effects of particle collision-coagulation.” Also: “This approach certainly limits the direct applicability of our results to real problems in atmospheric clouds, these limitations will be discussed in Sec. 4.5.”

Another goal of this study is to probe the accuracy of the drift-diffusion theory. In our view, considering the focus and goals of our present work, it is beneficial to keep the DNS setting idealized (and in the regime relevant for the theory) for the sake of clarity when interpreting results. Thus, we choose at this stage to not include various complexities that, although important for the ultimate application to real atmospheric clouds, would distracts the reader from the simple fundamental message derivable from our work, i.e. 1) Collision will fundamentally change the form and relations of RDF and MRV; 2) RDF and MRV are strongly coupled (in general); 3) RDF is not zero at r=d despite appearance; 4) the drift-diffusion theory is, in principle, extendable to scenario beyond non-colliding particles. 5) Simple phenomenology could reproduce the main features of MRV (at least in the regime considered). 6) The differential version of the theory should not have been accurate in some commonly encountered (studied) scenario, thus, pressing theoretical works is needed to understand how it was able to produce reasonable results.

We have now clarified these points in the manuscript (in Sec. 1 and Sec. 2).

However, we appreciate the point of view of the reviewer on the importance of gravitational effects. To that end, we have dedicated a section in the later part of the manuscript to briefly survey the effects of gravitational body force on the main qualitative findings/conclusion of our present work. A detailed quantitative analysis is beyond the scope of this manuscript and should be a subject of future studies.

To address the general concern of the reviewer on the relevance to realistic clouds, we have added a section that clarify the limitations of the present study. In short, our results are not intended to be directly applicable to real applications but are intended as a tool to explore several fundamental questions related to the problem. It is hoped that these fundamental understandings will inform future, more realistic, accounts of the complex problem.

5. Definition of τ_p (page 3, line 73) is not mentioned in the article.

>> We thank the reviewer for pointing out this error. A definition is now given.

6. Detail information about particles number concentration at the time of injection,

surrounding temperature and relative humidity of particles is also missing in the article.

>> Question regarding temperature and humidity is already addressed in point 2 and 4 above. The initial number of particles is now reported in Sec. 2. Also, more technical details about the simulation are now given in a dedicated section in the Supplement.

7. Justification behind the consideration of certain values of Stokes number (St = 0.22, 0.54, 0.054, 0.001) (page-5 line113) is missing in the article. While presenting an accuracy of a theory, a large number of simulations are needed at all possible complex scenarios. I found it missing in authors' present work.

>> A motivation for choice of St is now given in the second paragraph of Sec. 4.

>> As the theory is derived based on the assumption of St<<1. We are exploring the Stokes number values close to this regime only, with the exception of St=0.54, which is included to show that the general trend we observed (on the DNS produced RDF and MRV) extends beyond the St<<1 regime. An extension of the theory to finite Stokes number is definitely of great interest (even without including gravity and other inter-particle forces) and has been pursued for decades now, it is hoped that our results could shed light on this endeavor.

8. As per my understanding, authors considered particles of size 94.9 micrometre (0.000949 dm) or bigger. If so then mention the DNS domain size and each grid box length. Also, at what stage, the reverse effect of particle collision was considered is missing in this article.

>> Domain size (of the simulation cube) was already given in the original manuscript in Table 1 (i.e. 2*pi dm on each side). The number of grid points (i.e. 256^3) was also already given in Sec. 2 (just after Eq. 1). The reverse effect is always present, as a consequence of collision-coagulation, we do not directly control or impose this.

9. The statement made by authors in section 4.3 (page 10, line 211) is not justified. The numerical simulation is done at a very basic scenario, which may lead to an insignificant impact of Stokes numbers on W_r , calculated using DNS.

>> We understand that here the reviewer means that since our DNS setup is fairly idealized, we may not be able to capture any extra impact (on the MRV) caused by forces omitted in the DNS (e.g. gravity), thus the said statement would not hold for the general, real, system. We basically agree with that point of view, and thus we have added a footnote to clarify this limitation.

10. In my opinion, authors need to work more to justify their work and give a more clear explanation about the results.

At this stage, the article is not ready for the publication and requires major changes.

>> We thank the reviewer for these comments, and we have carefully edited the text, added sections and scientific contents to address these comments (details above). In our view, the updated manuscript should satisfy the required standards. We reiterate that this study focuses on the fundamental aspects of the problem (details in respond to point 4 above and the manuscript) and it is in the interest of scientific clarity that the DNS is done in a relatively

idealized setting. Also, it is our opinion that the fundamental findings our manuscript conveys merit rapid publication so that the community could benefit from them, before a more detailed quantitative investigation involving more complexities could be done.

References:

Kumar, B., Schumacher, J., and Shaw, R. A.: Lagrangian Mixing Dynamics at the Cloudy-Clear Air Interface, J. Atmos. Sci., 71, 2564–2580, https://doi.org/10.1175/JAS-D-13-0294.1, 2014.

Kumar, B., Janetzko, F., Schumacher, J., and Shaw, R. A.: Extreme responses of a coupled scalar–particle system during turbulent mixing, New J. Phys., 14, 115020, https://doi.org/10.1088/1367-2630/14/11/115020, 2012.

Thomas, L., Grabowski, W. W., and Kumar, B.: Diffusional growth of cloud droplets in homogeneous isotropic turbulence: DNS, scaled-up DNS, and stochastic model, Atmos. Chem. Phys., 20, 9087–9100, https://doi.org/10.5194/acp-20-9087-2020, 2020.