On Clouds and Turbulence: Causality and Theory of Particle Collision, Relative Motion and Clustering.

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Supplementary Material

S1 Estimation of Leading Order Terms in the Drift Flux, e.g $A_{ik}^{(1)}$

Using the DNS data, we estimate e.g. the value of

$$\int\limits_{-\infty}^{t} A_{ik}^{(1)} dt' \equiv \int\limits_{-\infty}^{t} \tau_\eta \left\langle \Gamma_{ik}(t)\Gamma_{lm}(t')\Gamma_{ml}(t')\right\rangle dt'.$$

Note: the averaging is done over fluid particles (the theory assumed $St \ll 1$ limit, such that all velocity statistics are tied to the fluid's), the integrand is non-vanishing only for t' in the vicinity of $t-\tau_\eta$ to t (where the turbulent velocity gradient Γ_{ij} retains correlation), thus this quantity may be approximated as: ${\tau_\eta}^2 \left\langle \Gamma_{ik}(t)\Gamma_{lm}(t)\Gamma_{ml}(t)\right\rangle$. As shown in Chun et al. (2005), $\left\langle \Gamma_{ik}(t)\Gamma_{lm}(t)\Gamma_{ml}(t)\right\rangle$ is by definition zero in fully developed turbulence due to the fact that the small-scale statistics of turbulent flows are almost isotropic Kolmogorov (1941). However, the coagulation constraint dictates that at $r=d$, such averages must be taken with the condition that only fluid-particle pairs with negative radial velocity ($w_r < 0$) are taken into account (that the inertial particles' motion being tied to the fluid's does not imply that inertial pairs sample the fluid particle pairs's motion uniformly). Under this condition, the DNS data gives ${\tau_\eta}^2 \left\langle \Gamma_{ik}(t)\Gamma_{lm}(t)\Gamma_{ml}(t)\right\rangle \approx -0.171\times10^{-3}/d_*$, ($d_* = 9.49\times10^{-4}$); here, it is of value to point out that without such constraint or condition, the result for this quantity from the DNS is two orders of magnitude smaller. Similarly, we found $\int_{-\infty}^{t} A_{ki}^{(2)} dt' \approx \tau_\eta^3 \left\langle \Gamma_{ij}(t)\Gamma_{jk}(t)\Gamma_{lm}(t)\Gamma_{ml}(t)\right\rangle \approx 2.32\times10^{-3}/d_*$; for this quantity, the DNS gives roughly the same values with or without the constraint.

S2 Full Definition of the Function $f_I(R_0,\mu,t_f)$ in the Model for Non-local Diffusive Flux.

Derived in Chun et al. (2005), summarized here (with typo corrected), the diffusive action of the turbulence on the particle-pairs is assumed to consist of a random sequence of uniaxial extensional or compressional flows defined, and:

$$f_I(R_0,\mu,t_f) \equiv f_+ I_+(R_0,\mu,t_f) + f_- I_-(R_0,\mu,t_f),$$

where $R_0 \equiv r_0/r$, r_0 is the initial separation distance of a particle pair before a straining event, r is the independent variable of the equation for $g(r)$; f_+ and $f_- \equiv 1 - f_+$ are the fractions of those flows that are extensional and compressional, respectively. Chun et al. (2005) calibrated with help from DNS that, and we adopt, $f_+ = 0.188$. $I_\pm$ is an indicator function such that it takes the value $+1$ (-1) when a secondary particle leaves (enters) a sphere of radius r centered on the primary particle, and otherwise zero. μ is the cosine of the angle between the axis of symmetry of the straining flow event and the separation vector of the particle pair, t_f is the lifetime if the event. To obtain a strain rate correlation function that decays exponentially with a characteristic time scale τ_S, Chun et al. (2005) set the probability density function for t_f to be:

$$F(t_f) = \frac{f_s t_f}{{\tau_S}^2} \exp(-t_f/\tau_S)\,.$$

The indicator function is used to count the net loss of particles from within the sphere over the duration of an (extensional or compressional) event and can be expressed as:

$$I_\pm(R_0,\mu,t_f) = H(1-R_0)H(R_{f\pm}-1) - H(R_0-1)H(1-R_{f\pm}),$$

where $H(x)$ is the heaviside function (zero for $x<0$, unity for $x \geq 0$), $R_{f\pm}$ is the non-dimensional final position of a particle pair with an initial position of R_0 and can be written as:

$$R_{f+} = R_0 \left[\mu^2 {\theta_t}^2 + \frac{(1-\mu^2)}{\theta_t}\right]^{1/2},$$

$$R_{f-} = R_0 \left[\frac{\mu^2}{{\theta_t}^2} + (1-\mu^2)\theta_t\right]^{1/2},$$

for uniaxial extension and compression respectively, where:

$$\theta_t \equiv \exp\left(\frac{t_f}{\tau_\eta \sqrt{3 f_s}}\right).$$

S3 Derivation and Role of c_{st} .

In this work, we deviate from the CK theory Chun et al. (2005) by introducing an extra factor c_{st} (positive, of order unity or less) in the model of non-local diffusion:

$$q_r^D = c_{st}\, r \int d\Omega \int_0^\infty dt_f F(t_f) \int_{d/r}^\infty dR_0\, {R_0}^2 \langle P\rangle(r R_0)\, f_I(R_0,\mu,t_f)\,. \tag{S1}$$

To determine what c_{st} is (or should be), we begin from an important finding in Chun et al. (2005) that if $\langle P\rangle$ is power-law of r, i.e. $\langle P\rangle = C r^{-c_1}$, then the non-local diffusion q_r^D can be cast into a differential form (which is usually only true for local diffusion):

$$q_r^D = -B_{nl}\, {\tau_\eta}^{-1} r^2 \frac{\partial \langle P\rangle}{\partial r}, \tag{S2}$$

where:

$$B_{nl} = \tau_\eta \int d\Omega \int_0^\infty dt_f \, F(t_f) \int_{d/r}^\infty dR_0 \, {R_0}^{2-c_1} f_I(R_0, \mu, t_f) \,. \tag{S3}$$

This, together with: $q_i^d = -A_{ck} \, {\tau_\eta}^{-1} \, r \, \langle P \rangle$, eventually leads to the first order equation differential equation for the RDF $g(r) \equiv V \langle P \rangle$, that has (only) power-law solutions: $g(r) = VCr^{-c_1}$. This result (i.e. $g(r)$ or equivalently $\langle P \rangle(r)$ are power-laws) has seen compelling validations from both experiments (e.g. Saw et al. (2012b); Lu et al. (2010); Yavuz et al. (2018)) and DNS (e.g. Chun et al. (2005); Bec et al. (2007); Saw et al. (2012a)). We now begin from this experimentally validated result and work backward to derive an expression for c_{st}. We plug the power-law form for $\langle P \rangle$ into (S2):

$$\begin{aligned}
q_r^D &= -B_{nl} \, {\tau_\eta}^{-1} \, r^2 \, \frac{\partial (Cr^{-c_1})}{\partial r} \\
&= -B_{nl} \, {\tau_\eta}^{-1} \, r^2 C(-c_1) r^{-c_1 - 1} \\
&= B_{nl} \, {\tau_\eta}^{-1} \, r c_1 \, C \, r^{-c_1} \\
&= {\tau_\eta}^{-1} \, r c_1 \, C \, r^{-c_1} \tau_\eta \int d\Omega \int_0^\infty dt_f \, F(t_f) \int_{d/r}^\infty dR_0 \, {R_0}^{2-c_1} f_I(R_0, \mu, t_f) \\
&= r c_1 \int d\Omega \int_0^\infty dt_f \, F(t_f) \int_{d/r}^\infty dR_0 \, {R_0}^2 \, C(rR_0)^{-c_1} f_I(R_0, \mu, t_f) \\
&= c_1 \, r \int d\Omega \int_0^\infty dt_f \, F(t_f) \int_{d/r}^\infty dR_0 \, {R_0}^2 \, \langle P \rangle(rR_0) \, f_I(R_0, \mu, t_f) \,.
\end{aligned}$$

Comparing with (S1), we have:

$$c_{st} = |-c_1| \equiv |c_1| \,,$$

which is found in experiments (and theories) to be of order 0 to 1 and a function of particle Stokes number St; in words, this means c_{st} is given by the modulus of the power-law exponent of the RDF that would arise in the collision-less case; in the case with collision and sufficiently small particle ($d/\eta \lesssim 1$), such as in this study, c_{st} equals the modulus of the power-law exponent of the RDF the range of $d \ll r \ll 20\eta$ (note: power-laws RDF are empirically observed for $r \ll 20\eta$ Saw et al. (2008, 2012a)). Note: we have chosen to define c_{st} using the 'modulus' (instead of the 'negative' of the power-law exponent) since it guarantees that q_r^D is negative (positive) when $g(r)$ is an increasing (decreasing) function of r, so that we are consistent with the fact that q_r^D is a diffusion flux. We note that both the CK theory and the current modified version assume $St \ll 1$.

Chun et al. Chun et al. (2005) went further to provide a solution for c_1 (for collision-less particles, in the $St \ll 1$ limit):

$$c_1 = \frac{A_{ck}}{B_{nl}} \equiv \frac{A(St, r \gg d) \, \tau_\eta}{B_{nl}} \,, \tag{S4}$$

where have made explicit the dependence of A in this work and that it has a different definition (and dimension) compared to its counterpart in Chun et al. (2005). In the current context, c_1 maybe obtained via (S4) or alternatively directly from the power-law exponent of $g(r)$ in the range $d \ll r \ll 20\eta$ as discussed above. Using values of the relevant parameters in our DNS, we found $\frac{A\,\tau_\eta}{B_{nl}} \approx \frac{2.4St^2\times.0925}{.0397} = 5.6St^2$, which is 15% smaller than the one found in Chun et al. (2005), i.e. $\frac{A_{ck}}{B_{nl}} \approx \frac{.61St^2}{.0926} = 6.6St^2$. However, we have observed in our DNS that the direct method (by fitting power-laws to the RDFs in the suitable r-range) gives c_1 which is 3.2 (1.9) times larger than the one obtained using (S4) for the case of $St = 0.054$ (0.11).

A plausible interpretation of the discrepancy described just above is that there may be another missing dimensionless factor (of order unity, possibly weakly dependent on Reynolds-number) in the correct definition of q_r^D . This could be a good subject for a detailed future study, here we close by noting that, by inspection, we found that if we further include a factor of $1/3.2$ in the definition of q_r^D, then the predicted $\langle W_r \rangle$ using the integro-differential version of the theory (cf. main text, case $St = 0.054$) is detectably closer to the DNS result in the $r \sim d$ regime, but the agreement is strikingly better in the $r \gg d$ limit (the latter should not come as a surprise as this is the regime of power-law RDFs and the factor-3.2 is exactly designed to reproduce the correct c_1); for the differential version of the theory, the improvement is decisively strong for all r.

S4 Relation Between $g(r)$ and $\langle P \rangle$.

In the main text, we state that $g(r) \equiv V\langle P \rangle$, where V is the spatial volume of the full domain of the problem i.e. $(2\pi)^3$ in the DNS. Justification: let $g(\boldsymbol{r})$ be the ratio of probability of finding a second particle at $\boldsymbol{r}$ from a particle, to the probability of such finding in a perfectly random distributed particle population, thus: $g(\boldsymbol{r}) \equiv \frac{\langle P \rangle \delta x \delta y \delta z}{(\delta x \delta y \delta z)/V} \equiv \langle P \rangle V$. Further, since system is isotropic, $g(\boldsymbol{r}) \equiv g(r)$.

S5 Lowest Order Phenomenological Model for Distribution of Particle Approach Angles $P(\theta)$.

We imagine the particles are small i.e. $d \ll \eta$ and $St \ll 1$. The latter implies their trajectories are almost like fluid particles', while the former implies that, viewed at the scale of interest $r \sim d$, their trajectories are almost rectilinear (since the radii of curvature are proportional to η). Thus in the reference frame of a primary particles, no secondary particle could have a trajectory, being straight-line, that has a history of collision with the volume of the primary (otherwise coagulation would have occurred and the secondary particle in question would cease to exist). In trigonometric terms, let θ be the angle between the secondary particle's velocity and its vector position in the rest frame of the primary particle, then we must have: $\sin^{-1}(d/r) \leq \theta \leq \pi$, with the convention that $\sin^{-1}(x) \in [-\frac{\pi}{2}, \frac{\pi}{2}]$.

From the above, we could then compute the MRV, $\langle w_r \rangle_*$, as a sum of the positive (i.e. $w_r > 0$) and negative branches (with proper statistical weights $p_\pm$ to account for possible skewness of the probability distribution of velocity):

$$\langle W_r \rangle \equiv \langle w_r \rangle_* = p_- \langle w_r \,|w_r < 0 \rangle_* + p_+ \langle w_r \,|w_r \geq 0 \rangle_* \,.$$

The negative branch $p_- \langle w_r \,|w_r < 0 \rangle_*$ is unaffected by collision-coagulation and we thus express it as a simple linear function of r that follows from the K41-phenomenology (Kolmogorov, 1941), i.e. $-p_- \,\xi_- \,r$, where $\xi_\pm \sim \sqrt{\varepsilon/(15\nu)}$, ε is the (kinetic) energy dissipation rate of the flow. For the positive branch, we further assume that the joint probability density function (PDF) of $|w_r|$ and approach-angle θ is separable (note: $w_r \equiv |w_r|\cos(\theta)$), hence:

$$\begin{aligned}
& p_+ \langle w_r \,| w_r \geq 0 \rangle_* \\
&= \int_0^\infty d|w_r| \int_{\theta_m}^{\frac{\pi}{2}} d\theta \, P(|w_r|, \theta)\, |w_r| \cos(\theta) \\
&= \int_0^\infty d|w_r| \, P_w(|w_r|)\, |w_r| \int_{\theta_m}^{\frac{\pi}{2}} d\theta \, P_\theta(\theta) \cos(\theta) \\
&= p_+ \int_0^\infty d|w_r| \, P_w(|w_r|)\, |w_r| \int_{\theta_m}^{\frac{\pi}{2}} d\theta \, P_\theta^+(\theta) \cos(\theta) \,,
\end{aligned}$$

where all the P's are PDFs, note that $p_+ \equiv \int_0^{\frac{\pi}{2}} P_\theta \, d\theta$, $\int_0^{\frac{\pi}{2}} P_\theta^+ d\theta \equiv \int_0^{\frac{\pi}{2}} (P_\theta / p_+) \, d\theta = 1$ and $\int_0^\pi P_\theta \, d\theta = 1$, also note that $P_\theta^+ \equiv P_\theta(\theta \,|\, w_r \geq 0)$; more importantly $\theta_m = \sin^{-1}(d/r)$ as previously explained. Further:

$$\begin{aligned}
& p_+ \langle w_r \,| w_r \geq 0 \rangle_* \\
&= p_+ \int_0^\infty d|w_r| \, P_w(|w_r|)\, |w_r| \int_{\theta_m}^{\frac{\pi}{2}} d\theta \, P_\theta^+(\theta) \cos(\theta) \\
&= p_+ \int_0^\infty d|w_r| \, P_w(|w_r|)\, |w_r| \left[\int_0^{\frac{\pi}{2}} d\theta \, P_\theta^+(\theta) \cos(\theta) + \int_{\theta_m}^{0} d\theta \, P_\theta^+(\theta) \cos(\theta) \right] \\
&= p_+ \int_0^\infty d|w_r| \, P_w(|w_r|)\, |w_r| \int_0^{\frac{\pi}{2}} d\theta \, P_\theta^+(\theta) \cos(\theta) \left[1 + \frac{\int_{\theta_m}^0 d\theta \, P_\theta^+(\theta) \cos(\theta)}{\int_0^{\frac{\pi}{2}} d\theta \, P_\theta^+(\theta) \cos(\theta)} \right] \\
&= p_+ \xi_+ r \left[1 + \frac{\int_{\theta_m}^0 d\theta \, P_\theta^+(\theta) \cos(\theta)}{\int_0^{\frac{\pi}{2}} d\theta \, P_\theta^+(\theta) \cos(\theta)} \right] ,
\end{aligned}$$

where in the last line, we have replaced the first two integrals, combined, with the Kolmogorov (1941) estimate, where $\xi_\pm \sim \sqrt{\varepsilon/(15\nu)}$.

S6 Prediction of the Peak Location of the RDF Using the Differential Form of the Drift-Diffusion Equation.

$$-\tau_\eta^{-1} B_{nl} \, r^4 \frac{\partial g}{\partial r} + \; g(r) \left[r^2 \langle W_r \rangle - A r^3 \right] = -R_c^* \,, \tag{S5}$$

A finite R_c^* inhibit us from locating the peak of the RDF using (S5) à la Lu et al. (2010) i.e. without knowing $g(r)$, since $g(r)$ could no longer be factored out when $\frac{\partial g}{\partial r} = 0$. However, we argue that (S5) could still give a reasonably accurate account of the peak location. For the case of $St = 0.05$, at $r = 3d$ (the approximate peak location), we found the DNS data gives $-\tau_\eta B_{nl} \, r^4 \frac{\partial g}{\partial r}\Big|_{\approx 0} + g(r) \left[r^2 \langle W_r \rangle - A r^3 \right] \approx -1.05 \times 10^{-9}$ and $-R_c^* \approx -1.01 \times 10^{-9}$

S7 General Analytical Solution for the Differential Form of the Drift-Diffusion Equation.

The general solution for the first-order non-homogenous ordinary differential equation (see e.g. Arfken and Weber (1999)), with $\langle w_r \rangle_*$ given by the model in the main text, is:

$$g(r) = \frac{1}{\beta(r)} \left[\int \beta(r) q(r) dr + C \right], \tag{S6}$$

with $q(r) = R_c^* / (\tau_\eta B_{nl} r^4)$; $\beta(r) = \exp\left[\int p(r) dr\right]$ and $p(r) = [\, Ar - \langle w_r \rangle_* \,] / (\tau_\eta B_{nl} r^2)$. For the current model described in the main text, the integral in (S6) could not be expressed in terms of simpler canonical functions. Hence, for specific applications, we currently anticipate that some sort of power-law expansion or asymptotic reduction (if not numerical integration) would be needed to produce problem specific analytical approximations.

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