

Scale-dependent droplet clustering in turbulent clouds

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(Received 5 December 2000 and in revised form 27 January 2001)

The current understanding of fundamental processes in atmospheric clouds, such as nucleation, droplet growth, and the onset of precipitation (collision–coalescence), is based on the assumption that droplets in undiluted clouds are distributed in space in a perfectly random manner, i.e. droplet positions are independently distributed with uniform probability. We have analysed data from a homogeneous cloud core to test this assumption and gain an understanding of the nature of droplet transport. This is done by examining one-dimensional cuts through clouds, using a theory originally developed for x-ray scattering by liquids, and obtaining statistics of droplet spacing. The data reveal droplet clustering even in cumulus cloud cores free of entrained ambient air. By relating the variance of droplet counts to the integral of the pair correlation function, we detect a systematic, scale-dependent clustering signature. The extracted signal evolves from sub- to super-Poissonian as the length scale increases. The sub-Poisson tail observed below mm-scales is a result of finite droplet size and instrument resolution. Drawing upon an analogy with the hard-sphere potential from the theory of liquids, this sub-Poisson part of the signal can be effectively removed. The remaining part displays unambiguous clustering at mm- and cm-scales. Failure to detect this phenomenon until now is a result of the previously unappreciated cumulative nature, or ‘memory,’ of the common measures of droplet clustering.

1. Introduction

The assumption of statistically uniform and independently distributed positions of droplets in homogeneous unmixed clouds underlies much of cloud physics (Pruppacher & Klett 1997; Rogers & Yau 1989). For example, the stochastic collection equation, which describes the growth of droplets via collision and coalescence, assumes that the cloud droplets are spatially distributed in a perfectly random fashion, that is, according to the Poisson process (Telford 1955). Similarly, it is assumed in the theory of droplet growth by condensation that, at least in unmixed cloud cores, droplets are Poisson-distributed within the vapour field (Srivastava 1989).

From a traditional turbulence perspective, however, it is not obvious that the perfect randomness assumption holds even in cloud cores free of boundary inhomogeneities. Indeed, it is known that high Reynolds number turbulence is characterized by spatial correlations of scalar concentration (Monin & Yaglom 1975; Sreenivasan & Antonia 1997; Warhaft 2000). It is therefore plausible that mixing of droplets (if regarded as passive tracers) by turbulence, or droplet–turbulence interactions, cause ‘patchiness’ of the droplet suspensions, thus violating the perfect randomness assumption. The presence and physical origin of particle clustering, if observed, can perhaps be interpreted in the context of the traditional scalar cascade picture. However, there

may be additional causes of patchiness, such as memory of the injection process or particle inertia. (From this perspective, recent results concerning passive scalars decoupled from turbulent flow (e.g. Shraiman & Siggia 2000) might not appear quite so surprising.)

Whatever the cause of patchiness (if observed) might be, it is noteworthy that the Poisson process assumption, so fundamental to cloud physics and, more generally, multiphase flows (e.g. Sundaram & Collins 1997), is consistent with neither Obukhov's $-5/3$ nor Batchelor's -1 power law scaling of the passive scalar spectrum, so fundamental to turbulence theory. Indeed, the defining feature of the Poisson process is statistical independence on all scales. Therefore, it is δ -correlated and has a flat white-noise spectrum. On the other hand, the weak scalar 'dissipation' suggests that there may not be sufficient time to establish the scalar cascade in the traditional sense – cloud droplets diffuse extremely slowly, with a time scale of weeks for diffusion across a Kolmogorov eddy. Rather than attempting to find a general theory for particle clustering, we confine ourselves to phenomenology; that is, establishing conclusively whether droplet clustering actually occurs in clouds, itself a matter of some controversy.

To test the Poisson process assumption we have selected data from an atmospheric flow characterized by a droplet concentration profile free of any trends or mean gradients: the core of a cumulus cloud. We find, however, that beyond the expected fluctuations resulting from a counting process ('shot noise'), there are enhanced fluctuations over a broad range of spatial scales. This form of 'intermittency', or departure from perfect randomness, is conveniently analysed using mathematical tools somewhat different from those typically employed in fluid mechanics. For example, it is common to discuss intermittency in turbulence in terms of deviations from a Gaussian process. However, continuous random variables typically encountered in turbulence and the 'countable', non-diffusing, random variables characterizing droplet distributions in clouds are distinct. Here, we suggest that a more appropriate standard for characterizing 'goodness of mixing' (Brodkey 1967, Sec. 14-4.G), or for detecting intermittency of 'countable scalars' such as droplets, should be a Poisson rather than a Gaussian process. The two standards can be quite different – for example, Poissonian tails are not as steep as Gaussian ones. One can still study departures from perfect randomness with the aid of a correlation (or structure) function, but this is the *pair* correlation function, specifically defined as the departure from perfect randomness at a given scale. The pair correlation function has been used to study clustering in widely differing physical processes, including photon bunching (Loudon 1983), clustering of galaxies (Peebles 1980), and inertial clustering of droplets in turbulence (Sundaram & Collins 1997; Wang, Wexler & Zhou 2000).

2. Theoretical background

Let the random variable N represent the fluctuating number of particles in a given fluid volume, and assume that the random process is *statistically homogeneous*, that is, the moments of N are unaffected by shifts in the choice of the origin. Note that the 'patchiness' of clouds (or fine-scale inhomogeneity in any given cloud) does not preclude statistical homogeneity. The state of perfect randomness is represented by the particle positions being independently distributed with uniform probability in the fluid volume. In terms of the fluctuating number of droplets, this state corresponds to the homogeneous Poisson process. Visually, Poissonian droplet distributions appear devoid of structure and resemble an ideal gas of molecules. More precisely, the

assumptions behind the Poisson process (e.g. Ochi 1990) are: (i) the process is statistically homogeneous; (ii) the probability of detecting more than one particle in a given volume dV is vanishingly small for sufficiently small dV ; (iii) particle counts in non-overlapping volumes are statistically independent random variables at any length scale. The last assumption is the true origin of the Poisson process.

Given these assumptions, the probability distribution of particle counts is given by

$$p(N) = \frac{\bar{N}^N \exp(-\bar{N})}{N!}, \quad (2.1)$$

where $p(N)$ is the probability of finding N particles in a test volume, and \bar{N} is the mean over all test volumes. For the Poisson distribution the variance of counts $(N - \bar{N})^2 \equiv (\delta N)^2$ equals the mean. Note that $(\delta N)^2 = \bar{N}$ can hold only for unitless, integer-valued random variables such as counts. This leads to the definition of the clustering index $CI \equiv (\delta N)^2 / \bar{N} - 1$, which is used to characterize departures from pure randomness in many fields of science.

We will study clustering of cloud droplets by analysing a time series of particle arrival times and will describe the required mathematical tools in this setting. Typically, the data are collected during an aircraft traverse through a cloud and droplet positions are recorded as they enter the probe. Hence, the cloud volume sampled by the probe is a long, narrow tube. To test the assumption of perfect randomness, we divide this tube into segments of length l , count the (random) number N of droplets in each segment, and compute the mean \bar{N} and variance $(\delta N)^2$ of droplet counts. The tube is then subdivided into different length segments and the procedure is repeated for each l . In this manner CI has been used to attribute droplet clustering to a given scale (e.g. Baker 1992; Chaumat & Brenguier 2000). Here we will show, however, that CI must be interpreted with care, as it has inherent ‘memory’ because its value at a given scale contains contributions from all smaller scales.

For segments so short as to contain either one or no particles, the clustering question can be stated as follows: Is the probability of detecting a droplet within the next segment enhanced by having just detected a droplet? The defining feature of pure randomness (Poisson process) is that such a conditional probability equals the unconditional probability of simply detecting a droplet within the next segment of the probe path. Consider two such tube segments of length dl_1 and dl_2 and let n denote linear droplet concentration (i.e. $n = cA$, where c is the droplet concentration and A is the constant cross-section of the data tube). The probability that a particle is in dl_1 is ndl_1 , which is also the average number of particles in dl_1 (Landau & Lifshitz 1980). Then, for a statistically homogeneous cloud with independently distributed particles, the joint probability $P(1, 2)$ of finding a droplet in each of the two segments is simply the product $(ndl_1)(ndl_2)$. If, however, droplets are clustered, then correlations are present and the joint probability is given by

$$P(1, 2) = (ndl_1)(ndl_2)[1 + \eta(l)], \quad (2.2)$$

where $\eta(l)$ is the pair correlation function for volumes separated by distance l . Thus, $\eta(l)$ measures the deviation from a perfectly random distribution of droplets. We emphasize that (2.2) is the most fundamental definition of the pair correlation function (Peebles 1980; Landau & Lifshitz 1980, p. 352). This definition makes clear that the range of η is $(-1, \infty)$, with $\eta = 0$ corresponding to perfect randomness. For example, $\eta(l) = 3$ yields a factor of 4 enhancement of finding another droplet distance l away from a given droplet. Likewise, $\eta(l) = -1$ represents impossibility of encountering another droplet distance l away from a given droplet.

It is natural to ask whether there is a connection between the two just-described measures of departure from perfect randomness. Such a connection does indeed exist. In the course of their studies of x-ray scattering by liquids, Ornstein & Zernike (1914) discovered that the mean-squared fluctuation $(\delta N)^2$ of particle counts (variance of N) in a given volume is related to the pair correlation function integrated over the same volume. Using our data tube geometry this can be written as

$$\frac{(\overline{\delta N})^2}{\bar{N}} - 1 = n \int_0^l \eta(l') dl', \quad (2.3)$$

where $n = \bar{N}/l$ is the linear concentration. This result is completely general in the sense that the derivation includes no assumptions regarding physical mechanisms (Landau & Lifshitz 1980). Recalling that for the Poisson distribution, $(\overline{\delta N})^2 = \bar{N}$, we see that the left-hand side of (2.3) is a natural measure of departure from a perfectly random distribution of particles, i.e. a measure of clustering. Hence, the previously measured clustering index (left-hand side of (2.3)) contains ‘memory’ of small scales – pair correlations at sub-resolution distances contribute to the volume integral. As will be shown, the cumulative effects can be reduced by using a volume-averaged pair correlation function defined as

$$\bar{\eta} = \frac{1}{l} \int_0^l \eta(l') dl' = \frac{(\overline{\delta N})^2}{\bar{N}^2} - \frac{1}{\bar{N}}. \quad (2.4)$$

The search for particle clustering on progressively finer scales eventually leads to the question of the importance of finite particle size. This also provides a convenient illustration of the relationship between η , $\bar{\eta}$, and CI . For example, for a uniform probability distribution of particle centres in the data tube, the spacing between nearest neighbours must obey the exponential distribution. If the mean separation is λ , then the probability density function of consecutive particle separations l is given by (e.g. Ochi 1990, pp. 432–433)

$$p(l) = \frac{1}{\lambda} \exp\left(-\frac{l}{\lambda}\right). \quad (2.5)$$

The most probable separation is zero and finite size will prevent this; that is, there should be no spacing less than the particle diameter D . The excluded volume fraction is then approximately $\phi = D/\lambda$. The probability of encountering a particle in a forbidden region is then simply ϕ because centre coordinates are uniformly distributed. For example, if λ is about 1 mm and D about 50 microns, ϕ is about 5% and we expect one ‘violation’ every 20 particles. Hence, we expect slightly sub-Poissonian variance, which can be expressed in the language of equation (2.3). Similar finite size effects arise in the context of turbulent collision rates (Reade & Collins 2000).

We can examine the possible effects of excluded volume on the otherwise perfectly random particle positions with the aid of a simple ‘hard sphere’ model. We treat the droplets as impenetrable spheres and use the definition (2.2) to give

$$\eta(l) = \begin{cases} -1 & \text{for } 0 < l < D \\ 0 & \text{for } l \geq D \end{cases} \quad (2.6)$$

where l is the distance between particle centres. Indeed, the lowest possible value of η is -1 , corresponding to impenetrability as in the case of hard-sphere incompressible fluids (Landau & Lifshitz 1980). Integration of η using equation (2.4) yields $\bar{\eta} = -D/l$ and $CI = -nD$, for l greater than D . These results quantify the sub-Poissonian vari-

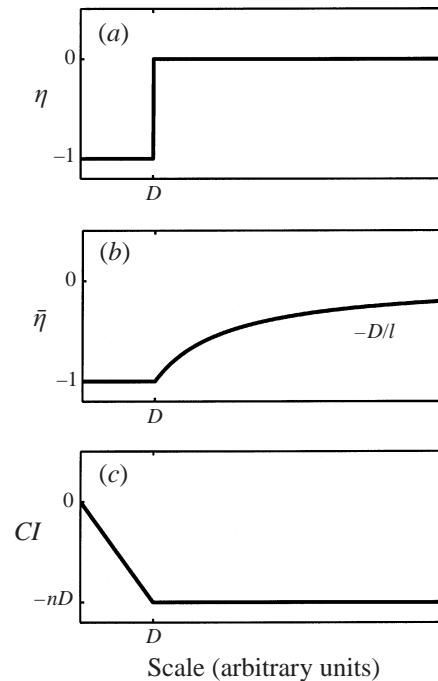


FIGURE 1. Schematic illustration of the effect of excluded volume on an otherwise perfectly random distribution of particle centres: (a) the pair correlation function η ; (b) the volume-averaged pair correlation function $\bar{\eta}$; and (c) the clustering index $CI = \overline{(\delta N)^2} / \bar{N} - 1$. Here, D corresponds to the diameter of a hard sphere or, for instruments, finite measurement resolution. Note that η , $\bar{\eta}$, and CI would be identically zero for a Poisson process.

ance caused by the excluded volume for an otherwise perfectly random distribution, and are illustrated in figure 1. We note that η is a local, scale-dependent measure of particle clustering, while $\bar{\eta}$ and CI are cumulative in nature. These cumulative effects persist indefinitely for CI , as seen in figure 1(c). In other words, even for perfect randomness at *all* scales larger than D one expects a negative, not zero, value of the clustering index as a result of droplet finite size.

3. Data analysis

We now proceed to analyse cloud droplet data collected during a single traverse by the Meteo-France Merlin IV research aircraft through a cumulus cloud encountered during the Small Cumulus Microphysics Study. Droplet positions were measured with the Meteo-France Fast Forward Scattering Spectrometer Probe (Fast FSSP) operating at a sampling frequency of 16 MHz, roughly equivalent to a spatial resolution of $6 \mu\text{m}$ (Chaumat & Brenguier 2000). The droplet concentration calculated at a resolution of approximately 1 m for the cloud traverse under consideration is shown in figure 2.

The region between approximately 500 and 700 m corresponds to the unmixed core of the cloud, characterized by statistically homogeneous conditions (see Kostinski & Jameson 2000, Appendix A) and defines the extent of the data used here. These data can be interpreted as a realization of a stationary but correlated random process. The cloud core is characterized by a root-mean-square vertical velocity fluctuation of approximately $U_z \approx 0.3 \text{ m s}^{-1}$ and a width of $L \approx 200 \text{ m}$, resulting in a Reynolds

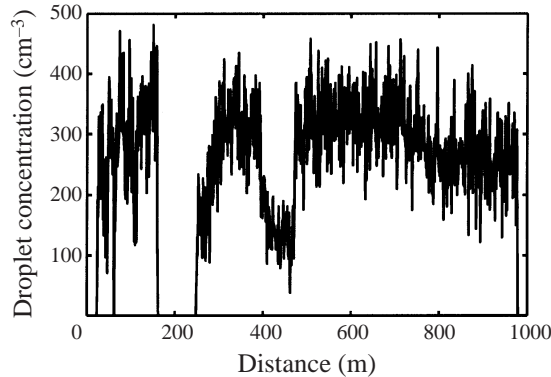


FIGURE 2. Droplet concentration in a single traverse of a cumulus cloud during the Small Cumulus Microphysics Study. Droplet concentration data are from the Fast FSSP operating at a sampling frequency of 100 Hz (approximately 1 m resolution). The droplet spacing data used in this study are from the unmixed, statistically homogeneous region of cloud located between approximately 500 and 700 m. The sampled cloud volume consists of about 2×10^5 droplets with radii between 12 and 13 μm , distributed in a long, narrow tube with cross-section of about 2 mm^2 .

number of order $Re \sim U_z L / \nu \sim 10^7$ and weak energy dissipation rate of order $\epsilon \sim U_z^3 / L \sim 10^{-4} \text{ m}^2 \text{ s}^{-3}$. Thus, while we would expect strong scalar intermittency as is observed in high-Reynolds-number turbulence, droplet–turbulence interactions will be weak because of the low dissipation rate. For example, the droplets encountered in this cloud have radii between 12 and 13 μm , resulting in a particle Stokes number of order $St = \rho_d d^2 \epsilon^{1/2} / 18 \rho \nu^{3/2} \sim 10^{-2}$, where ρ_d and ρ are the droplet and air mass densities, d is the droplet diameter, and ν is the kinematic viscosity of air (Shaw *et al.* 1998).

The Fast FSSP data tube cross-section is about 2 mm^2 and there are approximately 2×10^5 particles. Beginning with the first droplet, the distances to all subsequent droplets were recorded for the entire unmixed region. The data analysis consists of dividing the long tube of cloud air sampled by the Fast FSSP into segments of length l . This converts the data into a time series of droplet number counts per path increment (corresponding to a sampling volume element of about $2 \text{ mm}^2 \times l$). We then count the (random) number N of droplets in each segment and compute the mean \bar{N} and variance $(\delta N)^2$ of the droplet counts. The procedure is repeated for a range of l and the clustering index (CI) and volume-averaged pair correlation function $\bar{\eta}$ are calculated as a function of segment scale l .

The dependence of the clustering index on scale is shown in figure 3(a) for these data. For mm-length segments, the variance of droplet counts is slightly sub-Poissonian but increases monotonically with the interval length, seemingly reaching significantly super-Poissonian values at scales of about 1 m. Two questions arise: (i) Is the barely noticeable dip at sub-cm scale significant? (ii) Can the large value of CI at m-scales be interpreted as dramatic clustering at those scales? Previously, these results would have yielded ‘No’ and ‘Yes’ answers, respectively. With the aid of the pair correlation function and equation (2.3), we will demonstrate that neither conclusion is safe.

In figure 3(b), the volume-averaged pair correlation function $\bar{\eta}$ for the data is compared with a realization of an equivalent Poisson process. The sub-Poisson variance below cm-scales is strikingly similar to the $-D/l$ dependence of the simple hard sphere model; see figure 1(b) and the solid line in figure 3(b). For the solid line in

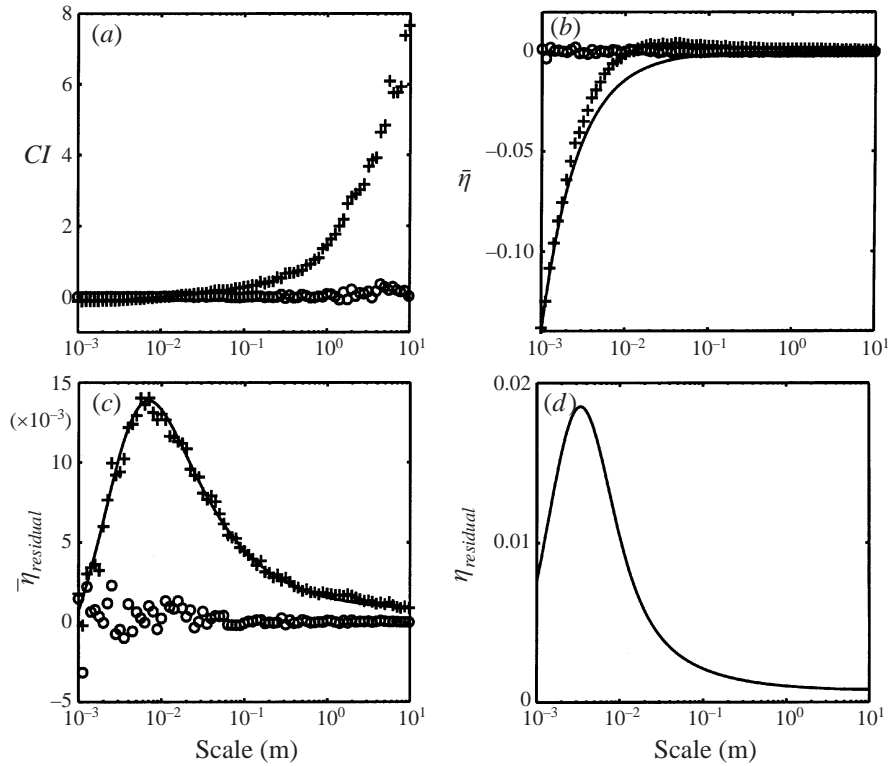


FIGURE 3. (a) CI versus l for the cloud data (+) and an equivalent-Poisson series (o), calculated as $CI = \overline{(\delta N)^2} / \bar{N} - 1$. The equivalent-Poisson series was generated with the same mean and number of particles as the data. (b) $\bar{\eta}$ versus l for the data (+) and an equivalent-Poisson series (o). The volume-averaged pair correlation function is computed via (2.3) as $\bar{\eta} = \overline{(\delta N)^2} / \bar{N}^2 - 1 / \bar{N}$. The sub-Poissonian values below cm-scales result from finite instrument resolution. The solid curve is similar to that shown in figure 1(b), where the effective exclusion diameter is $D_{eff} = 140 \mu\text{m}$, chosen to fit the negative tail of the data. (c) $\bar{\eta}_{res} \equiv \bar{\eta} + D_{eff}/l$ is shown (+) along with an equivalent-Poisson series (o). $\bar{\eta}_{res}$ quantifies clustering due to droplet-turbulence interactions with finite-resolution effects removed. The solid line is a fit to the data. (d) η_{res} versus l , calculated as $\eta_{res} = d(\bar{\eta}_{res}l)/dl$. To avoid numerical noise, the curve fit to $\bar{\eta}_{res}$ in (c) is used.

figure 3(b), the effective diameter is chosen as $D_{eff} = 140 \mu\text{m}$ to match the data. This is an order of magnitude greater than the actual diameter of a typical droplet but by systematically failing to resolve closely spaced droplets, the cloud probe volume introduces a larger excluded volume effect (in other probes this can also be created by the ‘dead time’ phenomenon). Indeed, a closer inspection of the Fast FSSP operation reveals that optical coincidences due to finite droplet and beam sizes are equivalent to the ‘repulsion’ of droplets on sub-mm scales. This is verified by a Monte Carlo computer experiment designed to imitate the Fast FSSP operation, as will be reported elsewhere in greater detail.

Figure 3(c) is a plot of the *residual* $\bar{\eta}$ (meaning excluded volume effects are removed) versus l , defined as $\bar{\eta}_{res} \equiv \bar{\eta} + D_{eff}/l$. This removes the effects of finite instrument resolution (figure 3(b), solid curve) and, therefore, can be interpreted more reliably in terms of particle clustering. The scale-dependent nature of the clustering signal is revealing, with super-Poissonian droplet clustering clearly peaked at a scale of about 1 cm and tapering off at larger scales. The trend of increasing pair correlation

function with decreasing scale observed at cm-scales is reminiscent of the signature for particle clustering in turbulence due to finite particle inertia (Sundaram & Collins 1997).

Ideally, one would calculate the pair correlation function η directly. With a limited data set and a single realization, such as that attainable in a cloud, however, this direct calculation lead to a noisy result, particularly at the smallest scales. We have, therefore, fitted a function to $\bar{\eta}_{res}$, as shown by the solid curve in figure 3(c). The residual pair correlation function η_{res} is calculated from $\bar{\eta}_{res}$ by rearranging (2.4) as $\eta_{res} = d(\bar{\eta}_{res}l)/dl$. The result of this calculation is shown in figure 3(d), yielding a persistent clustering signal at mm- and cm-scales. Extracting such a signal would not have been possible without the insight provided by equation (2.3). Furthermore, the equation allows us to compute the pair correlation function over a wide range of scales simply by counting droplets.

Note that $\bar{\eta}$ is a reasonable approximation of η . The magnitude of both is near 2% and it is understandably weak as the cloud encountered in the traverse used here was characterized by a very low energy dissipation rate (of order $10^{-4} \text{ m}^2 \text{ s}^{-3}$). However, the cumulative nature of clustering, as manifested by the clustering index, over the wide range of scales results in significantly non-Poissonian statistics at m-scales where $CI = (\overline{\delta N})^2 / \bar{N} - 1$ is approximately $\bar{N} \times \bar{\eta} = 10^3 \times 10^{-2} \sim 10$. Hence, the standard deviation of the population of m-segments of our data tube is about a factor of $\sqrt{10}$ more than that of an equivalent Poisson process. This is consistent with the conceptual picture of droplets being distributed in bunches interspersed with regions sparser in droplets (Kostinski & Jameson 2000, figure 1; Shaw *et al.* 1998, figure 1).

4. Concluding remarks

In this study we have attempted to advance the current understanding of particle–turbulence interactions in atmospheric clouds. Perhaps our findings can be interpreted with the aid of correlation functions and spectral densities commonly used in cascade arguments. Instead, we have chosen to work with the pair correlation function because it is specifically designed to measure, as a function of scale, the departure from perfect randomness for a countable scalar. The pair correlation function has been obtained here for droplets in a high Reynolds number turbulent flow. The crucial step, in our opinion, is the choice and application of theoretical tools most suited to the clearest, assumption-free interpretation of experimental data. To that end, (i) it is shown here how the pair correlation function can be interpreted and calculated, with the aid of equation (2.3), from a typical cloud traverse; (ii) sub-Poissonian variance is observed and explained via the excluded volume effect; (iii) the physical meaning of the clustering index is clarified by taking into account the inherent memory of this quantity; (iv) an unambiguous, scale-dependent clustering signal is extracted.

The results presented here suggest that previous measurements of droplet spatial distributions may have been ‘contaminated’ by the excluded volume effect. This may be one reason why a previous analysis of data from unmixed cloud cores in the same field experiment ended with the conclusion that there are no statistically significant departures from perfect randomness (Chaumat & Brenguier 2000). The super-Poissonian variance we detect in these homogeneous core data can be viewed as conclusive evidence of clustering. Because of the previously unappreciated cumulative nature of the clustering index (or ‘memory’ of small scales), the excluded volume effects at sub-cm scales weaken the signals at longer scales, which can then remain undetected. In view of the present findings, it is likely that cloud droplets are ‘mixed’

by turbulent air in an uneven and intermittent manner down to sub-mm scales. In other words, even in the homogeneous unmixed cloud cores, the droplets are distributed in bunches interspersed with regions depleted in droplets. This situation can be represented, for example, by the Poisson mixture approach (Kostinski & Jameson 2000).

The current study has focused on establishing whether droplet clustering exists and how it can be seen, but it is tempting to speculate briefly on possible mechanisms that lead to clustering. Aside from the traditional scalar cascade, one possibility is to regard cloud droplets as countable, non-diffusing scalars advected by turbulent air and yielding scalar fluctuations independent of the cascade. Another possibility is that clustering arises due to the inertial response of droplets to fluid accelerations in turbulence (Sundaram & Collins 1997; Shaw *et al.* 2000). This mechanism might be particularly important at large Re , as suggested by recent laboratory measurements of particle accelerations (Voth, Satyanarayan & Bodenschatz 1998).

In any case, the intrinsic cloud texture has implications for all theories of droplet growth because they rest, as mentioned in §1, on the Poissonian foundation. It is not surprising that numerical simulations based on such theories do not agree with observations of measures as essential as the width of droplet distributions in clouds (Lasher-Trapp & Cooper 2000). The problem is most severe in homogeneous, unmixed cloud cores where the liquid water content and the observed droplet sizes are both the largest. On the other hand, droplet clustering is expected to lead to broader droplet distributions through enhanced condensation and collision rates (Pinsky & Khain 1997; Sundaram & Collins 1997; Shaw *et al.* 1998). If true, this conclusion will have far-reaching consequences because the droplet distribution is of fundamental importance in understanding varied cloud processes such as precipitation formation, heterogeneous chemistry, and radiative transfer (Pruppacher & Klett 1997; Barker 1992; Jameson & Kostinski 2000).

The universality and quantitative range of the present observations on clustering can only be judged by making additional measurements. Most importantly, there is a need for measurements at higher dissipation rates and, preferably, with instruments less susceptible to resolution and coincidence counting problems. Implications of the clustering phenomenon for cloud physics and turbulence research will no doubt be better understood quantitatively as additional data become available.

We thank J.-L. Brenguier of Meteo-France for providing the Fast FSSP data and M. L. Larsen, K. R. Sreenivasan, and the anonymous reviewers for helpful comments. This work was supported by NSF grants ATM95-12685 (A. B. K.) and ATM99-84294 (R. A. S.).

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