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POSSIBLE IMPLICATIONS OF DROPLET CLUSTERING FOR RADIATIVE TRANSFER IN CLOUDS

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Exponential decay of radiation, described by the Beer-Lambert law, is of central importance in atmospheric radiative transfer. In the atmosphere, and especially for radiative transfer in clouds, there are physical scenarios where deviations from the Beer-Lambert law are expected (Barker, 1992; Cahalan et al., 1994; Davis and Marshak, 1997; Marshak et al., 1998; Weinman and Harshvardhan, 1982). It was shown in (Kostinski, 2001), using the language of the Poisson process, that correlations in the positions of particles in a random medium (e.g., droplets in a cloud) can lead to slower-than-exponential (sub-exponential) extinction of intensity with propagation depth. In this approach, exponential decay arises in a special case when the medium contains no correlations. A third possibility is that of *negative* correlations, which can lead to faster-than-exponential (super-exponential) extinction (Shaw et al., 2002).

Here we describe recent Monte Carlo studies of radiative transfer in an absorbing medium. The results confirm that sub-exponential decay occurs when the volume-averaged pair correlation function is greater than zero at the scale of interest and that the Beer-Lambert law is recovered when correlations vanish. Also, when the volume-averaged pair correlation function is negative super-exponential extinction with propagation distance occurs. These results are of special interest to the problem of radiative transfer in cloudy atmospheres, where the pair correlation function previously has been shown to be negative and positive at different scales (Kostinski and Shaw, 2001). For example, a turbulent mixing zone characterized by a $-5/3$ scalar spectrum may possess strong positive correlations in particle position. On the other hand, sedimenting particles in a laminar fluid possess strong negative correlations in particle position (Lei et al., 2001), and an atmospheric example might be droplets in a calm fog. It also is a simple matter to conceive of macroscopic cloud geometries that would possess either negative or positive correlations (e.g., cellular convection versus turbulent mixing at cloud top).

Extinction in a correlated medium: Theory

To begin with, we consider a random medium consisting of absorbing particles. To obtain deviations from the Beer-Lambert law, it is necessary to introduce correlations among positions of particles in the medium. Note that, in spite of correlations, the distribution of particles is still regarded as statistically homogeneous so that all moments (e.g., mean and variance) are invariant with respect to the shift of origin. Examples of spatially correlated particle distributions are shown in Fig. 1, which will be discussed in more detail later. Briefly, the left panel shows negatively-correlated particle positions, the middle panel shows uncorrelated particle positions for reference, and the right panel shows positively-correlated particle positions.

For the radiation extinction problem we must consider two 'countable' random variables: the number of particles or obstacles k, and the number of photon absorption events n. The degree of spatial correlation between particles is quantified by the pair correlation function, $\eta(r)$ (Larsen et al., 2002). For uncorrelated particle positions $\eta(r) = 0$ for all scales. When $\eta \neq 0$ the *conditional* probability of finding a second particle at a distance r from the first one is enhanced or inhibited by a factor of $(1 + \eta(r))$.

The spatial correlation of particles is related to the variance of particle counts in a fixed volume by (Kostinski, 2001)

$$\frac{\overline{(\delta K)^2}}{\bar{K}} - 1 = \frac{\bar{K}}{V} \int_V \eta dV, \quad (1)$$

where $\delta K \equiv K - \bar{K}$ is the deviation from the mean count in a given volume V and $\bar{K} = \bar{k}V$ where $\bar{k}$ is the local mean concentration. After multiplying both sides of Eq. (1) by $\bar{K}$ and rearranging we obtain $\overline{(\delta K)^2} = \bar{K} + \bar{\eta}\bar{K}^2$, where $\bar{\eta} = V^{-1} \int_V \eta dV$ is the volume-averaged pair correlation function. In the

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Poissonian case, $\eta = \bar{\eta} = 0$, and this gives the well-known result that $\overline{(\delta K)^2} = \bar{K}$ for a Poisson distribution. We note that the variance of counts can be less than or greater than $\bar{K}$, depending on the sign of $\bar{\eta}$. Indeed, $\bar{\eta}$ is negative at all scales for negatively correlated particle positions such as shown in the left panel of Fig. 1, and $\bar{\eta}$ is positive at all scales for positively correlated particle positions such as shown in the right panel of Fig. 1.

In (Kostinski, 2001) it was argued that variance of particle counts is related to the rate of extinction of radiation. As a specific example, super-Poissonian variance arising when $\bar{\eta} > 0$ was shown to result in slower-than-exponential extinction. Similarly, sub-Poissonian variance of obstacle number yields faster-than-exponential attenuation (Shaw et al., 2002). To understand these results we consider the relevance of the Poisson process to the Beer-Lambert law of exponential extinction. We assume perfect randomness in the distribution of obstacles so that the number of absorbed photons obeys the Poisson distribution:

$$p_n(x) = \frac{\overline{n(x)}^n exp(-\overline{n(x)})}{n!}, \qquad (2)$$

where n is the random number of *absorbed* photons in the test volume per unit time, $p_n(x)$ is a probability of having n photons absorbed in a given volume of a layer of depth x, and $\overline{n(x)}$ is the mean count over many realizations as a function of the depth x into the slab. Next, we consider the photon probability of transmission (no extinction) through the layer of depth x. That is, we need to find $p_0(x)$ from Eq. (2) by setting $n = 0$ ($\bar{n}$ held constant): $p_0(x) = \exp -\overline{n(x)} = \exp -x/\lambda$, where $\lambda^{-1} = \sigma\bar{k}$ with λ, σ and $\bar{k}$ being the mean free path, extinction cross-section per obstacle, and obstacle concentration, respectively. Now, using the law of large numbers to interpret $p_0(x)$, we can rewrite $p_0(x)$ as $N_{tr}/N_{inc} = \exp -x/\lambda$, which is the *stochastic equivalent* of the Beer-Lambert law. Here, N_{inc} and N_{tr} stand for the (large) number of incident and transmitted photons, respectively and x/λ is the unitless optical depth.

In Eq. (2) we used the fact that the number of absorbed photons (at a given location) is Poisson distributed because the number of obstacles is Poisson distributed. If, however, the number of obstacles is not Poisson-distributed, then $p_n(x)$ will also change. For example, if the variance in the number of obstacles decreases, so does the variance in the number of absorbed photons n. This means that the distribution $p(n)$ gets narrower (only 'well-behaved' densities are considered for the moment). Qualitatively, we expect that $p(0)$ will decrease as well because a narrower pdf will have 'lower tails'. Therefore, at a fixed mean free path, the numerical value of the narrow (sub-Poissonian) pdf produces lower $p(0)$ than does the Poissonian case. Furthermore, the argument holds at any $\bar{n}$ which, in turn, depends linearly on x, penetration distance into the medium. Hence, we conclude that negative correlations yield faster-than-exponential extinction. Analogous arguments hold for positive correlations and slower-than-exponential extinction.

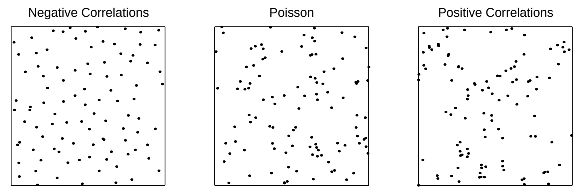

Figure 1: Thin slices (slice thickness is 1/10th the width shown), with n and σ the same in all cases, showing particle positions in typical realizations of the Monte Carlo model. Note that in all three panels the box width is equal to one free path, defined as $1/n\sigma$. The first panel corresponds to negatively correlated particle positions, achieved by specifying an 'excluded volume' around each particle. The second panel is for a completely uncorrelated medium, i.e., Poisson process. The third panel is for positively correlated particle positions, achieved via an exponential conditional probability of particle position.

Monte Carlo model and results

To test the theory outlined in the previous section, we built a straightforward Monte Carlo model that performs two functions. First, the model generates a random distribution of particle positions, which may be spatially correlated: this gives us variable k. Second, the model calculates a distribution of photon absorption events by 'shooting' photons through the distribution of particles: this gives us variable n. These procedures are described briefly below and additional details are available in (Shaw et al., 2002).

Three different types of particle distributions are shown in Fig. 1. Each panel is a thin slice (slice thickness is 1/10th the width shown) taken from one realization of particle positions generated by the model. The middle panel shows particle positions that are uncorrelated at all scales, or Poisson distributed. The Poisson distribution is made by selecting each particle's coordinate independently and at random from a uniform pdf. Thus each particle's position is uncorrelated with that of any other. The left panel shows particle positions that are negatively correlated over a wide range of scales. Negatively correlated distributions can be made by randomly selecting particle positions inside the box, but rejecting any position that is less than a specified distance away from any other previously placed particle. The right panel of Fig. 1 shows positively-correlated particle positions, achieved by using a conditional pdf to select a random distance from the most recently placed particle to the next particle's location. The scale-dependence of spatial correlations between particles is found by calculating the pair correlation function $\eta(r)$, where r is the radial distance from one particle to another. For a distribution such as in the middle panel of Fig. 1, $\eta(r) = 0$ for all r. For the right panel showing the positively-correlated model, $\eta(r) \geq 0$ for all r. For the 'excluded-volume' model shown in the left panel of Fig. 1, $\eta(r) = -1$ for $r \leq r_\circ$ and the volume-averaged pair correlation function $\overline{\eta(r)}$ is negative at all scales.

Once a particle distribution is generated, the extinction of radiation is calculated by shooting photons through the volume and keeping track of the number of photons remaining as a function of propagation depth x. Since the incident radiation is assumed to be incoherent, it could be represented by randomly positioned photons travelling along straight trajectories, all parallel to each other, until one passes through a particle and an absorption event occurs. All particles (obstacles) were assumed to be perfectly absorbing, so no scattering needed to be considered. In addition, all particles were assumed to have the same absorption cross section, σ.

All three particle distributions had an average particle density, $\bar{k}$, of 1000 particles per unit volume. The value of σ was then chosen to be 10^{-3} so that the optical depth, $\lambda = \sigma\bar{k}$ would be unity. For each distribution, 10^6 randomly positioned photons were sent into the particle cloud and 'measurements' of the number of photons that were not absorbed were made at several depths, yielding an extinction rate. Results for the three particle distributions are shown in Fig. 2, where the logarithm of relative intensity $I/I_\circ$ is plotted against propagation distance normalized by the mean free path x/λ. Lack of spatial correlations faithfully reproduces the exponential decay predicted by the Beer-Lambert law, with $\lambda^{-1} = \bar{k}\sigma$. Extinction of radiation propagating through the positively-correlated particle distribution is significantly slower than the Beer-Lambert law would predict. Finally, the model confirms that for negatively-correlated particle positions (more precisely, for $\bar{\eta} < 0$) the rate of extinction of radiation is faster than expected from the Beer-Lambert law.

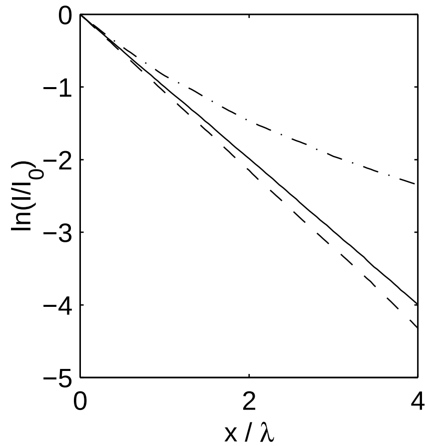

Figure 2: Relative intensity plotted as a function of propagation depth, calculated with the Monte Carlo model described in the text. The middle curve (solid) is for the uncorrelated medium and is identical to that expected from the Beer-Lambert law. The top curve (dot-dashed), representing slower-than-exponential decay, is for the positively correlated medium. The bottom curve (dashed), representing faster-than-exponential decay, is for the negatively correlated medium.

While the extinction rate shown in Fig. 2 for the negatively-correlated particle distribution is faster than the expected rate based on the particle number density and cross section, further examination reveals that after a transient regime the extinction rate does, in fact, approach negative exponential (e.g., it is a straight line in Fig. 2). A similar approach to exponential extinction, but with a modified optical depth $\lambda^* > \lambda$, occurs for positively-correlated particle positions characterized by a 'short-range' pair correlation function. Qualitatively, the final exponential extinction with an 'effective' cross-section results when all scales with non-zero correlation are averaged over.

These results are of special interest to the problem of radiative transfer in cloudy atmospheres, where the pair correlation function previously has been shown to be negative and positive at different scales. Additional work on relating the extinction rate and 'effective' optical depth to the pair correlation function is ongoing. This work must be done in conjunction with theoretical and observational studies to understand the scale dependence of the pair correlation function for particles in atmospheric clouds.

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