

# On the extinction of radiation by a homogeneous but spatially correlated random medium

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Exponential extinction of incoherent radiation intensity in a random medium (sometimes referred to as the Beer–Lambert law) arises early in the development of several branches of science and underlies much of radiative transfer theory and propagation in turbid media with applications in astronomy, atmospheric science, and oceanography. We adopt a stochastic approach to exponential extinction and connect it to the underlying Poisson statistics of extinction events. We then show that when a dilute random medium is statistically homogeneous but spatially correlated, the attenuation of incoherent radiation with depth is often slower than exponential. This occurs because spatial correlations among obstacles of the medium spread out the probability distribution of photon extinction events. Therefore the probability of transmission (no extinction) is increased. © 2001 Optical Society of America

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## 1. INTRODUCTION

Propagation of radiation in a random medium is a ubiquitous problem, and the Beer–Lambert law of exponential extinction is one of its basic elements. However, possible deviations from exponential attenuation have been recently explored in several studies, particularly in the context of the radiative transfer equation. To the best of the author's knowledge, the earliest study of the effects of medium heterogeneity on transmission characteristics was that of Romanova in 1975.<sup>1</sup> In 1982, Weinman and Harshvardhan<sup>2</sup> concluded that internal cloud geometry was a major factor in deducing the radiative properties. Integration over the probability distribution of optical depths and the Jensen inequality for the exponential function [ $\exp(-\tau) \geq \exp(-\bar{\tau})$ ], where  $\tau$  is the optical depth, were used by Stephens *et al.*<sup>3</sup> and Newman *et al.*,<sup>4</sup> respectively, to calculate the effective transmission. A little later, the Jensen inequality and fractal modeling of clouds were used in Marshak *et al.*<sup>5</sup> to interpret the observation that spatially heterogeneous clouds transmit more and reflect less than “equivalent” (same-water-content) homogeneous clouds. The effects of heterogeneity and nonexponential transmission were also considered by Davis *et al.* in Refs. 6 and 7 from the kinetic theory point of view, relying on fractal modeling. A fractal model was employed in Ref. 8 as well, in order to model small-scale structure, and it was concluded that exponential extinction is modified in radiative transfer through vegetation canopy. The related research in the field of stochastic transport is summarized in Ref. 9. Finally, for an example of nonexponential transmission that is due to line-shape effects, the reader is referred to the monograph on atmospheric radiation by Goody and Yung (Ref. 10, p. 132).

The purpose of this contribution is to examine the validity of and possible departures from exponential attenuation of incoherent (no-phase) radiation propagating in a statistically homogeneous but spatially correlated dilute

random medium. The examination proceeds at a fundamental level, a single particle at a time, in the spirit of classical kinetic theory. Neither a specific radiative transfer formalism nor the concept of optical depth is used in the approach. This allows consideration of correlation lengths comparable with the mean free path. Furthermore, no assumptions of fractal behavior or any other scale invariance are required. The concept of the pair correlation function is employed to describe the structure of a dilute random medium, and the notion of the Poisson random process is used as a bridge to the Beer–Lambert law from the stochastic viewpoint.

To distill the essence of the argument, we consider the simplest example: a parallel beam of incoherent radiation, incident normally on a slab containing a random collection of obstacles. Let us also assume that the interparticle distances are much larger than particle size (dilute random medium) and that when photons are absorbed, re-emission occurs at a different frequency and does not contribute to the original beam, and that scattering is negligible compared with absorption. It would appear that, with all these simplifying assumptions, exponential extinction must surely hold, but one subtlety remains.

Our random medium is assumed to be statistically homogeneous, that is, its statistical characteristics, such as moments, are invariant with respect to translation. However, this still leaves the possibility of correlations in the spatial positions of the obstacles. Let us quote from (Ref. 11, p. 351): “The assertion that the particles of a homogeneous isotropic body (liquid or gas) are equally likely to be at any position in space, applies to each separate particle on condition that all the other particles can have arbitrary positions. This assertion certainly does not contradict the fact that, owing to their interaction, there must exist some correlation between the relative positions of the different particles. This means that if we consider, say, two particles at the same time, then for a

given position of one particle, different positions of the other will not be equally probable.”

The question addressed in this paper is the following: Does the correlation in the positions of obstacles affect exponential extinction? Our purpose is to show that the attenuation of radiation with penetration depth into a spatially correlated but statistically homogeneous dilute random medium is often slower than exponential.

To that end, we begin by establishing the close connection between exponential extinction and Poissonian statistics in the simplest correlation-free (perfectly random) medium. The stochastic interpretation of the Beer–Lambert law is arrived at by regarding the number of extinction events (e.g., absorbed photons) in a given volume as the fundamental random variable. We then proceed to show that spatial correlations among obstacles cause deviations from the Poisson distribution of extinction events. Finally, it is shown that super-Poissonian fluctuations yield slower-than-exponential extinction.

To motivate the development intuitively before embarking on a formal treatment, perhaps it is helpful to consider the following simple analogy. Compare the photon’s view of going through the random layer with visibility in a forest. Imagine a line of observers parallel to and looking through a layer of sparsely populated woods. The photon free path is analogous to the line of sight.

For the perfectly random forest (without any clumps of trees), an exponential distribution of free paths (distribution among observers of the lengths of the lines of sight) holds; e.g., see Ref. 12, p. 257. This exponential distribution is equivalent to the Poisson distribution of the (random) number of obstacles per area and yields exponential extinction. Let  $\Lambda$  denote the mean free path or average visibility. Next, while preserving statistical homogeneity and without changing concentration, redistribute the trees to form patches and voids throughout the layer. The distribution of lines of sight will change. For example, there is now a higher probability of going through  $3\Lambda$  because some lucky observers happen to be looking through several aligned voids, and this is why one might expect the attenuation with distance to be slower than exponential (more photons here than in the Poissonian case leak through a slab of  $3\Lambda$ ). The probability of the free path being much lower than  $\Lambda$  is also higher than in the Poissonian case because of the patches of high concentration. Therefore the path-length distribution is broader in the patchy case, i.e., has larger variance (the mean held constant).

## 2. STOCHASTIC INTERPRETATION OF THE BEER–LAMBERT LAW

Consider a uniform parallel incoherent photon beam of cross-sectional area  $A$ , incident normally on an infinite slab of a dilute random medium of depth  $x$ . The slab is sufficiently deep to contain many randomly positioned obstacles (e.g., cloud droplets). Let the number of extinction events (e.g., photons absorbed by the particles of the medium) per unit volume and time be our fundamental random variable. Furthermore, we assume that obstacle positions are purely random, i.e., statistically independent

of each other. Therefore the obstacle count per unit volume is described by the Poisson distribution. This is so also for the probability distribution of the extinction events (as in kinetic theory of the classical ideal gas, where the number of collisions satisfies Poissonian statistics; e.g., see Ref. 13). In other words, the number of extinction events satisfies the following conditions: (a) Extinction by the slab represents a statistically homogeneous (stationary) random process (that is, the statistics are independent of the location in the slab); (b) the probability of extinction of more than one photon in a given width  $\delta x$  is vanishingly small for sufficiently small slab width  $\delta x$ ; and (c) extinction events in nonoverlapping volumes are statistically independent random variables. These three assumptions define the Poisson process; e.g., see Ref. 14. Therefore the number of absorbed photons obeys the Poisson distribution:

$$p_n(x) = \frac{\overline{n(x)^n \exp[-\overline{n(x)}]}}{n!}, \quad (1)$$

where  $n$  is the random number of, say, absorbed photons in the test volume per unit time,  $p_n(x)$  is the 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. Either we can view  $x$  as a random variable and  $n$  as a parameter or vice versa or both. A possible question might be, for  $n = 2$  (e.g., doubly scattered photon or two absorption events of a photon if a photon is reintroduced after first absorption), What is the distribution of a random variable  $x$ ? In this study, we will adhere to the other interpretation, however, in which  $x$  (and therefore  $\overline{n}$ ) are held constant and the focus is on the distribution of  $n$ .

The Poisson distribution satisfies  $(n - \overline{n})^2 = (\delta n)^2 = \overline{n}$  (the variance equals the mean). Note that this important relation can hold only for unitless integer-valued random variables (counts) because it is not invariant with respect to scaling. It will be shown below that natural variability (patchiness) yields overdispersion through an additional term  $\propto \overline{n}^2$ .

What is the relevance of this to the Beer–Lambert law of exponential extinction? Consider next the photon probability of transmission (no extinction) through the layer of depth  $x$ . That is, we need to find  $p_0(x)$  from Eq. (1) by setting  $n = 0$  ( $\overline{n}$  held constant). We obtain

$$p_0(x) = \exp[-\overline{n(x)}] = \exp(-\beta x), \quad (2)$$

where  $\beta = \sigma c = \Lambda^{-1}$ , with  $\Lambda$ ,  $\sigma$ , and  $c$  being the mean free path, the extinction cross section per obstacle, and the obstacle concentration, respectively. This is in complete analogy with the kinetic theory of ideal gas [Ref. 13 (p. 12) or Ref. 15]. The average number of absorbed photons is  $\overline{n(x)} = \beta x = pN$ , where  $p$  is the probability of absorption (ratio of extinction cross section to beam cross section,  $\sigma/A$ ) and  $N$  is the total number of obstacles in the volume  $V$ , so that  $cV = cAx$ . Hence  $\beta = \sigma c$ .

Now, by invoking the law of large numbers to interpret  $p_0(x)$ , we can rewrite Eq. (2) as

$$\frac{N_{\text{tr}}}{N_{\text{inc}}} = \exp(-\beta x), \quad (3)$$

which is the stochastic equivalent of the Beer–Lambert law. Here  $N_{\text{inc}}$  and  $N_{\text{tr}}$  stand for the (large) number of incident and transmitted photons, respectively, and  $\beta x$  is the optical depth.

Recall that the exponential distribution is memoryless (e.g., see Ref. 16), that is, it satisfies factorization of transmission probabilities:  $\exp[-(x+y)] = \exp(-x)\exp(-y)$  for nonoverlapping layers  $x$  and  $y$ . The close connection with the statistical independence assumption is seen in the following well-known and more direct derivation of Eq. (3).

Subdivide the layer of depth  $x$  into many differential layers of depths  $dx_1$ ,  $dx_2$ , etc. Then the probability of transmission through each layer is only slightly less than unity and is given by  $1 - \beta dx_1$ , while the probability of transmission through the whole layer is given as (note the assumption of statistical independence among the sublayers)

$$p_{\text{tr}} = (1 - \beta dx_1)(1 - \beta dx_2) \cdots (1 - \beta dx_m). \quad (4)$$

Next, take logarithms of both sides, expand in Taylor series, and keep the first-order term in the expansion to obtain

$$\ln(p_{\text{tr}}) = -\beta dx_1 - \beta dx_2 - \cdots - \beta dx_m = -\beta x, \quad (5)$$

and, after exponentiation, Eq. (3) results.

We now ask: What happens if assumption (c) above (statistical independence) is relaxed? Now correlations appear, and there is spatial memory. Does this memory affect the distribution of photon extinction events? Indeed, it does, by causing deviations from the Poissonian process and, therefore, departures from the exponential extinction, as is shown in Section 3.

### 3. SPATIAL CORRELATIONS AMONG OBSTACLES

Let us introduce correlations among positions of obstacles in the slab; e.g., cloud droplets can cluster and exhibit voids elsewhere. Physically, the formation of patches or filaments can be caused by, for example, turbulent advection of the droplets or interplay of vorticity and inertia; e.g., see a brief review of the literature in Refs. 17–19. Note that, in spite of correlations, the distribution of particles is still regarded as statistically homogeneous (all moments are invariant with respect to the shift of origin), as pointed out in the quote from Ref. 11, given in Section 1. Below, we will use  $k$  for the random number of obstacles (to distinguish it from the random number of photon absorption events  $n$ ).

Let two volume elements  $dV_1$  and  $dV_2$  be sufficiently small that they can contain either zero or one obstacle only and that the probability of containing two or more particles is negligible. Hence the average number of particles  $\bar{k}dV$  is also the probability that a particle is in the volume element  $dV$  (Ref. 11, p. 351).

Then, for a statistically homogeneous random field, the joint probability  $P(1, 2)$  of finding a particle in each of the two volumes  $dV_1$  and  $dV_2$  is

$$P(1, 2) = \bar{k}^2 dV_1 dV_2 [1 + \eta(l)], \quad (6)$$

where  $\bar{k}dV$  is the probability of finding a particle in  $dV$ ,  $\eta(l)$  is the pair correlation function, and  $l$  is the separation distance between two volumes [statistical isotropy is implied by  $\eta = \eta(l)$ ]. A somewhat less direct but perhaps a more practical definition of  $\eta(l)$  is given by

$$\eta(l) \equiv \frac{\overline{[K(l)K(0) - \bar{K}]^2}}{\bar{K}^2} = \frac{\overline{K(l)K(0)}}{\bar{K}^2} - 1, \quad (7)$$

where  $K$  is the random number of obstacles in a test volume<sup>20</sup> and  $\bar{K} = \bar{k}V$  is the expected number of such particles. We see from Eq. (6) that the assumption of statistical independence of counts in nonoverlapping volumes implies that  $\eta(l) = 0$  because only in this case is the joint probability simply a product of the individual ones. However, in the presence of correlations, the *conditional* probability of finding the second particle, given that the first is there, is enhanced (or inhibited) by a factor of  $1 + \eta$ . Equivalently, from the definition of  $\eta(l)$  in Eq. (7), it can be seen that the pair correlation function is identically zero in the absence of correlations (the Poissonian case).

If we interpret a patch or a cluster as a region of positive correlation, then the Poisson process is ideally random in the sense that only in the case of the Poisson process are there no patches or voids at any length scale. Presumably, the stronger the correlation, the more substantial the violation of the statistical independence assumption required in Poissonian statistics and, therefore, the larger the deviations from the Poissonian variance. In other words, the following question arises: Is there a relationship between the variance of obstacle counts in a fixed sampling volume and the spatial correlation of obstacles?

It turns out that a powerful formula is available, which indeed relates the variance of counts in a given volume to the pair correlation function, integrated over the same volume. It was originally developed for the case of density fluctuations in gases and liquids, but the derivation is completely general, as can be found in Ref. 11, p. 352, Eq. 116.5:

$$\frac{\overline{(\delta K)^2}}{\bar{K}} - 1 = \frac{\bar{K}}{V} \int_V \eta dV, \quad (8)$$

where, as in Eq. (6),  $\eta$  is the pair correlation function between particle counts in volumes  $V_1$  and  $V_2$  within  $V$ ,  $\delta K \equiv K - \bar{K}$  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. (The  $\eta$  here differs from the  $\nu$  in Ref. 11 by the factor  $\bar{K}/V$ .) Note that in the limiting case of no correlation, we recover the Poisson relation  $\overline{(\delta K)^2} = \bar{K}$ .

After multiplying through by  $\bar{K}$  both sides of Eq. (8) and rearranging, we obtain

$$\overline{(\delta K)^2} = \bar{K} + \bar{\eta} \bar{K}^2, \quad (9)$$

where  $\bar{\eta} = V^{-1} \int_V \eta dV$  is the volume-averaged pair corre-

lation function. In the Poissonian case,  $\eta = \bar{\eta} = 0$ . Another interpretation of the Poissonian limit can be obtained by letting the dimensions of the sampling volume  $V$  increase well beyond the correlation distance while keeping  $\bar{K}$  fixed. For such large distances,  $\eta$  is nearly zero, most of the volume in the expression for  $\bar{\eta}$  does not contribute to the integral, and  $\bar{\eta}$  approaches zero. Finally, cancellations of positive and negative  $\eta$  regions can also occur. Then the relation  $(\delta K)^2 = \bar{K}$  is approached. While Eq. (9) is completely general, in Section 4 we derive it with the aid of a simple model and then use the associated probability density function to obtain nonexponential extinction.

#### 4. SUPER-POISSONIAN MODEL YIELDS SLOWER-THAN-EXPONENTIAL ATTENUATION

We now come back to  $n$  (the number of removed photons) as the fundamental random variable. Naturally, the spatial correlations discussed in Section 3 yield spatial correlations in the number of absorbed photons in nearby volumes. Indeed, the number of extinction events will obey Eq. (1) as long as the local mean concentration of obstacles remains constant. However, on longer spatial scales, the local concentration itself will fluctuate as the lines of sight and the detectors move along the slab from a cluster to a void. The local mean number of absorption events will fluctuate along with the local concentration. Alternatively, we can imagine [in the spirit of Eq. (4)] that various sublayers of the slab are correlated spatially with each other, so that their voids are more likely to be partially aligned and more radiation can leak through the layer.

Let us, for simplicity of notation, use the shorthand  $p(n)$  for  $p_n(x)$  and  $\bar{n}$  for  $\bar{n}(x)$  until the new distribution is derived and then come back to the original notation to emphasize the spatial dependence in  $n(x)$ .

It can be seen, in view of Section 3 (also see Ref. 19, Sect. 6) that the local mean number of adsorbed photons  $\bar{n}$  associated with a given patch must itself be regarded as a random variable when correlations are present. Thus Eq. (1) holds only when conditioned upon a constant mean extinction rate of a given blob, and to obtain the total distribution, one must integrate over the distribution of local mean extinction rates [call it  $p(\bar{n})$ ] as follows:

$$p(n) = \int_0^\infty p(n|\bar{n})p(\bar{n})d\bar{n} = \int_0^\infty \frac{\bar{n}^n \exp(-n)}{n!} p(\bar{n})d\bar{n}, \quad (10)$$

where the vertical bar denotes conditional probability. Hence the process is doubly stochastic; i.e., Poissonian fluctuations in the number of absorbed photons per unit volume ride on top of the longer-scale cluster-to-void fluctuations of local scattering rate. This approach (known as Mandel's formula; e.g., see Refs. 14 and 20–22) has been used in photon optics to describe chaotic light. A physically plausible  $p(\bar{n})$  needs to be chosen next. Again, following the photonics example,<sup>14</sup> we pick the exponential function. There are other reasons such as the fact that the exponential satisfies the lack-of-memory

property mentioned in Section 2. Also, for cloud physics applications, we note that recent research in fluid dynamics indicates that the concentration probability density of passive contaminants suspended in turbulence tends to an exponential form under a rather wide set of conditions [see Ref. 19 (Sect. 6) or Ref. 23 for a brief review of the literature and a discussion of basic physics]. Furthermore, there is no real loss of generality in choosing the exponential, as will be discussed below. Therefore set

$$p(\bar{n}) = \frac{1}{\mu} \exp\left(-\frac{\bar{n}}{\mu}\right), \quad (11)$$

where  $\mu$  is the global (averaged over experiments with many correlation volumes) mean number of absorbed photons. The result of integration in Eq. (10) with Eq. (11) inside the integral is the geometric probability distribution (e.g., see Ref. 14, p. 473):

$$p(n) = \frac{1}{\mu + 1} \left( \frac{\mu}{\mu + 1} \right)^n. \quad (12)$$

Note that there is (as in the case of the Poisson distribution) only a single parameter here—the mean extinction rate  $\mu$ —but the shape of this distribution is completely different from the Poissonian one (much longer tail). The geometric variance can be much larger than the Poissonian value of  $\mu$  and is given by

$$\overline{(\delta n)^2} = \mu + \mu^2, \quad (13)$$

which is in agreement with the correlation-fluctuation relation (9) when  $\bar{\eta} = 1$ . Had we assumed a more general  $\Gamma$ -distribution family (rather than the exponential distribution) for the distribution of concentration, the answer (e.g., see Refs. 14 and 18) would have been modified only by a constant in front of the  $\mu^2$  term and would have been similar to the completely general equation  $(\delta n)^2 = \mu + \bar{\eta}\bar{\mu}^2$ , derived in Section 3. As discussed above, there are two independent sources of randomness whose contributions therefore add: the regular Poissonian fluctuations [ $\overline{(\delta n)^2} = \mu$ ] and the longer-scale patch fluctuations [ $\overline{(\delta n)^2} = \mu^2$ ].

We now come back explicitly to the question of spatial attenuation. Recall that  $\mu(x) = \beta x$ , where again  $\beta = \sigma c$  but the concentration  $c$  is averaged over many patches of local concentration  $\bar{k}$ . Next, we compute the probability of no extinction ( $n = 0$ ) from Eq. (12). This yields [switching to the original notation  $p(n) = p_n(x)$  to stress the spatial dependence]

$$p_0(x) = \frac{1}{\mu + 1} = \frac{1}{\beta x + 1}, \quad (14)$$

which can be rewritten as

$$\frac{N_{\text{tr}}}{N_{\text{inc}}} = \frac{1}{1 + \beta x}. \quad (15)$$

Hence the attenuation with distance is slower than exponential [compare with Eq. (3)].

It is important to note that any other function  $p(\bar{n})$  in



Eq. (10) still yields larger-than-Poissonian variance and, therefore, slower-than-exponential attenuation. For example, a more elaborate  $\Gamma$ -distribution model (rather than the exponential one) yields a negative-binomial distribution of counts (e.g., see Refs. 18 and 21) and, thereby, power-law attenuation  $N_{\text{tr}}/N_{\text{inc}} \propto 1/(1 + \beta x)^m$ , where  $m$  is the parameter of the  $\Gamma$ -function. Since the variance of the negative-binomial distribution is  $\overline{(\delta n)^2} = \mu + \mu^2/m$ , it is identical to the completely general correlation-fluctuation result  $\overline{(\delta n)^2} = \mu + \bar{\eta}\bar{\mu}^2$  when  $\bar{\eta} = m^{-1}$ . Hence the model presented here entails no significant loss of generality.

Geometrically, any description of the patchiness  $p(\bar{n})$  still implies spatial correlations and, by Eq. (8), increased variance, which, in turn, leads to deviations from the exponential attenuation. Indeed, physically, the super-Poissonian variance implies that, for a given depth  $x$ , probabilities of having zero and “many” absorbed photons are both larger than the Poissonian values (at the same mean  $\mu$ ) because of the broadening of the probability density  $p(n)$ , that is, because of the variance enhancement (e.g., see Fig. 1 in Ref. 19). This is so because the likelihood of several voids to conspire and align along the line of sight is enhanced by the spatial correlations among obstacles.

Given the importance of the Beer–Lambert law, the general (insofar as it is insensitive to a particular correlation model) conclusion of power-law attenuation with distance appears important. It is natural to inquire whether the deviations have ever been observed experimentally. For a large amount of absorbing material (but not so large that the signal is not detectable), the difference between  $\exp(-\beta x)$  and  $\propto 1/(1 + \beta x)$  may be observable as long as absorption dominates. However, when  $\beta x$  is small, Eqs. (3) and (15) both yield  $1 - \beta x$  to first order, while to second order the expressions are  $1 - \beta x + (\beta x)^2$  and  $1 - \beta x + (\beta x)^2/2$  respectively. This may be important when, for example, one wants to stay in a single-scattering regime (if scattering rather than absorption dominates). For instance, the difference at  $\beta x = 0.1$  is only approximately 0.5% and is 2% at  $\beta x = 0.2$ . Encouraging experimental results have recently been presented in Ref. 24 showing that the power-law photon free path distributions proposed in Ref. 7 and consistent with Eq. (15) here or with the attenuation decaying as  $1/(1 + \beta x)^m$  appear realistic when clouds are present.

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