



Contents lists available at ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

A generalized linear Boltzmann equation for non-classical particle transport

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ARTICLE INFO

Available online 13 July 2010

Keywords:

Particle transport

Random media

Correlated scattering centers

ABSTRACT

This paper presents a derivation and initial study of a new *generalized linear Boltzmann equation* (GLBE), which describes particle transport for random statistically homogeneous systems in which the distribution function for chord lengths between scattering centers is non-exponential. Such problems have recently been proposed for the description of photon transport in atmospheric clouds; this paper is a first attempt to develop a Boltzmann-like equation for these and other related applications.

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1. Introduction

In the standard (classical) theory of linear particle transport, the incremental probability dp that a particle at point \mathbf{x} with energy E will experience an interaction while traveling an incremental distance ds is given by

$$dp = \Sigma_t(\mathbf{x}, E) ds,$$

where Σ_t (the cross section) is independent of the direction of flight Ω and the path length s , defined by

$$s = \text{the distance traveled by the particle since its previous interaction (birth or scattering)}. \quad (1.1)$$

(In this definition, the instant after a particle is born or scatters, its value of s is 0.) The assumption that Σ_t is independent of Ω and s is valid when the locations of the scattering centers in the system are uncorrelated. However, to explain experimental observations of solar radiation in atmospheric clouds, researchers have recently suggested that the locations of the water droplets in clouds in fact *are* correlated, in ways that measurably affect radiative transfer within the cloud [1–10]. If a cloud is modeled by taking the water droplets to be randomly positioned (but correlated), with potential scattering

centers at all points within the cloud droplets, then Σ_t is no longer independent of both s and Ω . If the positions of the scattering centers are correlated and the correlations are independent of direction Ω , then Σ_t is independent of Ω but not s : $\Sigma_t = \Sigma_t(\mathbf{x}, E, s)$. In this situation, a Boltzmann-type equation for the radiation field has not previously been derived. The purpose of this paper is to derive and analyze such an equation.

For simplicity, we do not consider the most general problem here. Our analysis is based on five primary assumptions:

- A₁** The physical system is infinite and statistically homogeneous. (This paper presents an initial theory, intended to be valid in the absence of statistical inhomogeneities and finite system boundaries.)
- A₂** Particle transport is monoenergetic. (The inclusion of energy- or frequency-dependence is straightforward.)
- A₃** Particle transport is driven by a specified isotropic source $Q(\mathbf{x})$ satisfying $Q \rightarrow 0$ as $|\mathbf{x}| \rightarrow \infty$, and the particle flux $\rightarrow 0$ as $|\mathbf{x}| \rightarrow \infty$.
- A₄** The ensemble-averaged total cross section $\Sigma_t(s)$, defined as

$\Sigma_t(s) ds$ = the probability (ensemble – averaged over all physical realizations) that a particle, scattered

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or born at any point \mathbf{x} , and traveling in any direction Ω , will experience a collision between $\mathbf{x}+s\Omega$ and $\mathbf{x}+(s+ds)\Omega$,

(1.2)

is known; see discussion below. (For problems in general random media, $\Sigma_t(s)$ depends also on \mathbf{x} and Ω . In this paper, the statistics are assumed to be homogeneous and independent of the direction of flight, in which case Σ_t depends only on s .)

A₅ The distribution function $P(\Omega \cdot \Omega')$ for scattering from Ω' to Ω is independent of s . (The correlation in the scattering center positions affects the probability of collision, but not the scattering properties when scattering events occur. This assumption is valid when the system consists of “chunks” of two materials, one of which is a void; or when the materials consist of the same atomic species at different densities.)

In practice, $\Sigma_t(s)$ can be determined by the following “experimental” procedure, which applies for any situation in which computer realizations of the random system can be generated.

1. Construct a realization of the system, and let $\Sigma_t(\mathbf{x}) =$ the total cross section at point \mathbf{x} in the system.
2. Let: (i) \mathbf{x} be a random scattering center in the realization, (ii) Ω be a random direction of flight, and (iii) ξ be a uniformly distributed random number on the interval (0,1]. Then, from the standard Monte Carlo implementation of the Boltzmann transport equation [11,12], the equation

$$-\ln \xi = \int_0^s \Sigma_t(\mathbf{x}+s'\Omega) ds'$$

determines the random distance s to the next collision site.

Fig. 1 depicts this calculation. Here, the non-overlapping discs represent solid material, at each point within which particles can scatter, surrounded by void, within which particles freely stream and do not scatter. [The discs could be interpreted as water droplets in a cloud; each point within a disc (water droplet) is a potential scattering center.]

3. If $\mathbf{x}+s\Omega = \mathbf{y}$ is inside the realization, then “bin” the resulting value of s .

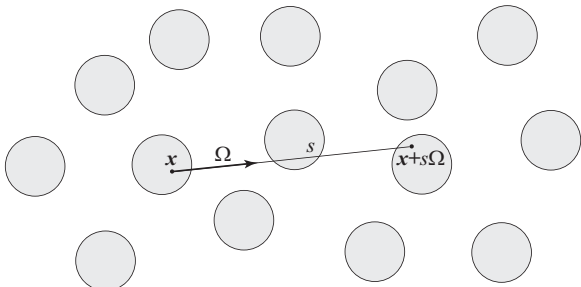


Fig. 1. Calculation of random distance s to collision.

4. Perform a large number of similar calculations of s , using a large number of different points \mathbf{x} , directions Ω , and realizations, to compile an accurate histogram approximation to $p(s)$, the ensemble-averaged distribution function for the distance to collision.
5. Finally, by Eq. (4.7) below, $\Sigma_t(s)$ is defined in terms of $p(s)$ by

$$\Sigma_t(s) = \frac{p(s)}{1 - \int_0^s p(s') ds'}$$

An alternative formulation of $p(s)$ can be obtained as follows. For a specific physical realization, let \mathbf{x} be a random scattering center (e.g. a random point inside one of the discs in Fig. 1) and Ω a random direction of flight. Then

$$P(\mathbf{x}, \Omega, s) = \Sigma_t(\mathbf{x}+s\Omega) e^{-\int_0^s \Sigma_t(\mathbf{x}+s'\Omega) ds'}$$

satisfies

$P(\mathbf{x}, \Omega, s) ds =$ the probability that a particle, released at \mathbf{x} in the direction Ω , will experience its first collision while traveling a distance between s and $s+ds$.

Thus, for the specified realization, $P(\mathbf{x}, \Omega, s)$ is the distribution function for the distance to collision s for a particle released at \mathbf{x} in the direction Ω . Then $p(s)$ is the ensemble average

$$p(s) = \langle P(\mathbf{x}, \Omega, s) \rangle_{(\mathbf{x}, \Omega, \mathcal{R})} \quad (1.3)$$

over all scattering centers \mathbf{x} in the realization, all directions of flight Ω , and all possible realizations \mathcal{R} .

For a specific random system, is the ensemble-averaged $p(s)$ exponential? To address this question, let us consider an almost trivial example: a system in which each realization is spatially uniform, but with probability p_1 has the total cross section $\Sigma_{t,1}$ and with probability $p_2 = 1 - p_1$ has the total cross section $\Sigma_{t,2}$. If the total cross section in the system is $\Sigma_{t,1}$, then

$$p(\mathbf{x}, \Omega, s) = \Sigma_{t,1} e^{-\Sigma_{t,1}s},$$

and if the total cross section in the system is $\Sigma_{t,2}$, then

$$p(\mathbf{x}, \Omega, s) = \Sigma_{t,2} e^{-\Sigma_{t,2}s}.$$

Therefore, the ensemble-averaged $p(s)$ is

$$p(s) = p_1 \Sigma_{t,1} e^{-\Sigma_{t,1}s} + p_2 \Sigma_{t,2} e^{-\Sigma_{t,2}s},$$

which is not exponential. Because even this trivial example has a non-exponential $p(s)$, it seems evident that the ensemble-averaged $p(s)$ for a general random system will not be exponential.

However, there are situations in which $p(s)$ is well-approximated by an exponential. For example, if the chunk size of the two materials is very small compared to a mean free path, then the atomic mix approximation—in which the cross sections are approximated by their volume averages—becomes accurate. In this approximation, the resulting linear Boltzmann equation with homogenized cross sections certainly has an exponential $p(s)$. Also, for systems similar to the depiction in Fig. 1, if the mean distance between the discs is large compared to the

radii of the discs, and if the centers of the discs are uncorrelated, then $p(s)$ is again well-approximated by an exponential.

For systems similar to the depiction in Fig. 1, a contributing factor to a non-exponential $p(s)$ is the correlation (or lack thereof) between the centers of the discs. For atmospheric clouds, the relationship between the experimentally observed non-exponential $p(s)$ and the correlations between the locations of the water droplets is an active area of research [1–10].

The fundamental purpose of this paper is to develop and discuss a Generalized Linear Boltzmann Equation (GLBE) for a random system satisfying the five assumptions stated above—in which Eq. (1.2) holds and $\Sigma_t(s)$ is known.

From the above discussion, the GLBE approximates the flux for any specific physical realization by the flux for a hypothetical problem in which all particles travel a distance s to collision that is consistent with the ensemble-averaged probability distribution function $p(s)$. The GLBE does not “know” about the particular geometrical structure of any specific realization; it treats all realizations identically through the ensemble-averaged $p(s)$.

The GLBE approximates a random medium by preserving the ensemble-averaged distribution function for distance-to-collision. The simpler atomic mix model approximates a random medium by shrinking the chunk sizes to zero. Because the GLBE preserves an important element of physics in random systems that is not preserved by the atomic mix approximation, we expect that the GLBE will generally be more accurate than the atomic mix equation. In this paper, we show by numerical simulations of a problem in nuclear engineering that this expectation is met.

This paper is an expanded version of a recent conference paper [13]. A summary of the remainder of the paper follows. In Section 2 we state definitions and derive the GLBE. In Section 3 we show that if $\Sigma_t(s)$ is independent of s , the GLBE yields the classical mono-energetic Boltzmann equation. In Section 4 we derive (i) the distribution function $p(s)$ for the distance s to collision in terms of $\Sigma_t(s)$, and (ii) the equilibrium path length spectrum. In Section 5 we reformulate the GLBE in terms of integral equations in which s (and for isotropic scattering, Ω) is absent; and in Section 6 we show that the GLBE has a straightforward asymptotic diffusion limit. Section 7 describes numerical results, based on Monte Carlo simulations, that confirm the validity of the GLBE. We conclude with a discussion in Section 8.

2. Derivation of the GLBE

Using the familiar notation $\mathbf{x} = (x, y, z)$ = position and $\Omega = (\Omega_x, \Omega_y, \Omega_z)$ = direction of flight (with $|\Omega| = 1$), and using Eq. (1.1) for s , we define:

$$n(\mathbf{x}, \Omega, s) dV d\Omega ds = \text{the number of particles in } dV d\Omega ds \text{ about } (\mathbf{x}, \Omega, s), \quad (2.1a)$$

$$v = \frac{ds}{dt} = \text{the particle speed}, \quad (2.1b)$$

$$\psi(\mathbf{x}, \Omega, s) = v n(\mathbf{x}, \Omega, s) = \text{the angular flux}, \quad (2.1c)$$

$$\Sigma_t(s) ds = \text{the probability that a particle that has traveled a distance } s \text{ since its previous interaction (birth as a source particle or scattering) will experience its next interaction while traveling a further distance } ds, \quad (2.1d)$$

$$c = \text{the probability that when a particle experiences a collision, it will scatter (} c \text{ is independent of } s), \quad (2.1e)$$

$$P(\Omega' \cdot \Omega) d\Omega = \text{the probability that when a particle with direction of flight } \Omega' \text{ scatters, its outgoing direction of flight will lie in } d\Omega \text{ about } \Omega \text{ (} P \text{ is independent of } s), \quad (2.1f)$$

$$Q(\mathbf{x}) dV = \text{the rate at which source particles are isotropically emitted by an internal source } Q(\mathbf{x}) \text{ in } dV \text{ about } \mathbf{x}. \quad (2.1g)$$

Then, classic manipulations directly lead to

$$\begin{aligned} \frac{\partial}{\partial s} \psi(\mathbf{x}, \Omega, s) dV d\Omega ds &= \frac{\partial}{v \partial t} v n(\mathbf{x}, \Omega, s) dV d\Omega ds \\ &= \frac{\partial}{\partial t} n(\mathbf{x}, \Omega, s) dV d\Omega ds \\ &= \text{the rate of change of the number of particles in } dV d\Omega ds \text{ about } (\mathbf{x}, \Omega, s), \end{aligned} \quad (2.2a)$$

$$\begin{aligned} |\Omega \cdot \mathbf{n}| \psi(\mathbf{x}, \Omega, s) dS d\Omega ds &= \text{the rate at which particles in } d\Omega ds \text{ about } (\Omega, s) \text{ flow through an incremental surface area } dS \text{ with unit normal vector } \mathbf{n}, \\ \Omega \cdot \nabla \psi(\mathbf{x}, \Omega, s) dV d\Omega ds &= \text{the net rate at which particles in } d\Omega ds \text{ about } (\Omega, s) \text{ flow (leak) out of } dV \text{ about } \mathbf{x}, \end{aligned} \quad (2.2b)$$

$$\begin{aligned} \Sigma_t(s) \psi(\mathbf{x}, \Omega, s) dV d\Omega ds &= \Sigma_t(s) \frac{ds}{dt} n(\mathbf{x}, \Omega, s) dV d\Omega ds \\ &= \frac{1}{dt} [\Sigma_t(s) ds] [n(\mathbf{x}, \Omega, s) dV d\Omega ds] \\ &= \text{the rate at which particles in } dV d\Omega ds \text{ about } (\mathbf{x}, \Omega, s) \text{ experience collisions.} \end{aligned} \quad (2.2c)$$

The treatment of the in-scattering and source terms requires extra care. From Eq. (2.2c),

$$\begin{aligned} \left[\int_0^\infty \Sigma_t(s') \psi(\mathbf{x}, \Omega', s') ds' \right] dV d\Omega' \\ = \text{the rate at which particles in } dV d\Omega' \text{ about } (\mathbf{x}, \Omega') \text{ experience collisions.} \end{aligned}$$

Multiplying this expression by $cP(\Omega \cdot \Omega') d\Omega$, we obtain

$$\begin{aligned} cP(\Omega \cdot \Omega') \left[\int_0^\infty \Sigma_t(s') \psi(\mathbf{x}, \Omega', s') ds' \right] dV d\Omega' d\Omega \\ = \text{the rate at which particles in } dV d\Omega' \text{ about } (\mathbf{x}, \Omega') \text{ scatter into } dV d\Omega \text{ about } (\mathbf{x}, \Omega). \end{aligned}$$

Integrating this expression over $\Omega' \in 4\pi$, we get

$$\left[c \int_{4\pi} \int_0^\infty P(\Omega' \cdot \Omega) \Sigma_t(s') \psi(\mathbf{x}, \Omega', s') ds' d\Omega' \right] dV d\Omega$$

= the rate at which particles scatter
into $dV d\Omega$ about (\mathbf{x}, Ω) .

(2.2d)

Finally, when particles emerge from a scattering event, their value of s is “reset” to $s=0$. Therefore, the path length spectrum of particles that emerge from scattering events is the delta function, $\delta(s)$. Multiplying the previous expression by $\delta(s) ds$, we obtain

$$\left[\delta(s) c \int_{4\pi} \int_0^\infty P(\Omega' \cdot \Omega) \Sigma_t(s') \psi(\mathbf{x}, \Omega', s') ds' d\Omega' \right] dV d\Omega ds$$

= the rate at which particles scatter
into $dV d\Omega ds$ about (\mathbf{x}, Ω, s) .

(2.2e)

Also,

$$\delta(s) \frac{Q(\mathbf{x})}{4\pi} dV d\Omega ds = \text{the rate at which source particles are emitted into } dV d\Omega ds \text{ about } (\mathbf{x}, \Omega, s).$$
(2.2f)

To proceed, we use the familiar conservation equation [in each of the following terms, the phrase “of particles in $dV d\Omega ds$ about (\mathbf{x}, Ω, s) ” is omitted]:

$$\begin{aligned} \text{Rate of change} &= \text{Rate of gain} - \text{Rate of loss} \\ &= (\text{In-scatter rate} + \text{Source rate}) \\ &\quad - (\text{Net leakage rate} + \text{Collision rate}). \end{aligned}$$

Introducing Eqs. (2.2) into this expression and dividing by $dV d\Omega ds$, we obtain the following *generalized linear Boltzmann equation* (GLBE) for $\psi(\mathbf{x}, \Omega, s)$:

$$\begin{aligned} \frac{\partial \psi}{\partial s}(\mathbf{x}, \Omega, s) + \Omega \cdot \nabla \psi(\mathbf{x}, \Omega, s) + \Sigma_t(s) \psi(\mathbf{x}, \Omega, s) \\ = \delta(s) c \int_{4\pi} \int_0^\infty P(\Omega' \cdot \Omega) \Sigma_t(s') \psi(\mathbf{x}, \Omega', s') ds' d\Omega' + \delta(s) \frac{Q(\mathbf{x})}{4\pi}. \end{aligned}$$
(2.3)

To repeat, we have for simplicity assumed an infinite homogeneous system with a “local” source $Q(\mathbf{x})$; and we take $\psi(\mathbf{x}, \Omega, s) \rightarrow 0$ as $|\mathbf{x}| \rightarrow \infty$.

Eq. (2.3) can be written in a mathematically equivalent way, in which the delta function is absent. We write Eq. (2.3) for $s > 0$:

$$\frac{\partial \psi}{\partial s}(\mathbf{x}, \Omega, s) + \Omega \cdot \nabla \psi(\mathbf{x}, \Omega, s) + \Sigma_t(s) \psi(\mathbf{x}, \Omega, s) = 0. \quad (2.4a)$$

Then, we operate on Eq. (2.3) by

$$\lim_{\varepsilon \rightarrow 0} \int_{-\varepsilon}^{\varepsilon} (\cdot) ds,$$

and we use $\psi = 0$ for $s < 0$ and define

$$\psi(\mathbf{x}, \Omega, 0) = \lim_{s \rightarrow 0^+} \psi(\mathbf{x}, \Omega, s) = \psi(\mathbf{x}, \Omega, 0^+) \quad (2.4b)$$

to obtain

$$\psi(\mathbf{x}, \Omega, 0) = c \int_{4\pi} \int_0^\infty P(\Omega' \cdot \Omega) \Sigma_t(s') \psi(\mathbf{x}, \Omega', s') ds' d\Omega' + \frac{Q(\mathbf{x})}{4\pi}. \quad (2.4c)$$

Eqs. (2.4) are mathematically equivalent to Eq. (2.3). In particular, we emphasize that the existence of a delta function in Eq. (2.3) does not imply that the

solution of this equation is singular. In the present situation, it implies that ψ is discontinuous at $s=0$, causing $\partial \psi / \partial s$ to become singular. [Eq. (2.4b) is a useful *definition* of ψ at $s=0$ that we use in the “initial condition” (2.4c).]

To establish the relationship between the present work and the classic number density and angular flux, we integrate Eq. (2.1a) over s and obtain

$$\left[\int_0^\infty n(\mathbf{x}, \Omega, s) ds \right] dV d\Omega = \text{the total number of particles in } dV d\Omega \text{ about } (\mathbf{x}, \Omega).$$

Therefore, consistently with Eqs. (2.1), we have

$$N(\mathbf{x}, \Omega) = \int_0^\infty n(\mathbf{x}, \Omega, s) ds = \text{classic number density} \quad (2.5)$$

and

$$\Psi(\mathbf{x}, \Omega) = \nu N(\mathbf{x}, \Omega) = \int_0^\infty \psi(\mathbf{x}, \Omega, s) ds = \text{classic angular flux.} \quad (2.6)$$

3. The classic linear Boltzmann equation

If $\Sigma_t(s) = \Sigma_t$ is independent of s , we can operate on Eq. (2.3) by $\int_{-\varepsilon}^\infty (\cdot) ds$ to obtain an equation for the classic angular flux $\Psi(\mathbf{x}, \Omega) = \int_0^\infty \psi(\mathbf{x}, \Omega, s) ds$. Using $\psi(\mathbf{x}, \Omega, -\varepsilon) = \psi(\mathbf{x}, \Omega, \infty) = 0$ and $\Sigma_s = c \Sigma_t$, we easily obtain

$$\Omega \cdot \nabla \Psi(\mathbf{x}, \Omega) + \Sigma_t \Psi(\mathbf{x}, \Omega) = \Sigma_s \int_{4\pi} P(\Omega' \cdot \Omega) \Psi(\mathbf{x}, \Omega') d\Omega' + \frac{Q(\mathbf{x})}{4\pi}. \quad (3.1)$$

This, of course, is the classic linear Boltzmann equation.

4. The path length and equilibrium path length distributions

Let us consider a single particle, which is released from an interaction site at $x=0$ in the direction $\Omega = \mathbf{i}$ = direction of the positive x -axis. Eq. (2.4a) for this particle becomes

$$\frac{\partial}{\partial s} \psi(x, s) + \frac{\partial}{\partial x} \psi(x, s) + \Sigma_t(s) \psi(x, s) = 0. \quad (4.1)$$

For this particle, we have

$$x(s) = s \quad \text{and} \quad \psi(x(s), s) \equiv F(s). \quad (4.2a)$$

Therefore,

$$\frac{dF}{ds}(s) = \frac{\partial \psi}{\partial x}(x(s), s) \left(\frac{dx}{ds} \right) + \frac{\partial \psi}{\partial s}(x(s), s) = \frac{\partial \psi}{\partial x} + \frac{\partial \psi}{\partial s}. \quad (4.2b)$$

Eq. (4.1) simplifies to

$$\frac{dF}{ds}(s) + \Sigma_t(s) F(s) = 0. \quad (4.3a)$$

We apply the initial condition

$$F(0) = 1, \quad (4.3b)$$

because we are considering a single particle. The solution of Eqs. (4.3) is

$$\begin{aligned} F(s) &= e^{-\int_0^s \Sigma_t(s') ds'} \\ &= \text{the probability that the particle will travel the distance } s \text{ without interacting.} \end{aligned} \quad (4.4)$$

The probability of a collision between s and $s+ds$ is

$$\Sigma_t(s) F(s) ds = p(s) ds, \quad (4.5)$$

and therefore

$$p(s) = \Sigma_t(s) e^{-\int_0^s \Sigma_t(s') ds'} = \text{distribution function for the distance – to – collision.} \quad (4.6)$$

Eq. (4.6) expresses $p(s)$ in terms of $\Sigma_t(s)$. To express $\Sigma_t(s)$ in terms of $p(s)$, we operate on Eq. (4.6) by $\int_0^s (\cdot) ds'$ to get

$$\int_0^s p(s') ds' = 1 - e^{-\int_0^s \Sigma_t(s') ds'}$$

or

$$e^{-\int_0^s \Sigma_t(s') ds'} = 1 - \int_0^s p(s') ds'.$$

Hence,

$$\int_0^s \Sigma_t(s') ds' = -\ln\left(1 - \int_0^s p(s') ds'\right).$$

Differentiating with respect to s , we obtain

$$\Sigma_t(s) = \frac{p(s)}{1 - \int_0^s p(s') ds'}. \quad (4.7)$$

Eqs. (4.6) and (4.7) easily show that $p(s)$ is exponential if and only if $\Sigma_t(s)$ is independent of s .

For the case of an infinite medium, with an “equilibrium” intensity having no space or direction-dependence, Eq. (2.3) for $s > 0$ reduces to

$$\frac{d\psi}{ds}(s) + \Sigma_t(s)\psi(s) = 0, \quad (4.8)$$

which has the solution

$$\psi(s) = A e^{-\int_0^s \Sigma_t(s') ds'}. \quad (4.9)$$

Normalizing this solution to have integral = unity, we obtain

$$\chi(s) = \frac{e^{-\int_0^s \Sigma_t(s') ds'}}{\int_0^\infty e^{-\int_0^{s'} \Sigma_t(s'') ds''} ds'} = \text{“equilibrium” spectrum of path lengths } s. \quad (4.10)$$

From Eq. (4.6), the mean distance to collision (mean free path) is

$$\begin{aligned} \langle s \rangle &= \int_0^\infty s p(s) ds \\ &= \int_0^\infty s \left[\Sigma_t(s) e^{-\int_0^s \Sigma_t(s') ds'} \right] ds \\ &= s \left[-e^{-\int_0^s \Sigma_t(s') ds'} \right]_0^\infty - \int_0^\infty \left[-e^{-\int_0^s \Sigma_t(s') ds'} \right] ds \\ &= \int_0^\infty e^{-\int_0^s \Sigma_t(s') ds'} ds. \end{aligned} \quad (4.11)$$

Thus, Eq. (4.10) may be written as

$$\chi(s) = \frac{1}{\langle s \rangle} e^{-\int_0^s \Sigma_t(s') ds'}. \quad (4.12)$$

For classic particle transport, in which $\Sigma_t(s) = \Sigma_t = \text{constant}$, Eq. (4.12) yields

$$\chi(s) = \Sigma_t e^{-\Sigma_t s}.$$

Hence, for classic particle transport in an infinite medium problem with no space or angle-dependence, the distribution

of particles at each spatial point and direction of flight that have traveled a distance s from their previous collision is exponential. (This result is not surprising.)

5. Integral equation formulations of the GLBE

Let us now define

$$\begin{aligned} f(\mathbf{x}, \boldsymbol{\Omega}) &= \int_0^\infty \Sigma_t(s) \psi(\mathbf{x}, \boldsymbol{\Omega}, s) ds \\ &= \text{collision rate density} \end{aligned} \quad (5.1)$$

and

$$\begin{aligned} g(\mathbf{x}, \boldsymbol{\Omega}) &= c \int_{4\pi} P(\boldsymbol{\Omega}' \cdot \boldsymbol{\Omega}) f(\mathbf{x}, \boldsymbol{\Omega}') d\Omega' \\ &= \text{in-scattering rate density.} \end{aligned} \quad (5.2)$$

In the following, we derive integral equations for f and g that do not contain the path length variable s as an independent variable. Also, if scattering is isotropic [$P(\boldsymbol{\Omega}' \cdot \boldsymbol{\Omega}) = 1/4\pi$], then $g(\mathbf{x}, \boldsymbol{\Omega})$ in Eq. (5.2) becomes isotropic:

$$g(\mathbf{x}) = \frac{c}{4\pi} \int_{4\pi} f(\mathbf{x}, \boldsymbol{\Omega}') d\Omega' \equiv \frac{c}{4\pi} F(\mathbf{x}), \quad (5.3a)$$

where

$$F(\mathbf{x}) = \int_{4\pi} f(\mathbf{x}, \boldsymbol{\Omega}') d\Omega' = \text{scalar collision rate density.} \quad (5.3b)$$

In this case, we derive an integral equation for $F(\mathbf{x})$ which is independent of both s and $\boldsymbol{\Omega}$.

First, using the definition (5.1), we write Eqs. (2.4) as

$$\frac{\partial \psi}{\partial s}(\mathbf{x}, \boldsymbol{\Omega}, s) + \boldsymbol{\Omega} \cdot \nabla \psi(\mathbf{x}, \boldsymbol{\Omega}, s) + \Sigma_t(s) \psi(\mathbf{x}, \boldsymbol{\Omega}, s) = 0, \quad (5.4a)$$

$$\psi(\mathbf{x}, \boldsymbol{\Omega}, 0) = c \int_{4\pi} P(\boldsymbol{\Omega}' \cdot \boldsymbol{\Omega}) f(\mathbf{x}, \boldsymbol{\Omega}') d\Omega' + \frac{Q(\mathbf{x})}{4\pi}. \quad (5.4b)$$

Solving Eq. (5.4a) and using Eq. (5.4b), we obtain for $s > 0$

$$\begin{aligned} \psi(\mathbf{x}, \boldsymbol{\Omega}, s) &= \psi(\mathbf{x} - s\boldsymbol{\Omega}, \boldsymbol{\Omega}, 0) e^{-\int_0^s \Sigma_t(s') ds'} \\ &= \left[c \int_{4\pi} P(\boldsymbol{\Omega}' \cdot \boldsymbol{\Omega}) f(\mathbf{x} - s\boldsymbol{\Omega}, \boldsymbol{\Omega}') d\Omega' + \frac{Q(\mathbf{x} - s\boldsymbol{\Omega})}{4\pi} \right] e^{-\int_0^s \Sigma_t(s') ds'}. \end{aligned} \quad (5.5)$$

Operating on this equation by $\int_0^\infty \Sigma_t(s) (\cdot) ds$ and using Eqs. (5.1) and (4.6), we get

$$f(\mathbf{x}, \boldsymbol{\Omega}) = \int_0^\infty \left[c \int_{4\pi} P(\boldsymbol{\Omega}' \cdot \boldsymbol{\Omega}) f(\mathbf{x} - s\boldsymbol{\Omega}, \boldsymbol{\Omega}') d\Omega' + \frac{Q(\mathbf{x} - s\boldsymbol{\Omega})}{4\pi} \right] p(s) ds. \quad (5.6a)$$

Also, operating on Eq. (5.5) by $\int_0^\infty (\cdot) ds$ and using Eq. (2.6), we obtain

$$\begin{aligned} \Psi(\mathbf{x}, \boldsymbol{\Omega}) &= \int_0^\infty \left[c \int_{4\pi} P(\boldsymbol{\Omega}' \cdot \boldsymbol{\Omega}) f(\mathbf{x} - s\boldsymbol{\Omega}, \boldsymbol{\Omega}') d\Omega' \right. \\ &\quad \left. + \frac{Q(\mathbf{x} - s\boldsymbol{\Omega})}{4\pi} \right] e^{-\int_0^s \Sigma_t(s') ds'} ds. \end{aligned} \quad (5.6b)$$

Eq. (5.6a) is an integral equation for $f(\mathbf{x}, \boldsymbol{\Omega})$. If this equation is solved, then Eq. (5.6b) yields the classic angular flux $\Psi(\mathbf{x}, \boldsymbol{\Omega})$.

Next, we use the definition (5.2) and write Eq. (5.6a) as

$$f(\mathbf{x}, \boldsymbol{\Omega}) = \int_0^\infty \left[g(\mathbf{x} - s\boldsymbol{\Omega}, \boldsymbol{\Omega}) + \frac{Q(\mathbf{x} - s\boldsymbol{\Omega})}{4\pi} \right] p(s) ds. \quad (5.7)$$

Operating on this result by $c \int_{4\pi} P(\boldsymbol{\Omega} \cdot \boldsymbol{\Omega}') (\cdot) d\Omega'$, we obtain

$$g(\mathbf{x}, \boldsymbol{\Omega}) = c \int_{4\pi} P(\boldsymbol{\Omega} \cdot \boldsymbol{\Omega}') \int_0^\infty \left[g(\mathbf{x} - s\boldsymbol{\Omega}', \boldsymbol{\Omega}') + \frac{Q(\mathbf{x} - s\boldsymbol{\Omega}')}{4\pi} \right] p(s) ds d\Omega'. \quad (5.8)$$

Now we make the change of spatial variables from the 3-D spherical $(s, \boldsymbol{\Omega})$ to the 3-D Cartesian \mathbf{x}' , defined by $\mathbf{x}' = \mathbf{x} - s\boldsymbol{\Omega}'$.

Then

$$s = |\mathbf{x} - \mathbf{x}'|, \quad (5.10a)$$

$$\boldsymbol{\Omega}' = \frac{\mathbf{x} - \mathbf{x}'}{|\mathbf{x} - \mathbf{x}'|}, \quad (5.10b)$$

$$s^2 ds d\Omega' = dV', \quad (5.10c)$$

and Eq. (5.8) can be written as

$$g(\mathbf{x}, \boldsymbol{\Omega}) = c \int \int \int P\left(\boldsymbol{\Omega} \cdot \frac{\mathbf{x} - \mathbf{x}'}{|\mathbf{x} - \mathbf{x}'|}\right) \left[g\left(\mathbf{x}', \frac{\mathbf{x} - \mathbf{x}'}{|\mathbf{x} - \mathbf{x}'|}\right) + \frac{Q(\mathbf{x}')}{4\pi} \frac{p(|\mathbf{x} - \mathbf{x}'|)}{|\mathbf{x} - \mathbf{x}'|^2} \right] dV'. \quad (5.11a)$$

Also, by using the definition (5.2) in Eq. (5.6b), we obtain

$$\Psi(\mathbf{x}, \boldsymbol{\Omega}) = \int_0^\infty \left[g(\mathbf{x} - s\boldsymbol{\Omega}, \boldsymbol{\Omega}) + \frac{Q(\mathbf{x} - s\boldsymbol{\Omega})}{4\pi} \right] e^{-\int_0^s \Sigma_t(s') ds'} ds. \quad (5.11b)$$

Eq. (5.11a) is an integral equation for $g(\mathbf{x}, \boldsymbol{\Omega})$. If this equation is solved for g , then Eq. (5.11b) determines $\Psi(\mathbf{x}, \boldsymbol{\Omega})$.

If scattering is isotropic, then Eqs. (5.3) hold and Eq. (5.11a) reduces to

$$\frac{c}{4\pi} F(\mathbf{x}) = c \int \int \int \frac{1}{4\pi} \left[\frac{c}{4\pi} F(\mathbf{x}') + \frac{Q(\mathbf{x}')}{4\pi} \right] \frac{p(|\mathbf{x} - \mathbf{x}'|)}{|\mathbf{x} - \mathbf{x}'|^2} dV'$$

or

$$F(\mathbf{x}) = \int \int \int [cF(\mathbf{x}') + Q(\mathbf{x}')] \frac{p(|\mathbf{x} - \mathbf{x}'|)}{4\pi|\mathbf{x} - \mathbf{x}'|^2} dV'. \quad (5.12a)$$

Also, using Eq. (5.3a), we write Eq. (5.11b) as

$$\Psi(\mathbf{x}, \boldsymbol{\Omega}) = \frac{1}{4\pi} \int_0^\infty [cF(\mathbf{x} - s\boldsymbol{\Omega}) + Q(\mathbf{x} - s\boldsymbol{\Omega})] e^{-\int_0^s \Sigma_t(s') ds'} ds. \quad (5.12b)$$

Operating on this equation by $\int_{4\pi} (\cdot) d\Omega$ and using Eq. (5.9) and (5.10), we obtain

$$\Phi(\mathbf{x}) = \int \int \int [cF(\mathbf{x}') + Q(\mathbf{x}')] \frac{e^{-\int_0^{|\mathbf{x} - \mathbf{x}'|} \Sigma_t(s') ds'}}{4\pi|\mathbf{x} - \mathbf{x}'|^2} dV', \quad (5.12c)$$

where $\Phi(\mathbf{x})$ is the scalar flux. Eq. (5.12a) is an integral equation for $F(\mathbf{x})$. If it is solved, then $\Psi(\mathbf{x}, \boldsymbol{\Omega})$ is given by Eq. (5.12b) and $\Phi(\mathbf{x})$ is given by Eq. (5.12c).

Finally, if scattering is isotropic and $\Sigma_t(s) = \Sigma_t = \text{constant}$, then by Eqs. (5.1) and (2.6),

$$f(\mathbf{x}, \boldsymbol{\Omega}) = \Sigma_t \int_0^\infty \psi(\mathbf{x}, \boldsymbol{\Omega}, s) ds = \Sigma_t \Psi(\mathbf{x}, \boldsymbol{\Omega}),$$

and thus by Eq. (5.3b),

$$F(\mathbf{x}) = \Sigma_t \int_{4\pi} \Psi(\mathbf{x}, \boldsymbol{\Omega}') d\Omega' = \Sigma_t \Phi(\mathbf{x}).$$

Also, Eq. (4.6) gives:

$$p(s) = \Sigma_t e^{-\Sigma_t s}.$$

Using the previous two results in Eqs. (5.12c) and (5.12b), we obtain

$$\Phi(\mathbf{x}) = \int \int \int [\Sigma_s \Phi(\mathbf{x}') + Q(\mathbf{x}')] \frac{e^{-\Sigma_t |\mathbf{x} - \mathbf{x}'|}}{4\pi|\mathbf{x} - \mathbf{x}'|^2} dV' \quad (5.13a)$$

and

$$\Psi(\mathbf{x}, \boldsymbol{\Omega}) = \frac{1}{4\pi} \int_0^\infty [\Sigma_s \Phi(\mathbf{x} - s\boldsymbol{\Omega}) + Q(\mathbf{x} - s\boldsymbol{\Omega})] e^{-\Sigma_t s} ds. \quad (5.13b)$$

Eq. (5.13a) is the classic integral transport equation for the scalar flux $\Phi(\mathbf{x})$, and Eq. (5.13b) is the classic expression for the angular flux $\Psi(\mathbf{x}, \boldsymbol{\Omega})$ in terms of $\Phi(\mathbf{x})$.

Thus, for general anisotropic scattering, an integral equation formulation of the GLBE [Eqs. (5.11)] can be obtained which does not contain the path length variable s as an independent variable. If scattering is isotropic, then the classic scalar flux can be obtained using Eqs. (5.12a) and (5.12c), in which the direction variable $\boldsymbol{\Omega}$ also does not occur as an independent variable. Finally, if scattering is isotropic and $\Sigma_t(s) = \Sigma_t = \text{constant}$, then—as they must—these integral equations reduce to the classic integral equation (5.13a) for the scalar flux.

6. Asymptotic diffusion limit of the GLBE

To begin this discussion, we must first consider the Legendre-polynomial expansion of the distribution function $P(\boldsymbol{\Omega} \cdot \boldsymbol{\Omega}') = P(\mu_0)$ defined by Eq. (2.1f)

$$P(\mu_0) = \sum_{n=0}^{\infty} \frac{2n+1}{4\pi} a_n P_n(\mu_0), \quad (6.1)$$

where $a_0 = 1$ and $a_1 = \bar{\mu}_0 = \text{mean scattering cosine}$. We define $P^*(\mu_0)$ by

$$P^*(\mu_0) = cP(\mu_0) + \frac{1-c}{4\pi}, \quad (6.2)$$

which has the Legendre polynomial expansion:

$$P^*(\mu_0) = \sum_{n=0}^{\infty} \frac{2n+1}{4\pi} a_n^* P_n(\mu_0), \quad (6.3a)$$

$$a_n^* = \begin{cases} 1, & n=0, \\ ca_n, & n \geq 1. \end{cases} \quad (6.3b)$$

Using previous work [14–18] as a guide, we scale $\Sigma_t = O(1)$, $1-c = O(\varepsilon^2)$, $Q = O(\varepsilon^2)$, $P^*(\mu_0)$ is independent of ε , $\partial\psi/\partial s = O(1)$, and $\boldsymbol{\Omega} \cdot \nabla\psi = O(\varepsilon)$, with $\varepsilon \ll 1$. Eqs. (2.3) and (6.2) yield

$$\begin{aligned} \frac{\partial\psi}{\partial s}(\mathbf{x}, \boldsymbol{\Omega}, s) + \varepsilon \boldsymbol{\Omega} \cdot \nabla\psi(\mathbf{x}, \boldsymbol{\Omega}, s) + \Sigma_t(s)\psi(\mathbf{x}, \boldsymbol{\Omega}, s) \\ = \delta(s) \int_{4\pi} \int_0^\infty \left[P^*(\boldsymbol{\Omega} \cdot \boldsymbol{\Omega}') - \varepsilon^2 \frac{1-c}{4\pi} \right] \Sigma_t(s') \psi(\mathbf{x}, \boldsymbol{\Omega}', s') ds' d\Omega' \\ + \varepsilon^2 \delta(s) \frac{Q(\mathbf{x})}{4\pi}. \end{aligned} \quad (6.4)$$

The scaling in this equation implies the following:

1. The *large* $[O(1)]$ terms describe neutron scattering. The transport process is dominated by scattering, with the source rate, the absorption rate, and the leakage rates being asymptotically smaller. Also, the length scale for the problem is chosen so that a unit of length is comparable to a typical mean free path, and the system is many mean free paths thick.
2. The leakage $(\mathbf{\Omega} \cdot \nabla \psi)$ term is *small* $[O(\varepsilon)]$. Thus, the angular flux ψ varies a small $[O(\varepsilon)]$ amount over the distance of one mean free path.
3. The absorption term $1-c = \Sigma_a/\Sigma_t$ and the source term Q are *smaller* $[O(\varepsilon^2)]$, and are balanced in such a way that the infinite medium solution

$$\psi = \frac{Q}{4\pi\Sigma_a},$$

which holds when the source and cross sections are constant, is $O(1)$.

4. Because scattering is anisotropic, it is possible to scale the constants a_n in Eq. (6.1) in other ways with respect to ε . However, the scaling defined in Eq. (6.3) has the virtue of being one of the simplest possible—only the $n=0$ constant a_0 is “stretched” asymptotically; the higher-order ($n \geq 1$) terms are not stretched. Also, when this scaling is applied to a standard linear Boltzmann equation, one obtains the same diffusion equation that is obtained from the standard P_1 or spherical harmonics approximation.

The asymptotic derivation of diffusion approximations from the Boltzmann transport equation has been a topic of study over many years. We refer the reader to Refs. [14–18] as a representative sampling of the literature.

To proceed with the asymptotic analysis of Eq. (6.4), we use Eq. (4.12) to define $\Psi(\mathbf{x}, \mathbf{\Omega}, s)$ by

$$\psi(\mathbf{x}, \mathbf{\Omega}, s) \equiv \Psi(\mathbf{x}, \mathbf{\Omega}, s)\chi(s) = \Psi(\mathbf{x}, \mathbf{\Omega}, s) \frac{e^{-\int_0^s \Sigma_t(s') ds'}}{\langle s \rangle}. \quad (6.5)$$

Then Eq. (6.4) for $\psi(\mathbf{x}, \mathbf{\Omega}, s)$ becomes the following equation for $\Psi(\mathbf{x}, \mathbf{\Omega}, s)$:

$$\begin{aligned} \frac{\partial \Psi}{\partial s}(\mathbf{x}, \mathbf{\Omega}, s) + \varepsilon \mathbf{\Omega} \cdot \nabla \Psi(\mathbf{x}, \mathbf{\Omega}, s) \\ = \delta(s) \int_{4\pi} \int_0^\infty \left[P^*(\mathbf{\Omega} \cdot \mathbf{\Omega}') - \varepsilon^2 \frac{1-c}{4\pi} \right] p(s') \Psi(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega' \\ + \varepsilon^2 \delta(s) \langle s \rangle \frac{Q(\mathbf{x})}{4\pi}. \end{aligned} \quad (6.6)$$

This equation is mathematically equivalent to the following two coupled equations:

$$\frac{\partial \Psi}{\partial s}(\mathbf{x}, \mathbf{\Omega}, s) + \varepsilon \mathbf{\Omega} \cdot \nabla \Psi(\mathbf{x}, \mathbf{\Omega}, s) = 0, \quad s > 0, \quad (6.7a)$$

$$\begin{aligned} \Psi(\mathbf{x}, \mathbf{\Omega}, 0) = \int_{4\pi} \left[P^*(\mathbf{\Omega} \cdot \mathbf{\Omega}') - \varepsilon^2 \frac{1-c}{4\pi} \right] \int_0^\infty p(s') \Psi(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega' \\ + \varepsilon^2 \langle s \rangle \frac{Q(\mathbf{x})}{4\pi}, \end{aligned} \quad (6.7b)$$

where (as before) we have defined $\Psi(\mathbf{x}, \mathbf{\Omega}, 0) = \Psi(\mathbf{x}, \mathbf{\Omega}, 0^+)$. Integrating Eq. (6.7a) over $0 < s' < s$, we obtain

$$\begin{aligned} \Psi(\mathbf{x}, \mathbf{\Omega}, s) &= \Psi(\mathbf{x}, \mathbf{\Omega}, 0) - \varepsilon \mathbf{\Omega} \cdot \nabla \int_0^s \Psi(\mathbf{x}, \mathbf{\Omega}, s') ds' \\ &= \int_{4\pi} \left[P^*(\mathbf{\Omega} \cdot \mathbf{\Omega}') - \varepsilon^2 \frac{1-c}{4\pi} \right] \int_0^\infty p(s') \Psi(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega' \\ &\quad + \varepsilon^2 \langle s \rangle \frac{Q(\mathbf{x})}{4\pi} - \varepsilon \mathbf{\Omega} \cdot \nabla \int_0^s \Psi(\mathbf{x}, \mathbf{\Omega}, s') ds'. \end{aligned}$$

Introducing into this equation the ansatz

$$\Psi(\mathbf{x}, \mathbf{\Omega}, s) = \sum_{n=0}^\infty \varepsilon^n \Psi^{(n)}(\mathbf{x}, \mathbf{\Omega}, s)$$

and equating the coefficients of different powers of ε , we obtain for $n \geq 0$:

$$\begin{aligned} \Psi^{(n)}(\mathbf{x}, \mathbf{\Omega}, s) &= \int_{4\pi} P^*(\mathbf{\Omega} \cdot \mathbf{\Omega}') \int_0^\infty p(s') \Psi^{(n)}(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega' \\ &\quad - \mathbf{\Omega} \cdot \nabla \int_0^s \Psi^{(n-1)}(\mathbf{x}, \mathbf{\Omega}, s') ds' \\ &\quad - \frac{1-c}{4\pi} \int_{4\pi} \int_0^\infty p(s') \Psi^{(n-2)}(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega' \\ &\quad + \delta_{n,2} \langle s \rangle \frac{Q(\mathbf{x})}{4\pi}, \end{aligned} \quad (6.8)$$

with $\Psi^{(-1)} = \Psi^{(-2)} = 0$. We now solve these equations recursively, first for $n=0$, then $n=1$, etc. In doing this, we use the Legendre polynomial expansion (6.3) of $P^*(\mu_0)$.

Eq. (6.8) with $n=0$ is

$$\Psi^{(0)}(\mathbf{x}, \mathbf{\Omega}, s) = \int_{4\pi} P^*(\mathbf{\Omega} \cdot \mathbf{\Omega}') \int_0^\infty p(s') \Psi^{(0)}(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega'.$$

The general solution of this equation is

$$\Psi^{(0)}(\mathbf{x}, \mathbf{\Omega}, s) = \frac{\Phi^{(0)}(\mathbf{x})}{4\pi}, \quad (6.9)$$

where $\Phi^{(0)}(\mathbf{x})$ is, at this point, undetermined.

Next, Eq. (6.8) with $n=1$ is

$$\begin{aligned} \Psi^{(1)}(\mathbf{x}, \mathbf{\Omega}, s) &= \int_{4\pi} P^*(\mathbf{\Omega} \cdot \mathbf{\Omega}') \int_0^\infty p(s') \Psi^{(1)}(\mathbf{x}, \mathbf{\Omega}', s') ds' d\Omega' \\ &\quad - \frac{s}{4\pi} \mathbf{\Omega} \cdot \nabla \Phi^{(0)}(\mathbf{x}). \end{aligned} \quad (6.10)$$

This equation has a particular solution of the following form:

$$\Psi_{part}^{(1)}(\mathbf{x}, \mathbf{\Omega}, s) = \frac{g(s)}{4\pi} \mathbf{\Omega} \cdot \nabla \Phi^{(0)}(\mathbf{x}). \quad (6.11a)$$

Introducing this form into Eq. (6.10) and using $a_1^* = ca_1 = c\bar{\mu}_0$, we obtain the following equation for $g(s)$:

$$g(s) = c\bar{\mu}_0 \int_0^\infty p(s') g(s') ds' - s,$$

which has the solution:

$$g(s) = - \left(s + \frac{c\bar{\mu}_0}{1-c\bar{\mu}_0} \langle s \rangle \right), \quad (6.11b)$$

where $\langle s \rangle =$ mean free path is defined by Eq. (4.11). Hence, the general solution of Eq. (6.10) is

$$\Psi^{(1)}(\mathbf{x}, \mathbf{\Omega}, s) = \frac{\Phi^{(1)}(\mathbf{x})}{4\pi} - \frac{1}{4\pi} \left(s + \frac{c\bar{\mu}_0}{1-c\bar{\mu}_0} \langle s \rangle \right) \mathbf{\Omega} \cdot \nabla \Phi^{(0)}(\mathbf{x}), \quad (6.12)$$

where $\Phi^{(1)}(\mathbf{x})$ is undetermined.

We next consider Eq. (6.8) with $n=2$. This equation has a solvability condition, which is obtained by operating by $\int_{4\pi} \int_0^\infty p(s)(\cdot) ds d\Omega$. Using Eqs. (6.12) and (6.9) to obtain

$$\int_0^s \Psi^{(1)}(\mathbf{x}, \Omega, s') ds' = s \frac{\Phi^{(1)}(\mathbf{x})}{4\pi} - \frac{1}{4\pi} \left(\frac{s^2}{2} + \frac{c\bar{\mu}_0}{1-c\bar{\mu}_0} \langle s \rangle s \right) \Omega \cdot \nabla \Phi^{(0)}(\mathbf{x})$$

and

$$\int_{4\pi} P^*(\Omega \cdot \Omega) \int_0^\infty p(s') \Psi^{(0)}(\mathbf{x}, \Omega', s') ds' d\Omega' = \frac{\Phi^{(0)}(\mathbf{x})}{4\pi},$$

the solvability condition becomes

$$0 = \frac{1}{4\pi} \int_{4\pi} \int_0^\infty p(s) \left(\frac{s^2}{2} + \frac{c\bar{\mu}_0}{1-c\bar{\mu}_0} \langle s \rangle s \right) (\Omega \cdot \nabla)^2 \Phi^{(0)}(\mathbf{x}) ds d\Omega - \frac{1-c}{4\pi} \int_{4\pi} \int_0^\infty p(s) \Phi^{(0)}(\mathbf{x}) ds d\Omega + \langle s \rangle Q(\mathbf{x}).$$

Evaluating the angular integrals and rearranging, we obtain the following diffusion equation for $\Phi^{(0)}(\mathbf{x})$:

$$-\frac{1}{3} \left(\frac{\langle s^2 \rangle}{2 \langle s \rangle} + \frac{c\bar{\mu}_0}{1-c\bar{\mu}_0} \langle s \rangle \right) \nabla^2 \Phi^{(0)}(\mathbf{x}) + \frac{1-c}{\langle s \rangle} \Phi^{(0)}(\mathbf{x}) = Q(\mathbf{x}). \quad (6.13)$$

To summarize: the solution $\psi(\mathbf{x}, \Omega, s)$ of Eq. (6.4) satisfies

$$\psi(\mathbf{x}, \Omega, s) = \frac{\Phi^{(0)}(\mathbf{x}) e^{-\int_0^s \Sigma_t(\mathbf{x}, s') ds'}}{4\pi \langle s \rangle} + O(\varepsilon), \quad (6.14)$$

where $\Phi^{(0)}(\mathbf{x})$ satisfies Eq. (6.13). Also, integrating Eq. (6.14) over $0 < s < \infty$ and $\Omega \in 4\pi$, and using Eq. (4.11), we obtain to leading order:

$$\Phi^{(0)}(\mathbf{x}) = \int_{4\pi} \left(\int_0^\infty \psi(\mathbf{x}, \Omega, s) ds \right) d\Omega = \int_{4\pi} \psi(\mathbf{x}, \Omega) d\Omega, \quad (6.15)$$

where [see Eq. (2.6)] $\psi(\mathbf{x}, \Omega)$ is the classic angular flux. Thus, the solution $\Phi^{(0)}(\mathbf{x})$ of Eq. (6.13) is the classic scalar flux.

Eq. (6.13) is, of course, a much simpler equation than the generalized linear Boltzmann equation (2.3). In Eq. (6.13), the angular and path length variables are absent, and the non-exponential path length distribution manifests itself as a non-classical definition of the diffusion coefficient. If the path length distribution is exponential, then by Eq. (4.6), $p(s) = \Sigma_t e^{-\Sigma_t s}$, which gives $\langle s \rangle = 1/\Sigma_t$ and $\langle s^2 \rangle = 2/\Sigma_t^2$; and then, as it must, Eq. (6.15) reduces to the classic diffusion equation:

$$-\frac{1}{3\Sigma_t(1-c\bar{\mu}_0)} \nabla^2 \Phi^{(0)}(\mathbf{x}) + \Sigma_t(1-c)\Phi^{(0)}(\mathbf{x}) = Q(\mathbf{x}). \quad (6.16)$$

The “non-classical” diffusion coefficient in Eq. (6.13):

$$D = \frac{(1-c\bar{\mu}_0)\langle s^2 \rangle + 2c\bar{\mu}_0\langle s \rangle^2}{6(1-c\bar{\mu}_0)\langle s \rangle} \quad (6.17)$$

is obviously positive for $0 \leq \bar{\mu}_0 \leq 1$, but showing that $D \geq 0$ for $-1 \leq \bar{\mu}_0 < 0$ requires more effort. To do this, we use the Cauchy–Schwartz inequality to get

$$\begin{aligned} \langle s \rangle^2 &= \left(\int_0^\infty sp(s) ds \right)^2 \\ &= \left(\int_0^\infty [sp^{1/2}(s)][p^{1/2}(s)] ds \right)^2 \end{aligned}$$

$$\leq \left(\int_0^\infty s^2 p(s) ds \right) \left(\int_0^\infty p(s) ds \right) = \langle s^2 \rangle, \quad (6.18)$$

with equality holding only when

$$s^2 p(s) = (\text{constant}) p(s), \quad 0 < s < \infty,$$

and this holds only when

$$p(s) = \delta(s-s_0), \quad (6.19)$$

where δ is the familiar delta function and s_0 is any positive constant. (In this situation, particles travel a fixed distance s_0 between collisions.) Since $\bar{\mu}_0 < 0$, the inequality (6.18) implies

$$(\bar{\mu}_0)\langle s \rangle^2 \geq (\bar{\mu}_0)\langle s^2 \rangle,$$

so Eq. (6.17) gives

$$D \geq \frac{(1-c\bar{\mu}_0)\langle s^2 \rangle + 2c\bar{\mu}_0\langle s^2 \rangle}{6(1-c\bar{\mu}_0)\langle s \rangle} = \left(\frac{1+c\bar{\mu}_0}{1-c\bar{\mu}_0} \right) \frac{\langle s^2 \rangle}{6\langle s \rangle} \geq 0. \quad (6.20)$$

Interestingly, D limits to 0 when the following three conditions are met:

1. The inequality (6.18) becomes equality. [This happens only when $p(s) = \delta(s-s_0)$.]
2. $\bar{\mu}_0 \rightarrow -1$. [This happens when particles only back-scatter 180° .]
3. $c \rightarrow 1$. [This is already implied by the asymptotic analysis, which requires $1-c = O(\varepsilon^2)$.]

The first two conditions imply that particle histories consist of simple “bouncing” back and forth between two points that lie a fixed distance s_0 apart. Thus, particles become “trapped”—they cannot diffuse away from their point of birth, and the result $D=0$ is appropriate.

To summarize: for all $-1 \leq \bar{\mu}_0 \leq 1$ and $0 \leq c \leq 1$, the diffusion coefficient in Eq. (6.13) is non-negative, and in the one circumstance in which $D=0$, this result is physically correct.

We also note that if $p(s)$ decays algebraically as $s \rightarrow \infty$ as

$$p(s) \geq \frac{\text{constant}}{s^3} \quad \text{for } s \gg 1, \quad (6.21a)$$

then

$$\langle s^2 \rangle = \int_0^\infty s^2 p(s) ds = \infty. \quad (6.21b)$$

In this case, the asymptotic diffusion approximation developed above is invalid, because the asymptotic analysis tacitly requires $\langle s \rangle$ and $\langle s^2 \rangle$ to both be finite.

Physically, the asymptotic diffusion theory becomes invalid when $\langle s^2 \rangle = \infty$ because particles will travel large distances between collisions too often. When $\langle s^2 \rangle < \infty$, the probability that a particle will scatter between two distant points is sufficiently small that the diffusion process can occur. However, when $\langle s^2 \rangle = \infty$, sufficiently long flight paths will occur sufficiently often that the diffusion description developed here becomes invalid. It seems unlikely that the asymptotic analysis can be generalized to develop a standard diffusion description when $\langle s^2 \rangle = \infty$. This is because the occurrence of “long” flight paths between collisions (over distances in which

the flux can vary appreciably) is inherently a characteristic of transport, not diffusion processes.

We note that in [10], the cases $\langle s^2 \rangle < \infty$ and $\langle s^2 \rangle = \infty$ are, respectively, called *standard diffusion* and *anomalous diffusion*. Our analysis indicates that in the context of the GLBE, standard diffusion is an asymptotic approximation, while anomalous diffusion is not.

7. Numerical results

To test the theory developed above, we consider a 2-D random system that models a 3-D pebble-bed nuclear reactor core. In the real 3-D problem, the reactor core is randomly filled with about 500,000 roughly tennis-ball-sized “pebbles” (spheres), each composed of graphite moderator containing several thousand microspheres of nuclear fuel, surrounded by an outer ceramic shell. In the model 2-D problem considered here,

- Neutrons are monoenergetic, travel only in the (x,y) -plane [19], and scatter isotropically.
- Each spherical “pebble” is modeled as a homogeneous circular disc of radius r in the (x,y) -plane.
- The reactor core is modeled as a square box with side of length L , inside which the circular fuel discs are randomly placed using the “ballistic deposition method” described below.

Thus, the term “2-D” describes both the geometry and the particle transport of the model. We have written a Monte Carlo computer code that constructs random realizations of the 2-D core, and a second Monte Carlo code that performs 2-D neutron transport inside the heterogeneous core. In this manner, we have performed

2-D Monte Carlo particle transport simulations in the random cores and compared the resulting “exact” results to approximate results obtained from the GLBE and the atomic mix models. In the following, we give details of generating the random realizations of the system and the subsequent numerical results.

First, the reactor core is randomly filled using an adaptation of the “ballistic deposition model” presented in [20]. In this model, each disc is released at a random point above the box. The disc follows a steepest descent trajectory until it reaches a position that is stable under gravity, in which case it is frozen in place—once the position of the disc is locked, it can no longer move. Fig. 2 contains four snapshots of a packing performed with this process: (A) disc 1 descends vertically to the bottom of the box, where it becomes locked in place; (B) disc 2 descends until it touches the frozen disc 1; then it rolls down disc 1 until it touches the bottom of the box, where it is locked in place; (C) disc 3 descends vertically to the bottom of the box; (D) disc 4 descends until it touches the frozen disc 3; then it rolls down disc 3 until it touches disc 2; since discs 2 and 3 cannot move, disc 4 is stable and is locked in place.

Since frozen discs cannot move, the inclusion of a new disc will not cause the system to rearrange; thus cascading events (“avalanches”) will not occur. Also, no velocity or friction coefficients are taken into account; the only restriction is that a disc can never, at any point of its trajectory, overlap the boundaries of the box or another disc. Once a disc has reached its final stable position, it is frozen in place and a new disc is released; this process is repeated until the box is filled. An example of random piling with $L=40r$ is shown in Fig. 3.

Given a realization of the model core, we selected the disc closest to the center of the system to be the one in which particles are born (i.e., we focus on the transport of

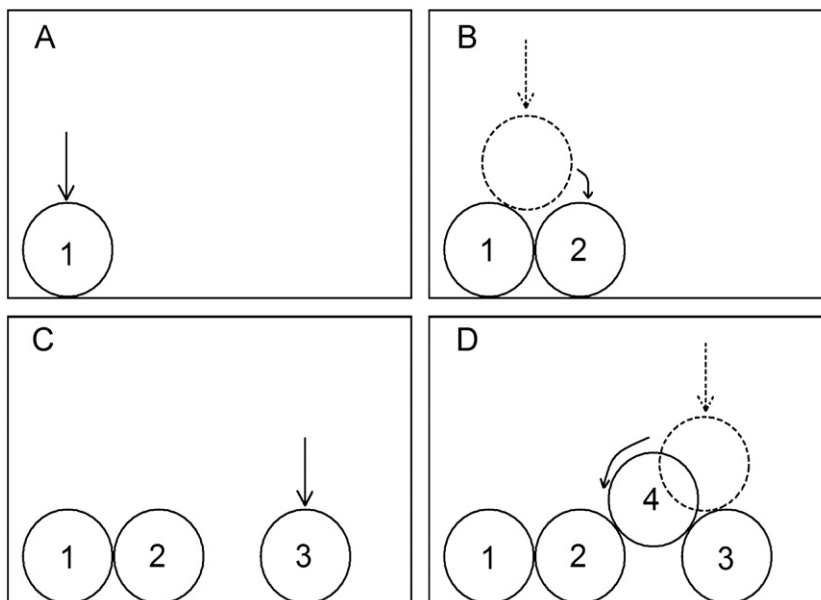


Fig. 2. Steps of a random 2-D packing process.

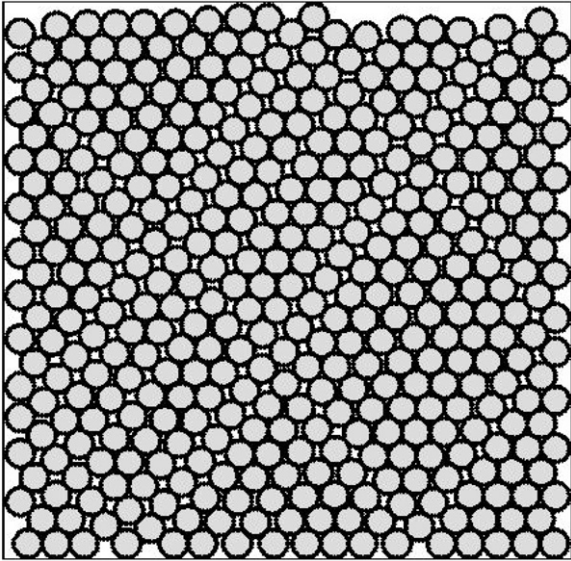


Fig. 3. Example of a 2-D random structure in a system with side $L = 40r$.

Table 1
2-D parameters for discs with radius r (Problem 1).

$2r\Sigma_{t,1}$	$2r\Sigma_{s,1}$	$2r\Sigma_{a,1}$	$c = \Sigma_{s,1}/\Sigma_{t,1}$	$P_1(\Omega \cdot \Omega')$
1.0	0.99	0.01	0.99	$1/2\pi$

particles generated by a single fuel disc). The particles' subsequent histories within the system are determined by our Monte Carlo transport code, which numerically calculates $\langle s \rangle$ = mean distance to collision, $\langle s^2 \rangle$ = mean squared distance to collision, and $\langle x^2 + y^2 \rangle$ = the mean-squared distance of a particle from its point of birth.

For the first of two problems that we simulated, we took the background material 2 in which the discs are piled to be a vacuum, with zero cross sections; the cross sections and other parameters for material 1 (inside the discs) are given in Table 1.

For each realization of the random system, we calculated the histories of 20,000 particles; the statistical error in each realization was found to be (with 97.5% confidence) less than 0.052% for all values of $\langle s \rangle$, less than 0.115% for all values of $\langle s^2 \rangle$, and less than 0.089% for all values of $\langle x^2 + y^2 \rangle$. We constructed 1024 different random packings in the 2-D system with $L = 600r$ (summing to a total of 20,480,000 particles' histories); the average Monte Carlo results and the statistical error bounds (with 95% confidence) are given in Table 2.

Also, the average packing fraction (the mean fraction of the area of the box occupied by fuel discs) was calculated as $f = 0.817$, with estimated standard deviation 0.00133.

Next, we consider the following 2-D diffusion equation, defined for an infinite 2-D planar system with a point source at the origin isotropically emitting Q particles

Table 2
Monte Carlo transport results (Problem 1).

	$\langle s \rangle / 2r$	$\langle s^2 \rangle / 4r^2$	$\langle x^2 + y^2 \rangle / 4r^2$
Ensemble average	1.22151	3.04209	303.517
Relative statistical error (%)	0.070	0.153	0.134

Table 3
Atomic Mix and GLBE Diffusion Coefficients.

	D	Σ_a
GLBE	$\frac{\langle s^2 \rangle}{4\langle s \rangle}$	$\frac{1-c}{\langle s \rangle}$
Atomic Mix	$\frac{1}{2f\Sigma_{t,1}}$	$f\Sigma_{a,1}$

per second:

$$-D \left(\frac{\partial^2}{\partial x^2} \Phi(x,y) + \frac{\partial^2}{\partial y^2} \Phi(x,y) \right) + \Sigma_a \Phi(x,y) = Q \delta(x) \delta(y). \quad (7.1)$$

For problems in which $\Sigma_a \ll \Sigma_s$, the GLBE and the atomic mix transport equation are both modeled by this diffusion equation, although with different prescriptions for D and Σ_a (see Table 3).

Here the GLBE expressions come from Eq. (6.13), and the atomic mix expressions come from homogenizing material 1 with the void material 2, with the volume fraction of material 1 taken as $f=0.817$. The factor $1/2$ (rather than $1/3$) in the atomic mix diffusion coefficient occurs because diffusion occurs in a 2-D plane.

Recalling that $\Phi(\mathbf{x}) \rightarrow 0$ and $\nabla \Phi(\mathbf{x}) \rightarrow 0$ as $|\mathbf{x}| \rightarrow \infty$, we can manipulate Eq. (7.1) to derive an exact (diffusion) formula for the mean square distance that particles travel from their point of birth. To do this, we multiply Eq. (7.1) by $x^2 + y^2$ and integrate over $-\infty < x, y < \infty$ to get

$$-D \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} (x^2 + y^2) \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \Phi(x,y) dx dy + \Sigma_a \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \Phi(x,y) dx dy = 0. \quad (7.2)$$

However, integrating by parts twice gives

$$\int_{-\infty}^{\infty} (x^2 + y^2) \frac{\partial^2 \Phi}{\partial x^2} dx = -2 \int_{-\infty}^{\infty} x \frac{\partial \Phi}{\partial x} dx = 2 \int_{-\infty}^{\infty} \Phi dx,$$

so Eq. (7.2) yields

$$\langle x^2 + y^2 \rangle = \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} (x^2 + y^2) \Phi(x,y) dx dy}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \Phi(x,y) dx dy} = \frac{4D}{\Sigma_a}. \quad (7.3)$$

Combining this with results from Table 2, we obtain

$$\langle x^2 + y^2 \rangle = \begin{cases} \frac{\langle s^2 \rangle}{1-c} & \text{GLBE,} \\ \frac{2}{f^2 \Sigma_{t,1} \Sigma_{a,1}} & \text{Atomic Mix.} \end{cases} \quad (7.4)$$

Not surprisingly, the GLBE and Atomic Mix approximations predict different mean-squared distances of particles from their point of birth. To test these predictions, we compare them to the "exact" values obtained

from the 2-D Monte Carlo transport simulations and given in Table 2.

Using Eq. (7.4), Table 2 for $\langle s^2 \rangle$; Table 1 for $\Sigma_{t,1}, \Sigma_{a,1}$, and c ; and $f=0.817$, we obtain the numerical estimates of the root mean square (RMS) distance of particles from their point of birth shown in Table 4.

Here the Monte Carlo error is estimated using the Central Limit Theorem with 95.0% confidence, and the GLBE and Atomic Mix errors are the percent relative differences between these RMS distance estimates and the Monte Carlo estimate.

The estimated Monte Carlo error (0.082%) is smaller than the estimated GLBE and Atomic Mix errors. Thus, most of the errors in the GLBE and Atomic Mix estimates are modeling errors caused by the fact that the GLBE and Atomic Mix models do not perfectly reproduce the transport physics. Table 4 shows that for the considered problem, the GLBE and Atomic Mix errors are both small (less than 0.6%), but the GLBE error is about five times smaller than the Atomic Mix error.

To test the GLBE results on a more difficult problem (a less diffusive problem which is further away from the atomic mix limit), we consider in Problem 2 exactly the same geometric situation as in Problem 1, except that the cross sections are all multiplied by a factor of 4 (see Table 5).

Just as before, for each realization of this random system we calculated the histories of 20,000 particles; the statistical error in each realization was now found to be (with 97.5% confidence) less than 0.039% for all values of $\langle s \rangle$, less than 0.086% for all values of $\langle s^2 \rangle$, and less than 0.063% for all values of $\langle x^2 + y^2 \rangle$. As before, we constructed 1024 different random packings in the 2-D system with $L = 600r$ (summing to a total of 20,480,000 particle histories); the average Monte Carlo results and the statistical error bounds (with 95% confidence) are given in Table 6.

For Problem 2, the average packing fraction was calculated to be $f = 0.817$, with the standard deviation 0.00133. (Not surprisingly, these numbers are identical to those calculated for Problem 1.) Using the above results, we calculate the RMS distance that particles travel from their point of birth; these are given in Table 7.

Table 4
RMS distance from point of birth (Problem 1).

	$\langle x^2 + y^2 \rangle^{1/2} / 2r$	% Relative error
Monte Carlo	17.422	0.082
GLBE	17.442	0.115
Atomic Mix	17.318	0.597

Table 5
2-D parameters for discs with radius r (Problem 2).

$2r\Sigma_{t,1}$	$2r\Sigma_{s,1}$	$2r\Sigma_{a,1}$	$c = \Sigma_{s,1} / \Sigma_{t,1}$	$P_1(\Omega \cdot \Omega')$
4.0	3.96	0.04	0.99	$1/2\pi$

Table 6
Monte Carlo transport results (Problem 2).

	$\langle s \rangle / 2r$	$\langle s^2 \rangle / 4r^2$	$\langle x^2 + y^2 \rangle / 4r^2$
Ensemble average	0.30492	0.20174	19.733
Relative statistical error (%)	0.067	0.157	0.129

Table 7
RMS distance from point of birth (Problem 2).

	$\langle x^2 + y^2 \rangle^{1/2} / 2r$	% Relative error
Monte Carlo	4.442	0.0645
GLBE	4.491	1.103
Atomic Mix	4.327	2.589

As before, we estimated the Monte Carlo error in this table using the Central Limit Theorem with 95.0% confidence, and we defined the GLBE and Atomic Mix errors to be the percent relative differences between these RMS distance estimates and the Monte Carlo estimate. The estimated Monte Carlo error (0.0645%) is again smaller than the estimated GLBE and Atomic Mix errors. Therefore, most of the errors in the GLBE and Atomic Mix estimates are again modeling errors. Table 7 shows that for Problem 2, the GLBE and Atomic Mix errors are still small (now less than 2.6%), but larger than for Problem 1. Problems 1 and 2 differ in the following two ways:

1. Because the solid material cross sections in Problem 2 are all larger than those in Problem 1 by a factor of 4, Problem 2 is further away from the atomic mix limit. This should cause the Atomic Mix solution to be less accurate in Problem 2 than in Problem 1.
2. Because the diffusion length $L = 1 / \sqrt{3\Sigma_a\Sigma_t}$ is smaller in Problem 2 by a factor of 4 than in Problem 1, Problem 2 is less “diffusive,” i.e. it is one in which a diffusion approximation is less likely to be able to accurately model the transport physics. [To maintain the diffusive character of Problem 1 in Problem 2, it would have been necessary to *decrease* (not increase) Σ_a by a factor of 4. We did not do this because it would have drastically increased the run time of our Monte Carlo simulations.] This reduction in the diffusive character of Problem 2 should cause our Atomic Mix and GLBE results to both degrade, since both are based on a diffusion approximation.

The GLBE solution has about 1/5 the error of the Atomic Mix solution in Problem 1, and about 1/2 the error of the Atomic Mix solution in Problem 2. In both problems, the Atomic Mix error is small (less than 3%), but the GLBE error is smaller. Due to the expense of generating numerical solutions, we did not test other problems.

Nonetheless, we can confidently assert the following: the 2-D diffusion model of a pebble bed reactor considered here is one for which the Atomic Mix result is reasonably accurate, but the GLBE result is more accurate. The probable reason for the increased accuracy

of the GLBE diffusion model is that it uses detailed physics properties of the random system ($\langle s \rangle$ and $\langle s^2 \rangle$) that are not used in the simpler Atomic Mix approximation. For this reason, the GLBE asymptotic diffusion model will likely continue to be more accurate than the Atomic Mix solution for other “diffusive” particle transport problems in random media.

8. Discussion

We have developed a new generalized linear Boltzmann equation (GLBE), which describes particle transport for infinite statistically homogeneous random media in which the ensemble-averaged distribution function $p(s)$ for the path length s between collisions is non-exponential. We have shown that the GLBE (i) can be cast as integral equations in which the path length variable s is absent, and (ii) has an asymptotic diffusion limit, in which s and Ω are both absent. Using Monte Carlo simulations, we have shown that the GLBE models a random, heterogeneous system (a 2-D model of a pebble bed reactor) more accurately than the standard atomic mix approximation; this indicates that the GLBE may be useful for other problems in which the atomic mix approximation is not considered to be sufficiently accurate.

Compared to the standard linear Boltzmann equation, the generalized linear Boltzmann equation contains one extra independent variable (the path length s). Therefore, the GLBE will be much more costly to numerically simulate using deterministic methods than the standard Boltzmann equation. (The standard linear Boltzmann equation is already costly to solve; adding an extra independent variable would only make matters worse.) However, Monte Carlo methods for the GLBE should be only slightly more costly to use than for the standard Boltzmann equation; the only difference is that the distance to collision will be sampled from a non-exponential distribution function. Also, approximate GLBE methods, such as the asymptotic diffusion approximation considered here, can have a phase space with the same dimensionality as approximations for the standard Boltzmann equation.

The GLBE is more costly to simulate than the Atomic Mix approximation. The Atomic Mix approximation only requires that one know the cross sections of the constituent materials and their volume fractions. The GLBE requires much more detailed information—which must be obtained by constructing realizations of the random system and developing an accurate estimate of the ensemble-averaged distribution function for distance to collision.

Nonetheless, because the GLBE method preserves certain statistical properties of the original random system, we believe that it represents a systematically more accurate alternative to the Atomic Mix approximation. It is possible that simplifications to the GLBE equation can be developed that will make the resulting theory less costly to implement. For example, given the values of $\langle s \rangle$ and $\langle s^2 \rangle$, the asymptotic diffusion approximation to the GLBE is no more difficult to

implement than any other diffusion approximation. Also, for the asymptotic diffusion method, it is not necessary to determine the entire distribution function $p(s)$; it is only necessary to determine two of its moments: $\langle s^n \rangle = \int_0^\infty s^n p(s) ds$ for $n=1$ and 2. Other simplifications of the GLBE method presented in this paper that preserve a limited number of moments of $p(s)$ are likely possible.

Generalizations of the present GLBE method are possible as well. For instance, the GLBE equation developed in this paper defines $p(s)$ to be independent of \mathbf{x} and Ω by ensemble-averaging $P(\mathbf{x}, \Omega, s)$ over all \mathbf{x}, Ω , and realizations \mathcal{R} [see Eq. (1.3)]:

$$p(s) = \langle P(\mathbf{x}, \Omega, s) \rangle_{(\mathbf{x}, \Omega, \mathcal{R})}. \quad (8.1)$$

A more accurate result could be obtained by ensemble-averaging over \mathbf{x} and \mathcal{R} but not Ω :

$$p(\Omega, s) = \langle P(\mathbf{x}, \Omega, s) \rangle_{(\mathbf{x}, \mathcal{R})}, \quad (8.2)$$

and a still more accurate result could be obtained by ensemble-averaging over \mathcal{R} but not Ω or \mathbf{x} :

$$p(\mathbf{x}, \Omega, s) = \langle P(\mathbf{x}, \Omega, s) \rangle_{\mathcal{R}}. \quad (8.3)$$

In fact, we have shown in a more complex analysis that by defining $p(\Omega, s)$ as in Eq. (8.2), the asymptotic diffusion limit described in this paper yields a more accurate anisotropic diffusion equation containing a diffusion tensor [21,22]. The rates of diffusion can differ in different directions because the physical structure of the random medium has a systematic asymmetry. For example, in the case of a pebble-bed reactor, the force of gravity always acts in an asymmetric manner, causing a slightly greater rate of diffusion in the vertical direction than in the horizontal direction. This result is not visually obvious in Fig. 3, but it is there nonetheless. (Details of this work will be published elsewhere.)

The present work was originally motivated by recent atmospheric sciences publications [1–10], in which the distribution function $p(s)$ for solar radiation in atmospheric clouds has been experimentally determined to be non-exponential, because of strong correlations between the scattering centers (water droplets) within the clouds. For the methodology developed in this paper to become applicable to these problems, the following generalizations must be made:

- The theory must account for finite clouds, in which the solar radiation enters the system through the boundary.
- The theory must account for statistically heterogeneous clouds, whose ensemble-averaged cross section may be dependent on both space and the direction of flight.
- A sufficiently inexpensive and accurate mathematical model of the GLBE must be developed for practical applications. [The asymptotic diffusion approximation developed in this paper will not be accurate for optically thin clouds, or for parts of clouds in which Eq. (6.21a) holds.]
- The physical mechanisms that generate the correlations between water droplets in clouds should be studied, and the theoretical relationship between these

correlations and the non-exponential distribution function $p(s)$ should be determined.

The first two of these tasks are reasonably straightforward, but the third and fourth may be significantly more difficult. As an initial attempt to address the fourth task, analytic expressions for $p(s)$ should be developed for specified (perhaps model) random systems, in order to better understand how the detailed structure of these systems can affect $p(s)$. These tasks cannot be considered here, but they should be pursued in future work.

Acknowledgments

Edward Larsen wishes to express his gratitude to Professor Alexander Kostinski, who provided strong encouragement for this initial work. Richard Vasques would like to thank CAPES (Coordenação de Aperfeiçoamento de Pessoal de Nível Superior) and the Fulbright Program for financial and administrative support. Both authors would like to thank the two referees for their significant and helpful comments.

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