



**HAL**  
open science

## From particles scale to anomalous or classical convection-diffusion models with path integrals

Catherine Choquet, Marie-Christine Néel

► **To cite this version:**

Catherine Choquet, Marie-Christine Néel. From particles scale to anomalous or classical convection-diffusion models with path integrals. 2013. hal-00842002

**HAL Id: hal-00842002**

**<https://hal.science/hal-00842002>**

Preprint submitted on 5 Jul 2013

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# From particles scale to anomalous or classical convection-diffusion models with path integrals\*

**Catherine Choquet**

Univ. La Rochelle, MIA CNRS EA 3165,  
F-17000 La Rochelle, France  
catherine.choquet@univ-lr.fr

**Marie-Christine Néel**

Univ. d'Avignon et des Pays de Vaucluse, UMR 1114 EMMAH,  
F-84018 Avignon Cedex, France  
mcneel@avignon.inra.fr

**AMS Subject Classification:** 60J70, 60J27, 60J75, 60G22, 82C21.

**Keywords:** fractal mobile-immobile model, fractional Fokker-Planck equation, fractional Feynman-Kac equation, diffusion *versus* sub-diffusion, up-scaling, continuum limit.

## 1 Introduction

The spread of a tracer in a shear flow, in a porous medium or in a spatially disordered set of streamlines, is often analyzed by averaging over microscopic scales. Classically the corresponding mesoscopic model is the advection-diffusion equation. Diffusion is characterized by an evolution in time of the tracer distribution variance proportional to  $t$ . Nevertheless, observations have emphasized the existence of regimes where the variance rather expands as  $t^{2\gamma}$ ,  $\gamma \neq 1/2$  (see *e.g.* the observations in a sand column described in [7]). This non-Fickian phenomenon is referred to as *anomalous diffusion*. In particular, if  $0 < \gamma < 1/2$  the dispersion is slower than diffusion and is thus termed *subdiffusion*. Subdiffusion has been observed for instance during the transport of charge carriers in amorphous semi-conductors, during the propagation of contaminants in groundwater, or in the movement of proteins in intracellular media.

---

\*Research of the authors was partially supported by the GDR MoMaS (PACEN/CNRS, ANDRA, BRGM, CEA, EDF, IRSN) as a part of the project “*Non-Fickian effects and fractional models for diffusion in porous media*”

The present paper is devoted to the rigorous upscaling of some particles displacement model with trapping events to the continuum scale. We focus especially on the transitions between subdiffusive and diffusive models.

A classical approach to go from the particles scale to a continuous description of the displacement consists in representing small scale motions of walkers by means of random processes. After the fundamental works of Brown [8], Einstein [15], Smoluchowski [52] and Perrin [41], literature on the subject is of course very abundant. Recently, a branch of the physical literature focused on non-Fickian settings. Let us give some typical references. A first statistical model yielding subdiffusion is the anti-persistent fractional Brownian motion with Hurst exponent in the range  $(0, 1/2)$  ([34]). Another approach to anomalous behavior consists in representing small scale motions of walkers by means of random processes whose (one-time) probability density function satisfies the Fractional Fokker Planck Equation (FFPE) or the fractal Mobile-Immobile Model (fMIM). FFPEs were derived *e.g.* in [56, 3, 36, 30, 29], using mainly *continuous time random walks* and Laplace transforms. In the present paper, we choose primarily the MIM approach that seems better suited for modeling mass transport in porous media ([49, 4, 38]). Indeed, fMIM's solutions behave as FFPE's ones at late times and as ones of the advection-dispersion equation at early times, in agreement with many heavy-tailed data. Yet we prove that our derivation lets us also recover FFPE.

Basically, MIM-like models considers the global displacement as a succession of Mobile periods (convection/diffusion) and Immobile periods (trapping events due to local –microscopic– heterogeneity, *e.g.* geometrical complexity or sorption sites) [57], [4]. Fractal MIM involves a generalization of Fourier's law that rules the density of Brownian motion.

In the present work, we aim at giving particular emphasis to the following points:

- *The waiting times distribution and its scaling. Anomalous versus classical diffusion.*

A quantity of fundamental importance in passing to the continuum limit in an interrupted process is the distribution of waiting times. One of the first illustrations of this importance was given in [55] by selecting a distribution which induces the same type of variance than the pde's model for the tracer distribution in a central pipe with many stagnant infinite side branches. See also the references therein.

On the one hand, classical diffusion equation has been justified by as-

suming deterministic residence times between jumps (Einstein’s derivation) or exponential distribution of residence times (see for instance [17]). On the other hand recent developments about anomalous diffusion are mainly associated to residence times with infinite mean (Mittag-Leffler residence time distribution, *e.g.* [48, 50]). These two types of result have led to the common idea that distributions with slowly decaying long tails produce anomalous diffusion while the classical diffusion model corresponds to distributions with short tails. In the present paper, we show that it does not always hold true. Actually, we introduce a simple model of geometrical heterogeneity leading to trapping events without characteristic time scale: the survival probability of the trapping asymptotically behaves as a power law (long tails). We then show that anomalous diffusion appears in MIM if and only if there is no scale separation between moving and trapping process. It means in particular that the mesoscale, that is the observation time, is calibrated in view of the magnitude of the sojourn times at the microscale. Furthermore we prove that the anomalous behavior in MIM is transient, without assuming that the microscopic heterogeneity asymptotically disappears as *e.g.* in [13, 5].

- *Path integrals.*

Let  $u$  be a given integrable function. We associate with any path of particle  $x(t)$  in the time interval  $(0, t)$  the random variable  $A$  defined as the functional:

$$A(t) = \int_0^t u(x(s)) ds.$$

Such path integral may represent various “observables” attached to trajectories of tracer particles: occupation time if  $u$  is a step function, advection in turbulent flow if  $u(x) = x$ , average width of an interface if  $u(x) = x^2$ , magnetization phase in NMR if  $u$  is the magnetic field *etc* (see the review [32] for various examples). Assuming Brownian particles, the Brownian functional  $A$  is widely studied. In the fundamental article [25], using Feynman’s path integral method, Kac derived the Schrödinger-like equation governing the distribution of Brownian functionals for any positive function  $u$ .

In the present paper, we extend the Feynman-Kac theory to some non-Brownian settings. We thus also recover the results of [9] without handling with double Laplace transforms. We believe our method more comfortable in view of numerical simulations.

- *Fractal Mobile-Immobile Model versus Fractional Fokker Planck Equation.*

We construct a microscopic random walk model that, thought based on a MIM approach, let us get at the mesoscopic limit both fMIM and FFPE. The key is the choice of the duration of the micro-convective step. Furthermore we prove at the mesoscale that FFPE may be viewed as a limit of fMIM.

Let us now describe the results presented in this work.

The present paper gives an uniform derivation of the appropriate form of the Feynman-Kac equation both in subdiffusive and diffusive settings. Moreover we describe the scalings leading to the appearance and disappearance of anomalous behavior. By the way, let us first focus on the anomalous setting. We state the following result.

**Theorem 1** *If the observation time is such that there is no scale separation between trapping and diffusion, the hydrodynamic limiting behavior of the probability density function (pdf)  $P(x, t, A)$  for a tagged particle to be at point  $x$  at time  $t$  with the value  $A$  for the path integral  $A(t)$  is governed by a non-Fickian equation in the form*

$$\partial_t P - D\partial_{xx}^2 \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P + \Lambda_v \partial_x (v \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P) + u \partial_A P = r, \quad (1)$$

where  $r$  is a source term and operator  $\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma$  is the inverse of some non-local in time mapping:

$$\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma = (\Lambda_v \text{Id} + \Lambda \mathcal{T}_{tu(x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-tu(x)}^A)^{-1}. \quad (2)$$

This relation entails a substantial fractional operator, that is the fractional integral of order  $1 - \gamma$ ,  $I_{0,+}^{1-\gamma}$ , composed with path translations  $\mathcal{T}_{\pm tu(x)}^A$ . Here Id denotes the identity operator. The real numbers  $\Lambda \geq 0$  and  $\gamma > 0$  are constant parameters characterizing the physics of the trapping process while the real number  $\Lambda_v \geq 0$  characterizes convective motions.

Furthermore, anomalous diffusion is a transient behavior. For larger times, pdf  $P$  is governed by the classical Fickian Feynman-Kac equation:

$$\partial_t P - D\partial_{xx}^2 P + \Lambda_v \partial_x (vP) + u \partial_A P = r. \quad (3)$$

We recall that the fractional integral of order  $\alpha > 0$ ,  $I_{0,+}^\alpha f$ , is defined by

$$I_{0,+}^\alpha f(t) = \frac{1}{\Gamma(\alpha)} \int_0^t (t - t')^{\alpha-1} f(t') dt'.$$

This fractional integral generalizes the Cauchy multiple integral to non-integer order [47, 45]. Operator  $I_{0,+}^\alpha$  is bounded in  $L^p(0, T)$ ,  $1 \leq p \leq \infty$  [47]. The corresponding Laplace symbol is  $\lambda^{-\alpha}$ . We also recall that  $I_{0,+}^\alpha I_{0,+}^\beta = I_{0,+}^{\alpha+\beta}$ . Hence  $I_{0,+}^\alpha$  may be viewed as a “ $\alpha$ th root” of the time integration operator. The substantial fractional operator  $\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma$  expresses *memory effects*. In short, the future state depends on the present state *and* on the past. Markovity fails.

We observe in Eq. (1) that the micro-trapping process induces two anomalous transport properties at the mesoscale, on the meso-diffusive law *and* on the meso-convection. First, due to the presence of a substantial fractional operator  $\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma$  in the diffusive term, the classical Fick’s law is perturbed and the model is subdiffusive. Second, the trapping process also perturbs the convective process, once again through the substantial fractional operator  $\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma$ .

Heuristically Theorem 1 states that some scaling conditions lead to a transient anomalous transport and hence to a subdiffusive version of the Feynman-Kac equation. By the way, the present paper also describes precisely the critical scalings leading to normal or anomalous transport behavior:

**Extended Theorem 1** *The choices of scaling parameters leading to normal or abnormal transport (or to a freezing of the system due to asymptotically dominant trapping) are basically summarized in Fig. 1.*

Finally, let us mention that choosing  $\Lambda_v = 0$  we recover the Feynman-Kac equation associated with FFPE, that is the equation derived in [9] with other tools. More precisely:

**Theorem 2** *Assume  $\Lambda_v = 0$ . If the observation time is such that there is no scale separation between trapping and diffusion, the hydrodynamic limiting behavior of the probability density function  $P^*(x, t, A)$  for a tagged particle to be at point  $x$  at time  $t$  with the value  $A$  for the path integral  $A(t)$  is governed by a non-Fickian equation in the form*

$$\begin{aligned} \partial_t P^*(x, t, A) - D \partial_{xx}^2 (\Lambda^{-1} \mathcal{T}_{tu(x)}^A D_{0,+}^{1-\gamma} \mathcal{T}_{-tu(x)}^A) P^*(x, t, A) \\ + u(x) \partial_A P^*(x, t, A) = r(x, t, A), \end{aligned} \quad (4)$$

where operator  $D_{0,+}^{1-\gamma}$  is the Riemann-Liouville derivative of order  $1-\gamma$ , that is a left inverse of the fractional integral  $I_{0,+}^{1-\gamma}$ . Hence FFPE appears as a

**Scale separation parameters :**

- $\tau$  : time separation parameter
- $\ell$  : distance separation parameter
- $\tau_w$  : trapping separation parameter

$$\ell \sim \tau^{1/2}, \tau_w \sim \tau^{1/\gamma}$$

**Parameter characterizing the heterogeneity leading to the trapping :  $\gamma$**

**Scalings :**

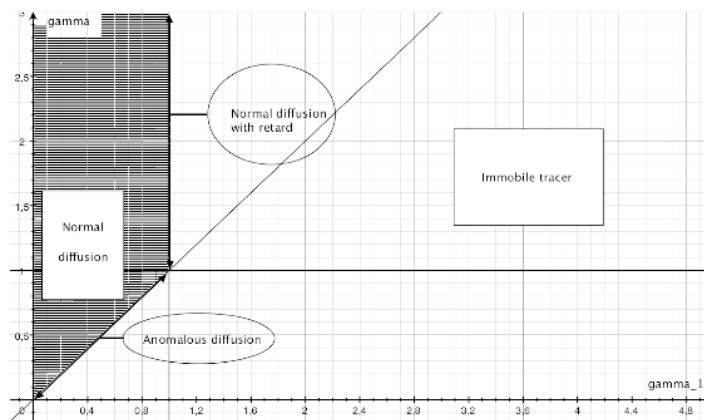
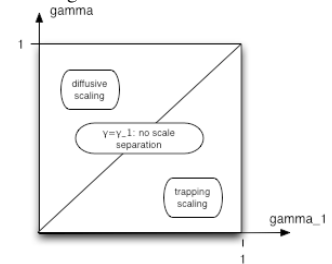


Figure 1: Governing models *versus* scaling choices. The scaling separation parameters are introduced in Section 2.2. Parameter  $\gamma_1$  scales trapping times with regard to observation times. Parameter  $\gamma$  characterizes the waiting times distribution of the trapping model and thus the heterogeneity degree leading to trapping (assumption  $\gamma < 1$  corresponds to long tails, assumption  $\gamma > 1$  corresponds to short tails).

*special case of our derivation. Moreover, convergence of the weak solutions of (1) to those of (4) is proved.*

The present paper is organized as follows. In Section 2, we construct a microscopic random walk model describing displacement of particles with trapping events. Our assumptions and especially the scaling parameters are introduced. Section 3 is devoted to the hydrodynamic limiting process. Anomalous (or not) behavior is induced by the relative importance of mobile and immobile densities. For each scaling choice, we derive the corresponding model at the mesoscale. In Section 4, we focus on the importance of scalings. We summarize when and why anomalous transport properties appear/disappear at the mesoscale and we interpret these phenomena in terms of operational and clock times. The mathematical analysis of the pde (1) is performed in Section 5. We also prove that solution of (4) is the limit of solution of (1). In Section 6 we derive the equation for observable quantities corresponding to the anomalous Feynman-Kac equation (1). Finally Annex A is devoted to the waiting times distributions with long tails. A power-law distribution is in particular justified using a basic geometrical model of heterogeneity.

## **2 A small-scale random walk model for flow with trapping processes**

### **2.1 Definition and assumptions**

The present study is based on a random walk process. Our starting point relies on some known facts in the classical Brownian setting. Assume that an average flow field  $v$  (here supposed constant) can be identified. A simple model for mass transport in homogeneous materials is obtained by considering that during successive time steps  $[n\Lambda_v\tau, (n+1)\Lambda_v\tau]$  fluid particles are advected over a distance  $v\Lambda_v\tau$ . Parameter  $\Lambda_v$  calibrates the duration of the convective step. Diffusion is taken into account by adding random independent instantaneous jumps of characteristic length scale  $\ell$  to the advective contribution. The total displacement length during time  $\Lambda_v\tau$  is thus  $v\Lambda_v\tau + \ell N^*$ , where  $N^*$  is a random variable with zero mean and unit variance. If  $\varphi$  represents the probability density function of  $N^*$  then

$$\varphi_\ell(\cdot) = \ell^{-1}\varphi(\cdot/\ell)$$



is the density of  $\ell N^*$ . We denote by  $x_t^\tau$  the random process that represents the position of a tagged walker at time  $t$ .

We now introduce an assumption on the scale separation parameters  $\ell$  and  $\tau$ . The so-called diffusive scaling consists in assuming that the microscopic and macroscopic distances (respect. times) satisfy  $x_{micro} = \ell^{-1}x_{macro}$  (respect.  $t_{micro} = \tau^{-1}t_{macro}$ ) and that the former scale separation parameters are related through

$$\ell = \sqrt{2D\tau}^{1/2}, \quad (5)$$

where  $D$  is a given nonnegative number. Letting  $\tau \rightarrow 0$ , the stochastic process  $x_t^\tau$  then asymptotically approaches the Brownian Motion  $x_t$  with diffusion coefficient  $D$  and drift  $v$  ([44]). Assuming that  $\varphi_\ell$  is a normal pdf with zero mean and standard deviation equal to  $\ell$  does not restrict the generality of this picture.

Suppose now that stagnation periods of random durations are superimposed to the above random walk due to small-scale heterogeneities. A natural way of accounting for such ‘‘sorption’’ events is that walkers are trapped at the end of each displacement of tracer or fluid parcels. For instance one may imagine a stream tube which winds tortuously through an array of persistent trapping eddies, or adsorption/desorption phenomena. We further assume that the sojourn time in the traps is itself a random variable  $t_w^*$ . Its average may be finite or not. As explained in the Introduction, we focus here on the second case. We thus assume that  $t_w^*$  obeys a power-law decaying pdf. More precisely, we assume:

**(H<sub>1</sub>) (waiting times distribution)** *The pdf  $\psi$  of the retention times is concentrated on  $\mathbb{R}_+$ , with survival probability  $\Psi$  of type*

$$\Psi(t) = \Lambda t^{-\gamma}/\Gamma(1 - \gamma) + K(t), \quad \gamma > 0.$$

*Function  $K$  is integrable in  $\mathbb{R}_+$  and such that  $\Psi$  is integrable in  $(0, 1)$ . The real number  $\gamma$  characterizes tails of the waiting time distribution: we assume*

$$\begin{aligned} (\mathbf{H}_1^{\text{long}}) \quad & 0 < \gamma < 1 \text{ for long tails,} \\ (\mathbf{H}_1^{\text{short}}) \quad & 1 < \gamma \text{ for short tails.} \end{aligned}$$

Prefactor  $\Gamma(\gamma - 1)$  is introduced for the sake of simplicity of computations while  $\Lambda$  is a constant introduced for dimensional consistency. Assumption **(H<sub>1</sub><sup>long</sup>)** is satisfied by pdf’s whose asymptotic behavior is a power-law. We might consider maximally skewed Lévy laws with exponent  $\gamma$  [27, 17, 35], which are concentrated on  $\mathbb{R}_+$  precisely for  $0 < \gamma < 1$ . The trivial degenerate

case of constant waiting times,  $\gamma = 1$ , corresponds to normal diffusion (see e.g. [39]).

Justification of Assumption  $(\mathbf{H}_1^{\text{long}})$  is postponed to Annex A. We now only give two fundamental remarks about this hypothesis.

**Remark 1** *Considering as in  $(\mathbf{H}_1^{\text{long}})$  waiting times distributions with no characteristic mean meets the ideas of [40] where subdiffusion was recovered through the asymptotic study of a stochastic differential equation after assuming no-scale separation and thus no possibility of a classical homogenization process (bear in mind that any homogenization process may be considered as the tracking of a mean behavior).*

**Remark 2** *A main ingredient of our fractal MIMs derivation is the choice  $(\mathbf{H}_1^{\text{long}})$  of power-law retention times. In FFPEs derivation such a power-law also characterizes residence times between diffusive jumps. Let us precise that the main difference between FPE and MIM is the subordinator which replaces the deterministic time by a random clock process. The equivalent Langevin description of both models is in terms of the subordinated process  $y(t) = x(S(t))$  where the parent process  $x(\cdot)$  is defined as the solution of the Itô stochastic differential equation*

$$dx(\tau) = \Lambda_v v d\tau + dB(\tau),$$

*driven by the standard Brownian motion  $B$ . In FFPE,  $S$  is the inverse stable subordinator  $S(t) = \inf\{s \in \mathbb{R}_+; W_{\mathcal{L}}(s) > t\}$ ,  $W_{\mathcal{L}}$  being a stable Levy process. In fractal MIM,  $S(t) = \inf\{s \in \mathbb{R}_+; \Lambda_v s + W_{\mathcal{L}}(s) > t\}$ . Hence our assumptions should lead to the FFPE derivation if  $\Lambda_v = 0$ . This point is rigorously confirmed in this work.*

*Microscopic models where the power-law characterizes diffusive jumps (Levy flights instead of Gaussian jumps) lead to limit models with a fractional space derivative ([53] for instance) which seems more suitable for super-diffusive settings.*

Let us now introduce a scale separation parameter  $\tau_w$  for the trapping process such that  $t_{micro}^{trap} = \tau_w^{-1} t_{macro}^{trap}$ . We choose to express it as a function of the time scale separation parameter  $\tau$  by setting

$$\tau_w = \tau^{1/\gamma_1}, \quad \gamma_1 > 0. \tag{6}$$

Note that the scaling relation (6) is constructed as the scaling relation (5). We have the classical diffusive scaling “distance=(time)<sup>1/2</sup>” and a trapping

scaling “trapping time=(time) $^{1/\gamma_1}$ ”. In short, we have the following assumption.

**(H $'_1$ ) (scaling of the waiting times)** *We introduce the rescaled variable*

$$t_w = \tau^{1/\gamma_1} t_w^*, \quad \gamma_1 > 0.$$

The real number  $\gamma_1$  is a parameter introduced to account for possibly different orders of magnitudes between sojourn times and observation time. This point is more detailed in Section 4 below.

The survival probability associated with  $t_w^*$  is

$$\int_t^{+\infty} \psi(t') dt'.$$

This quantity expresses the probability that the trapping time is longer than  $t$ . It follows that the rescaled pdf of  $t_w$  is

$$\psi_{\gamma_1}(t) = \tau^{-1/\gamma_1} \psi(t/\tau^{1/\gamma_1}),$$

while the rescaled survival probability is

$$\Psi_{\gamma_1}(t) = \Psi(t/\tau^{1/\gamma_1}) = \int_{t/\tau^{1/\gamma_1}}^{+\infty} \psi(t') dt'.$$

Assumptions **(H $_1$ , H $'_1$ )** provide a sufficient description of the walkers immobilization (immobile phase). For the time spent during displacements (mobile phase), several sceneries may be conceived, depending on the time of occurrence of the diffusive jump within a mobile period  $[t - \tau, t]$ . Since we assume here stochastic independence between waiting times and jumps, they are equivalent in the diffusive limit [38, 54]. For the sake of simplicity, we thus assume that walkers perform a single instantaneous diffusive jump at the end of each mobile period.

The present work aims at deriving an equation governing the limit behavior of the pdf  $P^\tau(x, t, A)$  for a tagged particle to be at point  $x$  at time  $t$ , with the value  $A$  for the path integral  $\int_0^t u(x^\tau(t')) dt'$ , as  $\tau \rightarrow 0$ . We assume:

**(H $_2$ ) (path integral integrand)** *The real function  $u$  is nonnegative and integrable, and belongs to  $L^\infty(\mathbb{R}_+)$ .*

We aim deriving the equation ruling the hydrodynamic limit  $P(x, t, A)$  of  $P^\tau(x, t, A)$ . We will show below that the existence of  $P$  is related to the behavior of the density distribution  $m^\tau(x, t, A)$  defined as the pdf of

just ending a stagnation period at time  $t$  and position  $x$  with value  $A$ . We assume:

**(H<sub>3</sub>) (mobility)** *There exists a limit function  $l_m \in W^{1,1}((0, T) \times \mathbb{R}^2)$  such that the sequence of density distribution  $(m^\tau)_{\tau>0} \subset \mathcal{D}'((0, T) \times \mathbb{R}^2)$  satisfies:*

$$\lim_{\tau \rightarrow 0} \tau^{-1/2} \|\tau m^\tau - l_m\|_{L^1(0, T; X)} = 0, \quad X = L^1(\mathbb{R}, L^1(\mathbb{R})).$$

We choose here to work within the framework of  $L^1$  functions instead of using measures. This choice is essentially motivated by the use of density functions and the corresponding simplicity of notations. Indeed our aim in the present paper is to focus on the importance of scalings. Integral representations are interpretable physically and clarify the structure of the limiting processes. The interested reader will find the details of the proof in the context of measures in [10].

**Remark 3** *In the present paper, the hydrodynamic limit  $\tau \rightarrow 0$  consists in studying weak limit in  $X$ , and therefore testing against functions that live on the macroscopic scale. Part of the microscopic information is lost through this mechanism and lead to the irreversibility of the macro-model, even in case of reversibility of the micro-model.*

## 2.2 Probability densities for the mobile and immobile phases at small scale

Particles performing such random walks with sorption can be thought of as belonging alternatively to two distinct “phases”: at each time step walkers are said to be in the immobile or in the mobile phase according they are trapped or not. Note that this succession of “Stop and Go” is in some sense comparable to the sequence of “On and Off” of network traffic models ([26]). We proceed to derive an explicit microscopic relation that links the particles densities in the two phases. The hydrodynamic limit will be addressed in the next Section.

Let  $\tau > 0$  be given. Let  $P_i^\tau(x, t, A)$  represent the density of trapped particles at location  $x$  and time  $t$  such that the path integral  $\int_0^t u(x^\tau(t')) dt'$  has the value  $A$ . Formally, quantity  $P_i^\tau(x, t, A) dx dA$  is the probability to find a tagged walker between  $x$  and  $x + dx$ , with the path integral between  $A$  and  $A + dA$ . Similarly,  $P_m^\tau(x, t, A)$  represents the density of mobile walkers. In order to establish a relation between densities  $P_i^\tau$  and  $P_m^\tau$ , we introduce the pdf's  $i^\tau(x, t, A)$  of just arriving and being immobilized at point  $x$  and time  $t$  with the value  $A$  for the path integral, and  $m^\tau(x, t, A)$  of just being

released by a sorbing site (at point  $x$ , time  $t$  with the value  $A$ ). We recall that density  $m^\tau$  is characterized in assumption **(H<sub>3</sub>)**.

Mobile particles at position  $x$  at time  $t$  have been trapped and then released at time  $t - t'$  and at a distance  $vt'$  from  $x$ , for some  $t' > 0$ . Moreover, particles attached (at point  $x$  and time  $t$ ) to the value  $A$  of the integral  $\int_0^t u(x^\tau(\theta))d\theta$  correspond at time  $t - t'$  (and at point  $x - vt'$ ) to the value  $A - \int_0^{t'} u(x - vt' + v\theta)d\theta$ . This implies

$$\begin{aligned} & P_m^\tau(x, t, A) \\ &= \int_0^{\Lambda_v \tau} m^\tau(x - vt', t - t', A - \int_0^{t'} u(x - vt' + vs)ds) dt' \\ &= \int_0^{\Lambda_v \tau} \mathcal{T}_{vt', t'} \mathcal{T}_{\int_0^{t'} u(x+vs)ds}^A m^\tau(x, t, A) dt', \end{aligned}$$

that is

$$\begin{aligned} P_m^\tau &= \Lambda_v P_m^{\tau, aux}, \\ P_m^{\tau, aux}(x, t, A) &= \int_0^\tau \mathcal{T}_{\Lambda_v vt', \Lambda_v t'} \mathcal{T}_{\int_0^{\Lambda_v t'} u(x+vs)ds}^A m^\tau(x, t, A) dt'. \end{aligned} \quad (7)$$

Convective displacements are represented through operator  $\mathcal{T}_{u,w}$  which denotes translation in space and time. We set

$$\mathcal{T}_{u,w}G(x, t) = H(t - w)G(x - u, t - w),$$

$H$  being the Heaviside step function. Translations of amplitude  $b$  in the direction of  $A$ , are denoted by  $\mathcal{T}_b^A$ . Note that all the translations used in the present paper trivially become  $Id$  if  $\Lambda_v = 0$ .

Immobile particles that are in  $x$  at time  $t$  must have jumped there previously at time  $t - t'$ , for some  $t' > 0$ , been trapped and stayed there up to  $t$ . During this immobile period, the increase of path integral  $A$  is  $u(x)t'$ . Hence, denoting time convolutions of functions in  $\mathbb{R}_+$  by  $*$ , *i.e.*

$$F * G(t) = \int_0^t F(t - t')G(t') dt',$$

we have

$$\begin{aligned} P_i^\tau(x, t, A) &= \int_0^t \Psi_{\gamma_1}(t') i^\tau(x, t - t', A - t' u(x)) dt' \\ &= \mathcal{T}_{u(x)t}^A \int_0^t \Psi_{\gamma_1}(t') i^\tau(x, t - t', A + (t - t')u(x)) dt' \\ &= \mathcal{T}_{u(x)t}^A [\Psi_{\gamma_1} * \mathcal{T}_{-u(x)\cdot t}^A i^\tau(x, \cdot, \cdot, A)](x, t, A) \end{aligned} \quad (8)$$

where  $\cdot_y$  designs a variable ( $y = t, x$  or  $A$ ) involved in convolutions or translations.

Equations (7) and (8) were gained upon addressing separately what happens to a tagged walker in the mobile and immobile phases. We now link these steps. Particles just arriving at  $x$  at time  $t > \Lambda_v \tau$  (where they will be immobilized) may have been trapped and released or have been injected into the system by the source, in each case at time  $t - \Lambda_v \tau$ . During this latter mobile period, that began at point  $x - y - \Lambda_v v \tau$ , path integral  $A$  was increased of  $\int_0^{\Lambda_v \tau} u(x - y - \Lambda_v v \tau + v \theta) d\theta = \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \int_0^{\Lambda_v \tau} u(x - y + v s) ds$ . Indeed, the diffusive jump was assumed to be instantaneous and to occur at the end of the current period. Hence, quantity  $i^\tau(x, t, A)$  reads, for  $t > \Lambda_v \tau$ ,

$$\begin{aligned} & i^\tau(x, t, A) \\ &= \int_{\mathbb{R}} m^\tau \left( x - y - \Lambda_v v \tau, t - \Lambda_v \tau, A - \int_0^{\Lambda_v \tau} u(x - y - \Lambda_v v \tau + v s) ds \right) \varphi_\ell(y) dy \\ &= (\varphi_\ell(\cdot_x) \star \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(\cdot_x + v s) ds}^A m^\tau)(x, t, A). \end{aligned} \quad (9)$$

We denote by  $\star$  the space convolution, that is

$$f \star g(x) = \int_{\mathbb{R}} f(x - x') g(x') dx'.$$

Quantity  $i^\tau$  is thus fully described by the knowledge of  $m^\tau$ .

According to Eq. (7), the weighted density  $\tau^{-1} P_m^{\tau, aux}(x, t, A)$  is the average of the quantity  $\mathcal{T}_{\Lambda_v v t', \Lambda_v t'} \mathcal{T}_{\int_0^{\Lambda_v t'} u(x + v s) ds}^A m^\tau(x, t, A)$  over an interval of amplitude  $\tau$ . Thus it approximates  $\mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(x + v s) ds}^A m^\tau(x, t, A)$  when  $\tau \rightarrow 0$ , at least for smooth functions of time. This latter assumption is actually not necessary to ensure that replacing  $\mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(x + v s) ds}^A m^\tau(x, t, A)$  by  $\tau^{-1} P_m^{\tau, aux}(x, t, A)$  results into a small error for  $P_i^\tau$ . This argument will be checked in the next subsection. We thus define the error  $\varepsilon^\tau$  corresponding to this approximation:

$$\begin{aligned} & \varepsilon^\tau(x, t, A) \\ &= \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(x + v s) ds}^A m^\tau(x, t, A) - \frac{P_m^{\tau, aux}(x, t, A)}{\tau} \\ &= \int_0^1 \left( \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(x + v s) ds}^A - \mathcal{T}_{\Lambda_v v \theta \tau, \Lambda_v \theta \tau} \mathcal{T}_{\int_0^{\Lambda_v \theta \tau} u(x + v s) ds}^A \right) m^\tau(x, t, A) d\theta. \end{aligned} \quad (10)$$

From relations (8) and (9) that read

$$\begin{aligned} & P_i^\tau(x, t, A) \\ &= \mathcal{T}_{u(x)t}^A [\Psi_{\gamma_1} * \mathcal{T}_{-u(x)\cdot t}^A (\varphi_\ell * \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(\cdot + vs) ds}^A) m^\tau](x, t, A), \end{aligned} \quad (11)$$

and from the decomposition (10)  $\mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(x+vs) ds}^A m^\tau = \tau^{-1} P_m^{\tau, aux} + \varepsilon^\tau$ , we infer:

$$P_i^\tau(x, t, A) = R^\tau P_m^{\tau, aux}(x, t, A) + E^\tau(x, t, A), \quad (12)$$

where

$$R^\tau g(x, t, A) = \tau^{-1} \mathcal{T}_{u(x)t}^A \Psi_{\gamma_1} * \mathcal{T}_{-u(x)\cdot t}^A [\varphi_\ell * g](x, t, A), \quad (13)$$

$$E^\tau(x, t, A) = \mathcal{T}_{u(x)t}^A \Psi_{\gamma_1} * \mathcal{T}_{-u(x)\cdot t}^A [\varphi_\ell * \varepsilon^\tau](x, t, A) = R^\tau [\tau \varepsilon^\tau](x, t, A). \quad (14)$$

In the next Section, we prove that the operator  $R^\tau$  may asymptotically lead to the introduction of a substantial derivative operator, quite similar to the one already introduced (with a different formulation) by [18] and [9]. We also check that  $E^\tau$  tends to zero as  $\tau \rightarrow 0$ . To this aim, we provide in the next subsection some auxiliary convergence results.

### 2.3 Some convergence results

We begin with an auxiliary result.

**Lemma 1** *Let  $g \in L^1(0, T; X)$ . We define application  $\mathcal{A}[g] : (0, T) \rightarrow L^1(0, T; X)$  by*

$$\mathcal{A}[g](t') = \mathcal{T}_{\Lambda_v v t', \Lambda_v t'} \mathcal{T}_{\int_0^{\Lambda_v t'} u(\cdot + vs) ds}^A [g], \quad t' \in (0, T).$$

*Then we have*

$$\lim_{\tau_1 \rightarrow 0} \left\| \mathcal{A}[g](\tau_1) - \tau_1^{-1} \int_0^{\tau_1} \mathcal{A}[g](t') dt' \right\|_{L^1(0, T; X)} = 0.$$

*If furthermore  $g \in W^{1,1}((0, T) \times \mathbb{R}^2)$ ,*

$$\lim_{\tau_1 \rightarrow 0} \tau_1^{-1/2} \left\| \mathcal{A}[g](\tau_1) - \tau_1^{-1} \int_0^{\tau_1} \mathcal{A}[g](t') dt' \right\|_{L^1(0, T; X)} = 0.$$

**Proof.** By continuity of translation operator, with in particular

$$\lim_{(x_1, t_1, A_1) \rightarrow 0} \|g(\cdot_x - x_1, \cdot_t - t_1, \cdot_A - A_1) - g(\cdot_x, \cdot_t, \cdot_A)\|_{L^1(0, T; X)} = 0,$$

and continuity of the integration operator, we check straightforward that application  $\mathcal{A}[g]$  is continuous in  $(0, T)$ . Its average is thus known to approximate it in  $(0, T)$ . More precisely, if  $\|\cdot\|_{L^1} = \|\cdot\|_{L^1(0, T; X)}$ ,

$$\begin{aligned} \left\| \mathcal{A}[g](\tau_1) - \tau_1^{-1} \int_0^{\tau_1} \mathcal{A}[g](t') dt' \right\|_{L^1} &= \left\| \int_0^1 (\mathcal{A}[g](\tau_1) - \mathcal{A}[g](\theta\tau_1)) d\theta \right\|_{L^1} \\ &\leq \left\| \max_{\theta \in (0, 1)} |\mathcal{A}[g](\tau_1) - \mathcal{A}[g](\theta\tau_1)| \right\|_{L^1} = \left\| |\mathcal{A}[g](\tau_1) - \mathcal{A}[g](\theta_o\tau_1)| \right\|_{L^1} \end{aligned}$$

for some  $\theta_o : \mathbb{R} \rightarrow (0, 1)$ . Since  $\mathcal{A}[g] : (0, T) \rightarrow L^1(0, T; X)$  is continuous, it follows that the right-hand side of the latter expression tends to zero as  $\tau_1 \rightarrow 0$ . If furthermore  $g \in W^{1,1}((0, T) \times \mathbb{R}^2)$ , we use a Taylor's type formula to assert that

$$\lim_{\tau_1 \rightarrow 0} \tau_1^{-1/2} \left\| \mathcal{A}[g](\tau_1) - \mathcal{A}[g](\theta_o\tau_1) \right\|_{L^1(0, T; X)} = 0.$$

This ends the proof of the lemma.  $\square$

We now study the limit behavior of the correction  $\varepsilon^\tau$  defined in (10). We prove the following result.

**Lemma 2** *The following result holds true:*

$$\tau^{1/2} \varepsilon^\tau \rightarrow 0 \text{ in } L^1(0, T; X).$$

**Proof.** Using application  $\mathcal{A}$  defined in Lemma 1, we note that  $\varepsilon^\tau = \mathcal{A}[m^\tau](\tau) - \tau^{-1} \int_0^\tau \mathcal{A}[m^\tau](t') dt'$ . Inserting the  $L^1$ -limit  $l_m$  of  $\tau m^\tau$  in the latter expression, we have

$$\begin{aligned} \tau \varepsilon^\tau &= (\mathcal{A}[\tau m^\tau](\tau) - \mathcal{A}[l_m](\tau)) + \left( \mathcal{A}[l_m](\tau) - \tau^{-1} \int_0^\tau \mathcal{A}[l_m](t') dt' \right) \\ &\quad + \tau^{-1} \int_0^\tau (\mathcal{A}[l_m] - \mathcal{A}[\tau m^\tau])(t') dt'. \end{aligned}$$

It remains to prove that the three terms in the right-hand side of the latter relation tend to zero in  $L^1(0, T; X)$  faster than  $\tau^{1/2}$ . Using the continuity of the translation operator in  $L^1$ , we compute:

$$\begin{aligned} &\tau^{-1/2} \left\| \mathcal{A}[\tau m^\tau](\tau) - \mathcal{A}[l_m](\tau) \right\|_{L^1(0, T; X)} \\ &= \tau^{-1/2} \left\| \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(\cdot_x + vs) ds}^A [\tau m^\tau - l_m] \right\|_{L^1(0, T; X)} \\ &\leq \tau^{-1/2} \left\| \tau m^\tau - l_m \right\|_{L^1(0, T; X)} \rightarrow 0 \text{ as } \tau \rightarrow 0. \end{aligned}$$



Since function  $l_m$  belongs to  $W^{1,1}((0, T) \times \mathbb{R}^2)$  and does not depend on  $\tau$  (see **(H3)**), we assert with Lemma 1 that

$$\tau^{-1/2} \left\| \mathcal{A}[l_m](\tau) - \tau^{-1} \int_0^\tau \mathcal{A}[l_m](t') dt' \right\|_{L^1(0, T; X)} \rightarrow 0 \text{ as } \tau \rightarrow 0.$$

Finally, we compute:

$$\begin{aligned} & \tau^{-1/2} \left\| \tau^{-1} \int_0^\tau (\mathcal{A}[l_m] - \mathcal{A}[\tau m^\tau])(t') dt' \right\|_{L^1(0, T; X)} \\ = & \tau^{-1/2} \left\| \tau^{-1} \int_0^\tau \mathcal{T}_{\Lambda_v v t', \Lambda_v t'} \mathcal{T}_{\int_0^{\Lambda_v t'} u(\cdot_{x+vs}) ds}^A [l_m - \tau m^\tau] dt' \right\|_{L^1(0, T; X)} \\ & \leq \tau^{-3/2} \int_0^\tau \|l_m - \tau m^\tau\|_{L^1(0, T; X)} dt' \\ = & \tau^{-1/2} \|l_m - \tau m^\tau\|_{L^1(0, T; X)} \rightarrow 0 \text{ as } \tau \rightarrow 0. \end{aligned}$$

Lemma 2 is proved.  $\square$

Now we claim and prove the following result.

**Proposition 1** *There exists some function  $P_m$  belonging to  $W^{1,1}((0, T) \times \mathbb{R}^2)$  such that the following convergence holds true as  $\tau \rightarrow 0$ :*

$$\tau^{-1/2} (P_m^{\tau, aux} - P_m^{aux}) \rightarrow 0 \text{ in } L^1(0, T; X).$$

**Proof.** Due to (10),

$$P_m^{\tau, aux} = \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\tau \Lambda_v} u(\cdot_{x+vs}) ds}^A [\tau m^\tau] - \tau \varepsilon^\tau. \quad (15)$$

The result of the present proposition thus follows from Assumption **(H3)** and Lemma 2.  $\square$

**Remark 4** *In view of (15) and of the previous results, Assumption **(H3)** on  $\tau m^\tau$  (and not on  $m^\tau$ ) is the minimal one to get a non-trivial limit behavior for  $P_m^\tau$ , that is  $P_m \neq 0$  if  $\Lambda_v \neq 0$ .*

### 3 Hydrodynamic limit: governing equations *versus* scaling

The keypoint in deriving the macroscopic equations governing the hydrodynamic limit of the small scale processes described above is the limit of Eq. (12). It is the purpose of the next two subsections. In the third one, we collect our results and give the appropriate form of the Feynman-Kac equation for each scaling choice (thus recovering results of Fig. 1).

### 3.1 Relation between mobile and immobile densities if $\gamma < 1$

We now show that the mapping  $R^\tau$  defined by Eq. (13) converges to a combination of translations in the  $A$ -direction with a fractional integral with regard to time.

To highlight the dominant singular term in the time convolution of kernel  $\tau^{-1}\Psi_\gamma$ , for any  $g \in L^1(0, T; X)$  we decompose  $R^\tau g$  in

$$R^\tau g = I_1^\tau g + I_2^\tau g, \quad (16)$$

where

$$\begin{aligned} & I_1^\tau g(x, t, A) \\ &= \frac{\Lambda \tau^{\gamma/\gamma_1 - 1}}{\Gamma(1 - \gamma)} \int_0^t (t')^{-\gamma} (\varphi_\ell \star g)(x, t - t', A - t' u(x)) dt', \end{aligned} \quad (17)$$

$$\begin{aligned} & I_2^\tau g(x, t, A) \\ &= \tau^{1/\gamma_1 - 1} \int_0^t \tau^{-1/\gamma_1} K\left(\frac{t'}{\tau^{1/\gamma_1}}\right) (\varphi_\ell \star g)(x, t - t', A - t' u(x)) dt', \end{aligned} \quad (18)$$

thanks to hypothesis  $(\mathbf{H}_1, \mathbf{H}'_1)$  that read:

$$\tau^{-1}\Psi_{\gamma_1}(t) = \Lambda \frac{t^{-\gamma}}{\Gamma(1 - \gamma)} \tau^{\gamma/\gamma_1 - 1} + \tau^{-1} K(t/\tau^{1/\gamma_1}).$$

Let us begin with some results about the limit behavior of operators  $I_i^\tau$ ,  $i = 1, 2$ .

The time convolution of kernel  $\Lambda t^{-\gamma}/\Gamma(1 - \gamma)$  is precisely the fractional integral  $\Lambda I_{0,+}^{1-\gamma}$ . Then  $I_1^\tau$  satisfies

$$\begin{aligned} I_1^\tau g(x, t, A) &= \tau^{\gamma/\gamma_1 - 1} \mathcal{T}_{tu(x)}^A \Lambda \left[ \frac{\cdot^{-\gamma}}{\Gamma(1 - \gamma)} * \mathcal{T}_{-u(x)\cdot}^A [\varphi_\ell(\cdot) \star g(\cdot)] \right] (x, t, A) \\ &= \tau^{\gamma/\gamma_1 - 1} \mathcal{T}_{tu(x)}^A \Lambda I_{0,+}^{\gamma-1} \mathcal{T}_{-u(x)\cdot}^A [\varphi_\ell \star g](x, t, A) \end{aligned} \quad (19)$$

for any  $g \in L^1(0, T; X)$ . Convolution of kernel  $\varphi_\ell$  tends to the identity operator  $\text{Id}$  as  $\ell \rightarrow 0$ . Thus, for any  $g \in L^1(0, T; X)$ , we have the strong convergence result

$$\langle \tau^{1-\gamma/\gamma_1} I_1^\tau g, h \rangle_{\mathcal{D}' \times \mathcal{D}} \rightarrow \langle \Lambda \mathcal{T}_{tu(x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x)\cdot}^A g, h \rangle \text{ in } L_x^1(\mathbb{R})$$

as  $\tau \rightarrow 0$ , for any *ad hoc* test function  $h$  defined in  $\mathbb{R}^2$ . Moreover, if  $(g^\ell)$  is a sequence of  $L^1(0, T; X)$  which is weakly convergent in  $L^1(0, T; X)$  to some

limit  $g$ , we recall that for any test function  $h$ ,

$$\begin{aligned} & \int_{(0,T) \times \mathbb{R}} \int_{\mathbb{R}} (\varphi_\ell \star g^\ell) h \, dx dt dA \\ &= \int_{(0,T) \times \mathbb{R}} \int_{\mathbb{R}} g^\ell (\widetilde{\varphi}_\ell \star h) \, dx dt dA \rightarrow \int_{(0,T) \times \mathbb{R}} \int_{\mathbb{R}} gh \, dx dt dA \end{aligned}$$

as  $\ell \rightarrow 0$ , where  $\widetilde{\varphi}_\ell = \varphi_\ell \circ (-\text{Id}) = \varphi_\ell$  because of symmetry. We infer from the two latter relations the following lemma.

**Lemma 3** *For any sequence  $(g^\tau)$  of  $L^1(0, T; X)$  which is weakly convergent in  $L^1(0, T; X)$  to some limit  $g$ , we have*

$$\tau^{1-\gamma/\gamma_1} I_1^\tau g^\tau(x, t, A) \rightharpoonup \Lambda \mathcal{T}_{tu(x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x),t}^A g(x, t, A) \text{ weakly in } L^1(0, T; X).$$

**Remark 5** *On the right-hand side of the latter convergence result, for any fixed  $x \in \mathbb{R}$ , we recognize a mapping whose double Laplace transform w.r.t.  $t$  and  $A$  has symbol  $(\lambda + pu(x))^{\gamma-1}$  when the range of  $A$  is restricted to  $\mathbb{R}_+$  (see Appendix B). Since [18] and [9] named “substantial fractional derivative” the mapping of symbol  $(\lambda + pu(x))^\alpha$ , we propose to consider that  $\mathcal{T}_{tu(x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x),t}^A$  is a **substantial fractional integral** of order  $1 - \gamma$ .*

We now consider the operator  $I_2^\tau$  defined as

$$I_2^\tau g(x, t, A) = \tau^{\frac{1}{\gamma_1}-1} \mathcal{T}_{tu(x)}^A \tau^{-\frac{1}{\gamma_1}} K\left(\frac{\cdot t}{\tau^{1/\gamma_1}}\right) * [\mathcal{T}_{-u(x),t}^A [\varphi_\ell \star g]](x, t, A),$$

where  $\tau^{-\frac{1}{\gamma_1}} K\left(\frac{\cdot t}{\tau^{1/\gamma_1}}\right)$  is the kernel of an approximation in time for Id. Hence the limit behavior of  $I_2^\tau g$  depends on the value of  $(1/\gamma_1 - 1)$ . More precisely, we have the following result.

**Lemma 4** *For any sequence  $(g^\tau)$  of functions in  $L^1(0, T; X)$  weakly convergent to  $g$  in  $L^1(0, T; X)$ , we have*

$$\tau^{1-\frac{1}{\gamma_1}} I_2^\tau g^\tau \rightharpoonup g \text{ weakly in } L^1(0, T; X).$$

Using the two previous lemmas with Proposition 1, we compute the limit behavior of  $R^\tau P_m^\tau$ . The limit behavior of the correction  $E^\tau = I_1^\tau [\tau \varepsilon^\tau] + I_2^\tau [\tau \varepsilon^\tau]$  is obtained using Lemmas 2, 3, and 4. We have proved the following result.

**Proposition 2** ( $\gamma < 1$ )

(i) If  $\gamma_1 = \gamma$ , the transport in the limit  $\tau \rightarrow 0$  evolves to a non-Fickian one. More precisely, the mapping  $R^\tau$  of  $L^1(0, T; X)$  with  $X = L^1(\mathbb{R}, L^1(\mathbb{R}))$  tends to the substantial fractional integral operator  $\Lambda \mathcal{T}_{\cdot u(\cdot x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(\cdot x)\cdot t}^A$  when  $\tau \rightarrow 0$ . Densities of immobile and mobile walkers are related through

$$P_i(x, t, A) = \Lambda \mathcal{T}_{tu(x)}^A I_{0,+}^{1-\gamma} [\mathcal{T}_{-u(x)\cdot t}^A P_m^{aux}](x, t, A), \quad (20)$$

while the total density of walkers  $P = P_m + P_i = \Lambda_v P_m^{aux} + P_i$  satisfies

$$P_m^{aux} = \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P, \quad (21)$$

$$\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma = (\Lambda_v Id + \Lambda \mathcal{T}_{\cdot u(\cdot x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-t u(\cdot x)}^A)^{-1}. \quad (22)$$

(ii) If  $\gamma_1 < \gamma$ , that is if the observation time scale is larger than the largest heterogeneity scale, the transport becomes normal and there is no influence of the trapping processes in the limit  $\tau \rightarrow 0$ :

$$\Lambda_v P_m^{aux} = P. \quad (23)$$

(iii) If  $\gamma < \gamma_1$ , tracer is asymptotically immobile.

All these convergence results hold true in  $L^1(0, T; X)$  with a convergence rate of order  $\tau^{1/2}$ .

We note that the rate of convergence  $\tau^{1/2}$  in Assumption **(H<sub>3</sub>)** is useless to prove items (i)-(iii). This rate assumption is only necessary in Subsection 3.3 for the study of the physical fluxes.

**Proof.** Proof of points (i) and (ii) based on Lemmas 3 and 4 is straightforward. We thus only detail point (iii). First we have

$$\begin{aligned} \tau^{1-\gamma/\gamma_1} P_i^\tau &= (\tau^{1-\gamma/\gamma_1} I_1^\tau P_m^{\tau, aux}) + \tau^{(1-\gamma)/\gamma_1} (\tau^{1-1/\gamma_1} I_2^\tau P_m^{\tau, aux}) \\ &\quad + (\tau^{1-\gamma/\gamma_1} I_1^\tau \tau \varepsilon^\tau) + \tau^{(1-\gamma)/\gamma_1} (\tau^{1-1/\gamma_1} I_2^\tau \tau \varepsilon^\tau). \end{aligned}$$

We infer from Lemmas 2, 3 and 4 that the right-hand side of the latter relation tends to  $P_m^{aux}$  as  $\tau \rightarrow 0$ . Using (11), we write the left-hand side in the form

$$\begin{aligned} \tau^{1-\gamma/\gamma_1} P_i^\tau &= \tau^{1-\gamma/\gamma_1} R^\tau [\tau \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(\cdot x + vs) ds}^A m^\tau] \\ &= \tau^{1-\gamma/\gamma_1} I_1^\tau [\mathcal{A}[\tau m^\tau](\tau)] + \tau^{(1-\gamma)/\gamma_1} \tau^{1-1/\gamma_1} I_2^\tau [\mathcal{A}[\tau m^\tau](\tau)] \end{aligned}$$

that tends to  $l_m$  in  $L^1(0, T; X)$ . Finally, we have

$$\begin{aligned} P_m^{\tau, aux} &\rightarrow P_m^{aux} = l_m \text{ in } L^1(0, T; X), \\ \tau^{1-\gamma/\gamma_1} P_i^\tau &\rightarrow l_m \text{ in } L^1(0, T; X), \end{aligned}$$

the second convergence result with  $1 - \gamma/\gamma_1 > 0$  meaning that immobile particles become asymptotically dominant.  $\square$

**Remark 6** Relation (20) for the density of immobile walkers is derived here from the microscopic scale. In [55] Young derived a similar relation, with  $\gamma = 1/2$ , by working directly at the mesoscopic scale on a particular flow geometry. He considered a pipe containing a mobile tracer of concentration  $c_m$  with many stagnant side branches where the tracer is immobilized and of concentration  $c_i$ . He computed  $c_i$  solution of a purely diffusive equation in the one-dimensional infinite branches with the continuous boundary condition  $c_i = c_m$  at the pipe/branches transitions. Thanks to the explicit integral representation of such a solution, he obtained  $c_i \sim I_{0,+}^{1/2} c_m$  (see (2.7) in [55]).

**Remark 7** Notice that we also have proved that if  $\gamma_1 < \gamma$ , transport becomes normal, without the usual technical assumption of a truncated in time pdf  $\psi$  (see for instance [13, 5]). This point deserves being detailed and explained, which is done in Section 4.

### 3.2 Relation between mobile and immobile densities if $\gamma > 1$

In the present subsection we assume for instance that  $\psi$  is such that  $\psi(t) \sim_{+\infty} t^{-\gamma-1}$ ,  $\gamma > 1$ , with a finite mean. The survival probability  $\Psi$  is thus integrable in  $\mathbb{R}_+$  and we can define the mean pause by

$$R_\gamma = \int_0^\infty \Psi(t) dt.$$

Note that  $R_\gamma \rightarrow 0$  as  $\gamma \rightarrow \infty$ . We turn back to relation (12), that is

$$P_i^\tau = R^\tau [P_m^{\tau,aux}] + R^\tau [\tau \varepsilon^\tau]$$

with  $R^\tau [g](x, t, A) = \tau^{-1+1/\gamma_1} \mathcal{T}_{u(x)t}^A \tau^{-1/\gamma_1} \Psi(\cdot/t/\tau^{1/\gamma_1}) * \mathcal{T}_{-u(x)_t}^A [\varphi_\ell * g](x, t, A)$ . Time convolution of kernel  $\tau^{-1/\gamma_1} \Psi(\cdot/\tau^{1/\gamma_1})$  tends to  $R_\gamma \text{Id}$  as  $\tau \rightarrow 0$ . The following result follows.

**Proposition 3** ( $\gamma > 1$ )

(i) If  $\gamma_1 = 1$ , limit densities are related through the following relations involving the retardation factor  $R_\gamma$ :

$$P_i = R_\gamma P_m^{aux}, \quad P = (\Lambda_v + R_\gamma) P_m. \quad (24)$$

(ii) If  $\gamma_1 < 1$ , transport is normal and we recover once again (23).

(iii) If  $\gamma_1 > 1$ , the tracer is asymptotically immobile.

All these convergence results hold true in  $L^1(0, T; X)$  with a convergence rate of order  $\tau^{1/2}$ .

### 3.3 Study of the fluxes: a p.d.e. for the evolution of $P$

In the present subsection, we do not assume that we can substitute directly the tracer concentration to the probability density and that the tracer concentration is smooth enough to be Taylor expendable, as in [6]. We do not either choose to restrict ourself to the asymptotic study of the moments. We prefer applying mass conservation principle and getting an expression of the flux of walkers in the space  $\{(x, A) \in \mathbb{R}^2\}$ . This physical argument leads to the generator of the process under interest.

For this purpose, we derive a variant of Fick's law for the hydrodynamic limit of our random walk model. Basically, we have to count the walkers that cross a point of given coordinate  $x$ , or a given level  $A$  of the path integral, during any time interval  $[t, t + dt]$ . Crossings may occur strictly inside the current mobile period or at the very end. The first alternative involves convective motions due to the average velocity field  $v$ , and the second alternative corresponds to diffusive jumps. The probability current is a vector with two coordinates ( $x$  and  $A$ ). The first one represents the averaged balance of walkers crossing point  $x$ : it is equal to the probability for a tagged walker to cross  $x$  to the right minus the probability to cross  $x$  to the left. The second one is equal to the probability for the path integral carried by a tagged walker to become larger than  $A$  minus the probability to become smaller than  $A$ .

Let us now translate the former intuitions into a mathematical formulation. Particles that cross  $x$  toward the right during the period are mobile and have spent a time  $t' \in (0, \Lambda_v \tau)$  in the mobile phase, resulting into a convective displacement of length  $vt'$ . Assume first that  $v$  is nonnegative. On the one hand, crossings through point  $x$  occur due to convection before the end of the current mobile period with probability

$$F_c^\tau = \int_0^{\Lambda_v \tau} \mathcal{T}_{vt', t'} \mathcal{T}_{\int_0^{t'} u(x+vs) ds}^A m^\tau v dt dt' = \Lambda_v v dt P_m^{\tau, aux}. \quad (25)$$

On the other hand, at the end of a mobile period for  $t' = \Lambda_v \tau$ , a diffusive random jump may allow the walker to cross  $x$  and this occurs with the probability:

$$\int_0^\infty \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \mathcal{T}_{\int_0^{\Lambda_v \tau} u(x-y+vs) ds}^A m^\tau(x-y, t, A) \Phi\left(\frac{y}{\sqrt{2D\tau}}\right) dy dt.$$

Indeed, we recall that we assume  $\ell = \sqrt{2D\tau}$ . Function  $\Phi$  defined by

$$\Phi(z) = \int_z^\infty \varphi(s) ds$$

represents the probability for  $N^*$  to take a value larger than  $z$  (see the notations in the beginning of Subsection 2.1). Accounting for crossings to the right yields the following global contribution of diffusive randoms jumps to the probability current:

$$F_D^\tau(x, t, A) = \int_0^\infty \mathcal{T}_{\Lambda_v v \tau, \Lambda_v \tau} \left( m^\tau(x - y, t, A - \int_0^{\Lambda_v \tau} u(x - y + vt') dt') \right. \\ \left. - m^\tau(x + y, t, A - \int_0^{\Lambda_v \tau} u(x + y + vt') dt') \right) \Phi\left(\frac{y}{\sqrt{2D\tau}}\right) dy.$$

Thanks to (15), the latter expression is rewritten using  $P_m^\tau$  and  $\varepsilon^\tau$ :

$$F_D^\tau = \int_0^\infty \frac{P_m^{\tau, aux}(x - y, t, A) - P_m^{\tau, aux}(x + y, t, A)}{\tau} \Phi\left(\frac{y}{\sqrt{2D\tau}}\right) dy \\ + \int_0^\infty \frac{\tau \varepsilon^\tau(x - y, t, A) - \tau \varepsilon^\tau(x + y, t, A)}{\tau} \Phi\left(\frac{y}{\sqrt{2D\tau}}\right) dy \\ = 2D \int_0^\infty \frac{P_m^{\tau, aux}(x - \sqrt{2D\tau}Y, t, A) - P_m^{\tau, aux}(x + \sqrt{2D\tau}Y, t, A)}{\sqrt{2D\tau}Y} Y \Phi(Y) dY \\ + \int_0^\infty \frac{\tau \varepsilon^\tau(x - \sqrt{2D\tau}Y, t, A) - \tau \varepsilon^\tau(x + \sqrt{2D\tau}Y, t, A)}{\sqrt{2D\tau}Y} Y \Phi(Y) dY. \quad (26)$$

The right-hand side of (26) tends to  $D\partial_x P_m^{aux}$  in the hydrodynamic limit. In particular, we have ensured through Assumption **(H<sub>3</sub>)** that the rate of convergence of  $P_m^{\tau, aux}$  to  $P_m^{aux}$  and  $\varepsilon^\tau$  to 0 is faster than  $\mathcal{O}(\tau^{1/2})$ . Thus, for instance,  $P_m^{\tau, aux}/\sqrt{\tau} = P_m^{aux}/\sqrt{\tau} + (P_m^{\tau, aux} - P_m^{aux})/\sqrt{\tau}$  behaves like  $P_m^{aux}/\sqrt{\tau}$ . Hence, letting  $\tau \rightarrow 0$  in contributions (25) and (26), we get straightforward the  $x$ -component of the flux  $\mathcal{F}$  of walkers. It is given by the following advective Fick's law applied to  $P_m^{aux}$ :

$$\mathcal{F}_x = -D\partial_x P_m^{aux} + \Lambda_v v P_m^{aux}.$$

Since  $u$  is nonnegative, immobile particles whose third coordinate becomes larger than  $A$  during  $[t, t + dt]$  fill the cylinder of basis  $u(x)dt$ . Corresponding mobile particles fill the cylinder of basis  $\int_0^{dt} u(x - vs)ds$ . Nevertheless, this latter integral asymptotically behaves as  $u(x)dt$  as  $dt \rightarrow 0$ . Finally, the  $A$ -component of the flux is thus analogous to a convective term for all particles:

$$\mathcal{F}_A = uP.$$

Finally, we use mass conservation principle with fluxes  $\mathcal{F}_x$  and  $\mathcal{F}_A$  to derive the Feynman-Kac type equation governing the system in the hydrodynamic limit. We obtain

$$\partial_t P + \partial_x (-D\partial_x P_m^{aux} + \Lambda_v v P_m^{aux}) + \partial_A(uP) = r, \quad (27)$$

function  $r$  being the source term. Combining the latter equation with the results of Propositions 2 and 3, we state the following result.

**Theorem 3**

(i) Assume  $\gamma = \gamma_1 < 1$ . Transport is anomalous (subdiffusive). Feynman-Kac equation corresponding to the limit fractal MIM is (1), that is:

$$\partial_t P - D\partial_{xx}^2 \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P + \Lambda_v \partial_x (v \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P) + u \partial_A P = r.$$

(ii) Assume  $\gamma_1 < \gamma$  and  $\gamma_1 < 1$ . The trapping process asymptotically has no influence. We observe normal diffusion and we recover the classical Feynman-Kac equation:

$$\Lambda_v \partial_t P - D\partial_{xx}^2 P + \Lambda_v v \partial_x P + \Lambda_v u \partial_A P = \Lambda_v r.$$

(iii) Assume  $\gamma_1 = 1 < \gamma$ . We observe normal diffusion with a retardation factor  $R_\gamma$  ( $R_\gamma \rightarrow 0$  as  $\gamma \rightarrow \infty$ ). Corresponding Feynman-Kac equation reads:

$$(1 + R_\gamma) \partial_t P - D\partial_{xx}^2 P + \Lambda_v v \partial_x P + (1 + R_\gamma) \partial_A (uP) = (1 + R_\gamma) r.$$

Note that the difference with item (ii) comes from the intensity of heterogeneity leading to trapping process via parameter  $\gamma$ .

(iv) The other scalings choices lead to an asymptotically immobile tracer.

We emphasize in particular that a divergent distribution of waiting times does not imply automatically a non-Fickian behavior in the hydrodynamic limit (see all the results for  $\gamma < 1$ ). Anomalous behavior lies on a precise relation between the scale separation parameters.

**Remark 8 (Fractal MIM versus FFPE)** As already noted in Remark 2, the usual microscopic context leading to FFPE corresponds to setting  $\Lambda_v = 0$  in the definition of our random walk. This is consistent with the asymptotic behaviors stated above. Indeed, all our convergence results are proven for any  $\Lambda_v \geq 0$ . In the particular case  $\Lambda_v = 0$ , all the translation operators reduce to identity and point (i) of Theorem 3 reduces to:

$$\partial_t P - D\partial_{xx}^2 (\Lambda \mathcal{T}_{\cdot, tu(\cdot, x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-\cdot, tu(\cdot, x)}^A)^{-1} P + u \partial_A P = r,$$

that is

$$\begin{aligned} \partial_t P(x, t, A) - D\partial_{xx}^2 (\Lambda^{-1} \mathcal{T}_{tu(x)}^A D_{0,+}^{1-\gamma} \mathcal{T}_{-\cdot, tu(x)}^A) P(x, t, A) \\ + u(x) \partial_A P(x, t, A) = r(x, t, A). \end{aligned}$$



Operator  $D_{0,+}^{1-\gamma}$  is the Riemann-Liouville derivative of order  $1-\gamma$ , that is a left inverse of the fractional integral  $I_{0,+}^{1-\gamma}$ . This latter equation is Eq. (11) of [9]. Hence FFPE appears as a special case of our derivation. Furthermore, we prove in the next subsection that letting  $\Lambda_v \rightarrow 0$  in (1) let us recover in some sense the FFPE formulation (4).

## 4 Scaling arguments, occurrence of anomalous versus normal transport

The present section is devoted to a physical interpretation of the scaling parameters involved in the present work. We aim to consider the most complete case where convection, diffusion and trapping occur. For the sake of clarity, we thus set  $\Lambda_v = 1$ .

An obvious scaling in our microscopic setting is linked with the time step  $\tau$ . By assuming  $\tau \rightarrow 0$ , we suppose that the observation time (or “clock time”) is much larger than other temporal scales present in the system. We also assume that  $v$  is a mesoscopic velocity and that the characteristic local Péclet number  $\mathbf{Pe}$  of the random walk model, defined as the rate between characteristic diffusive time  $T_D = \ell^2/D$  to the characteristic convective time  $T_c = \tau$ , satisfies

$$\mathbf{Pe} = \frac{T_D}{T_c} = \frac{\ell^2}{\tau D} = \mathcal{O}(1).$$

In the next subsections we precise the importance of the other scale parameters, that is of  $\gamma$ ,  $\gamma_1$  and  $\Lambda$  introduced in Assumptions  $(\mathbf{H}_1, \mathbf{H}'_1)$  on waiting times. We restrict this analysis to the non-classical context with stable law of exponent  $\gamma < 1$  as in assumption  $(\mathbf{H}_1^{\text{long}})$ .

### 4.1 Parameters $\gamma$ and $\gamma_1$ , observation time and heterogeneity scale

Assume a power law decrease as  $t^{-\gamma-1}$  for the pdf  $\psi$  of  $n$  independent random variables  $W_1, \dots, W_n$  distributed as  $t_w^*$  and representing successive stagnation times for a tagged particle. Stable laws (here of exponent  $\gamma$ ) play a central role within this context and cannot be regarded as being a simple particular case. Indeed they are attractors. Moreover, focusing on stable laws allows us to give sense to parameters  $\gamma$  and  $\gamma_1$  through essentially observation time and heterogeneity scale (itself described by  $\gamma$ ).

To this aim, let us compare the “clock time” with the “operational time”. The operational time is a process  $S(t)$  defined as the time spent by the walker

in the mobile phase before clock time  $t$ . It is easier and more natural to compute the statistics of its inverse process  $T(s)$ , expressing the observable clock time with regard to operational time. We can think of  $T(\cdot)$  as an unscaled, order one quantity. A tagged walker performing the above random walk ends the step of rank  $n$  (*i.e.* after  $n$  units of microscopic time) at macroscopic time  $t$

$$t = T(n\tau) = n\tau + \tau^{1/\gamma_1}(W_1 + \dots + W_n). \quad (28)$$

The stable Levy process  $W_t$  with the stability index  $\gamma$  being self-similar, we have  $W_j \stackrel{d}{=} j^{1/\gamma}W_1$ . The latter random variable is thus distributed as

$$T(n\tau) = n\tau + \tau^{1/\gamma_1}n^{1/\gamma}W_1 \quad (29)$$

if  $W_i$  is a stable variable of exponent  $\gamma$  [17]. Here  $\gamma$  represents the heterogeneity of the stagnation process. Term  $n\tau$  is the time spent moving until observation time  $t$ . Only  $t$  can be measured whereas time  $n\tau$  ranges between  $t$  itself and 0, according to the value of  $\gamma_1/\gamma$ . Of course, since clock time  $t = T(n\tau)$  is of order one,  $n \rightarrow \infty$  as  $\tau \rightarrow 0$ . The rate of convergence of  $n$  to infinity is crucial.

Let us be more precise. Let  $T$  be the macroscopic reference time. For sake of clarity, we begin with the classical diffusive random walk without trapping effects, that is when  $t = T(n\tau) = n\tau$ . In this case, it is well-known that the central limit theorem applies when choosing

$$n = n_D = \left[ \frac{T}{\tau} \right]$$

where  $[\cdot]$  denotes the integer part, leading to a diffusive limit as  $\tau \rightarrow 0$ . This latter choice for  $n$  is the classical diffusive scaling. Note that we use a renormalization by  $\tau^{-1}$  and not by  $n$ , therefore the limit depends on  $T$  (but of course not on  $\tau$ ). In the present paper, we consider the latter diffusive process coupled with a trapping process. We thus may choose either a diffusive scaling  $n_D$  or a trapping scaling  $n_{trap}$ . As emphasized by (29),  $n_D$  and  $n_{trap}$  are respectively such that  $n_D\tau = \mathcal{O}(1)$  and  $n_{trap}^{1/\gamma}\tau^{1/\gamma_1} = \mathcal{O}(1)$ . Since  $T = \mathcal{O}(1)$ , the previous choices induce of course some conditions on the scale separation parameters, and especially on the trapping scale parameter  $\gamma_1$  with regard to the trapping intensity parameter  $\gamma$ . More precisely, we

have

$$\begin{aligned}
\text{purely diffusive scaling :} & \quad n = n_D = \left\lceil \frac{T}{\tau} \right\rceil \quad \text{and } \gamma_1 < \gamma; \\
\text{purely trapping scaling :} & \quad n = n_{trap} = \left\lceil \frac{T^\gamma}{\tau^{\gamma/\gamma_1}} \right\rceil \quad \text{and } \gamma_1 > \gamma; \\
\text{no scale separation between} & \\
\text{trapping and diffusion :} & \quad n = n_D = n_{trap} \quad \text{and } \gamma_1 = \gamma.
\end{aligned} \tag{30}$$

More intuitively, on the (microscopic) scale of walkers,  $\gamma_1$  helps us comparing the time spent in the immobile phase with the time spent moving for each individual step of the random walk. For any given sampling of the  $W_i$ s, the fraction of observation time  $t = T(n\tau) = n\tau + (n\tau)^{1/\gamma} \tau^{1/\gamma_1 - 1/\gamma} W_1$  spent in the immobile phase becomes negligible when  $\tau \rightarrow 0$  if we assume  $\gamma_1 < \gamma$ , because  $n\tau < T(n\tau) = t$  and  $\tau^{1/\gamma_1 - 1/\gamma} \rightarrow 0$ . In this case, the influence of the heterogeneity disappears at the limit (see Theorem 3 (ii)). Assuming oppositely  $\gamma_1 > \gamma$  implies that the “mobile time” has to tend to zero in the hydrodynamic limit in order to keep quantity  $(n\tau)^{1/\gamma} \tau^{1/\gamma_1 - 1/\gamma}$  finite. Hence stagnation dominates in this case, and there is no transport at all on the macroscopic scale (see Theorem 3 (iv)).

In short we calibrate the operational time with regard to the heterogeneity degree of the stagnation process given by  $\gamma$  thanks to parameter  $\gamma_1$ .

## 4.2 Return to a Fickian behavior

In the present subsection, we show that the anomalous behavior is a temporary phenomenon. More precisely we check that the long time behavior is always Fickian, even if we use parameters  $\psi$ ,  $\gamma$  and  $\gamma_1$  leading to anomalous diffusion as in the former section. Our aim is not to study the asymptotic behavior of the anomalous pde model (1) (as in [31]). We rather are going to study our random walk model for larger observation times.

Our arguments are based once again on the comparison of clock time and operational time processes and on an appropriate large time scaling. We recall that the time scale separation parameters  $\tau$  and  $\tau_w$  of the present problem have been defined in section 2 by  $t_{micro} = \tau^{-1} t_{macro}$  and  $t_{micro}^{trap} = (\tau_w(\tau))^{-1} t_{macro}^{trap}$ , function  $\tau_w$  being defined by  $\tau_w(\tau) = \tau^{1/\gamma_1}$ . We now assume

$$\gamma = \gamma_1.$$

Indeed we have proved in the former sections that assuming  $\gamma = \gamma_1$  and the diffusive scaling  $n = \lceil T/\tau \rceil$  leads to an anomalous behavior at times of order  $T = \mathcal{O}(1)$ ,  $T$  corresponding to the clock time defined in (29) by

$$T(n\tau) = n\tau + n^{1/\gamma} \tau_w W_1 = n\tau + (n\tau)^{1/\gamma} W_1.$$

Now, imagine that we track the behavior of this random walk process with trapping after  $T$ , more precisely until time  $T_1 \gg T$ . In view of going back to time scales of order 1, we rescale the time variable by setting

$$T_1 = T_R T.$$

We thus have to rescale the time scale separation parameters. We first set

$$\tau_1 = \tau/T_R$$

(intuitively this means that we use shorter time steps to mimic a long time study even if the final time  $T$  is of order 1). This corresponds to the scaling  $t_{micro} = \tau^{-1} t_{1,macro} = \tau^{-1} T_R t_{macro}$ . Function  $\tau_w$  characterizing the trapping time scale separation is scaled similarly by  $1/T_R$ . We get

$$\tau_{1,w} = \frac{\tau_w(\tau_1)}{T_R} = \left(\frac{\tau}{T_R}\right)^{1/\gamma} \frac{1}{T_R}.$$

Corresponding rescaled clock time reads

$$T = n \frac{\tau}{T_R} + \left(n \frac{\tau}{T_R}\right)^{1/\gamma} \frac{1}{T_R} W_1. \quad (31)$$

Classical diffusive scaling consists in assuming  $n = [TT_R/\tau]$ .

Choosing  $T_R = \mathcal{O}(1)$  we recover of course at the limit the non-Fickian model (1). In view of tracking the behavior of this random walk model until  $T_1 \gg T$  we let  $T_R \rightarrow \infty$ . In (31) the trapping part  $(n\tau/T_R)^{1/\gamma} T_R^{-1} W_1$  of the clock time becomes obviously negligible as  $T_R \rightarrow \infty$ . We thus observe a return to the Fickian behavior, *i.e.* a classical advection-diffusion equation. Following the lines of Section 3 gives a rigorous derivation of this result.

### 4.3 Parameter $\Lambda$ , intensity of the trapping process. Weak coupling limit.

In the present subsection we focus on the rule of parameter  $\Lambda$ . We illustrate that this parameter does not influence the nature of the transport but the intensity of the anomalous behavior.

After the mobile period of rank  $n$ , the walker is immobilized during time  $\tau^{1/\gamma_1} W_n$  where the random variable  $W_n$  belongs to the attraction domain of a maximally skewed Levy law of exponent  $\gamma$ . More precisely, the pdf of  $W_n$  is defined in Assumption  $(\mathbf{H}_1^{\text{long}})$  which involves a parameter  $\Lambda \geq 0$ .

To highlight the rule of  $\Lambda$ , we choose two “toy” models for the trapping process.

1. *Reactive barriers.* Particles are retained by adsorption phenomena. If we increase sufficiently the observation time, the reactions become instantaneous and the trapping process is no more detectable.
2. *Presence of residence branches.* Assume trapping is due to presence of side branches as in the example developed in Annex A. Once again, if we increase sufficiently the observation time and keep the same order of time displacement in the branches, trapping becomes negligible.

The mechanism described in the two latter examples consists basically in assuming  $\Lambda = \Lambda(\tau)$ ,  $\Lambda(\tau) \rightarrow 0$  as  $\tau \rightarrow 0$ , and studying the weak coupling limit of the process (2.2) as  $\tau \rightarrow 0$ . We then recover a classical advection-diffusion model. Such weak coupling limit is comparable to what is done for Hamiltonian systems with a random Hamiltonian to preserve Markovity (see for instance [21, 16]).

Note finally that we may also include in  $\Lambda$  a trapping probability by replacing  $\Lambda$  by some space function of type  $\Lambda h(x)$  (see [38]).

## 5 Mathematical analysis of equation (1). Convergence of the fractal MIM formulation to the FFPE formulation

The present section is devoted to the mathematical analysis of the fractional pde (1), that is

$$\partial_t P - D\partial_{xx}^2 \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P + \Lambda_v \partial_x (v \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P) + u \partial_A P = r, \quad (32)$$

$$\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma = (\Lambda_v \text{Id} + \Lambda \mathcal{T}_{\cdot t u(\cdot x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-\cdot t u(\cdot x)}^A)^{-1}, \quad \gamma < 1. \quad (33)$$

We recall that this is the Feynman-Kac equation associated to the MIM derivation (see Theorem 3). Nevertheless, this equation is strongly connected with the one associated with the FPE approach:

$$\begin{aligned} \partial_t P^*(x, t, A) - D\partial_{xx}^2 (\Lambda^{-1} \mathcal{T}_{tu(x)}^A D_{0,+}^{1-\gamma} \mathcal{T}_{-\cdot t u(x)}^A) P^*(x, t, A) \\ + u(x) \partial_A P^*(x, t, A) = r(x, t, A). \end{aligned} \quad (34)$$

We have already mentioned in Remark 8 that the FFPE above corresponds to the limiting process of our random walk if  $\Lambda_v = 0$ . Here we also prove that solutions of (32)-(33) converge to a solution of (34) as  $\Lambda_v \rightarrow 0$ .

Let  $(0, T)$ ,  $T > 0$ , be the time interval of interest. Our first aim is the study of (32)-(33) for  $t \in (0, T)$ ,  $x \in (0, \infty)$ ,  $A \in (0, \infty)$ . For the mathematical analysis, we complete the equations with the following initial and boundary conditions:

$$P|_{t=0} = P^0, \quad P|_{x=0} = 0, \quad P|_{A=0} = 0, \quad (35)$$

with the compatibility conditions  $P^0|_{x=0} = 0$  and  $P^0|_{A=0} = 0$ . We focus on the parabolic in space setting by assuming a positive diffusion coefficient  $D > 0$ . We state the following result of existence.

**Theorem 4** *Assume  $P^0 \in H^2(\mathbb{R}_+^2)$ . Assume  $r \in L^2((0, T) \times \mathbb{R}_+^2)$ . There exists a unique weak solution  $P$  to the problem (32)-(33), (35). Specifically, we have*

$$P \in L^\infty(0, T; L^2(\mathbb{R}_+^2)), \quad \partial_x P \in L^2((0, T) \times \mathbb{R}_+^2).$$

We set

$$g_\gamma(t) = \frac{t^{-\gamma}}{\Gamma(1-\gamma)}, \quad t > 0.$$

Since  $\gamma < 1$ ,  $g_\gamma \in L^1(0, T)$ . We recall that  $I_{0,+}^{1-\gamma}$  is defined by

$$I_{0,+}^{1-\gamma}[g] = g_\gamma * g.$$

For the proof of Theorem 4, we recall the following results (see [28, 20]).

**Lemma 5** *Let  $\mathcal{X}$  be a Hilbert space (with scalar product denoted by  $\langle \cdot, \cdot \rangle$ ). Let  $v \in \mathcal{C}([0, T]; \mathcal{X})$  such that  $\partial_t v \in W^{(\gamma-1)/2, 2}(0, T; \mathcal{X})$ . The following relations hold true:*

$$(i) \quad \int_0^T \langle (g_\gamma * \partial_t v)(t), v(t) \rangle dt \geq \frac{1}{2} \int_0^T g_\gamma(t) (\|v(T-t)\|_{\mathcal{X}}^2 + \|v(t)\|_{\mathcal{X}}^2) dt \\ - \int_0^T g_\gamma(t) \langle v(0), v(t) \rangle dt;$$

$$(ii) \quad \int_0^T \langle \partial_t (g_\gamma * \partial_t v)(t), v(t) \rangle dt \geq \frac{1}{2} \int_0^T g_\gamma(t) (\|v(T-t)\|_{\mathcal{X}}^2 + \|v(t)\|_{\mathcal{X}}^2) dt.$$

There exists  $c > 0$  and  $\bar{g} \in L^1(\mathbb{R}_+)$ ,  $\bar{g}(t) = t^{-1+(1-\gamma)/2} e^{-t}$ , such that

$$(iii) \quad \int_0^T \langle (g_\gamma * v)(t), v(t) \rangle dt \geq \int_0^T \|\bar{g} * v\|_{\mathcal{X}}^2 dt;$$

$$\begin{aligned}
& (iv) \quad \int_0^T \langle \partial_t(g_\gamma * v)(t), \partial_t v(t) \rangle dt \geq \int_0^T \langle (g_\gamma * \partial_t v)(t), \partial_t v(t) \rangle dt \\
& + \int_0^T \langle g_\gamma(t)v(0), \partial_t v(t) \rangle dt \geq \int_0^T \|\bar{g} * \partial_t v\|_{\mathcal{X}}^2 dt + \int_0^T \langle g_\gamma(t)v(0), \partial_t v(t) \rangle dt ; \\
& (v) \quad \int_0^T \langle (g_\gamma * \partial_t v)(t), \partial_t v(t) \rangle dt = \frac{1}{2} \int_0^T \int_0^t \|v(t-s) - v(s)\|_{\mathcal{X}}^2 g'_\gamma(s) ds dt \\
& \quad - \int_0^T g'_\gamma(s) (\|v(s) - v(0)\|_{\mathcal{X}}^2 + \|v(T-s) - v(T)\|_{\mathcal{X}}^2) ds \\
& \quad \quad + g_\gamma(T) \|v(T) - v(0)\|_{\mathcal{X}}^2.
\end{aligned}$$

**Proof of Theorem 4.** Inspired by the derivation of the model, we define  $P_m^{aux}$  by

$$P_m^{aux} = \mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma P.$$

Instead of studying directly (32)-(33), we consider the following equivalent equation:

$$\begin{aligned}
& \Lambda_v \partial_t P_m^{aux} + \Lambda \partial_t [\mathcal{T}_{u(x)t}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x)\cdot t}^A] P_m^{aux} + \Lambda_v v \partial_x P_m^{aux} - D \partial_{xx}^2 P_m^{aux} \\
& \quad + \Lambda_v u \partial_A P_m^{aux} + \Lambda u \partial_A [\mathcal{T}_{u(x)t}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x)\cdot t}^A] P_m^{aux} = r. \quad (36)
\end{aligned}$$

This equation seems more cumbersome than the original one but it is actually more adapted to the exploitation of known mathematical results. We note that, at least formally,

$$\begin{aligned}
& \partial_t [\mathcal{T}_{u(x)t}^A g(t, A)] = \partial_t g(t, A - tu(x)) - u(x) \partial_A g(t, A - tu(x)) \\
& \quad = \mathcal{T}_{u(x)t}^A \partial_t g(t, A) - u(x) \mathcal{T}_{u(x)t}^A \partial_A g(t, A).
\end{aligned}$$

Equation (36) thus reduces to

$$\begin{aligned}
& \Lambda_v \partial_t P_m^{aux} + \Lambda \partial_t [I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x)\cdot t}^A] P_m^{aux}(t, x, A - tu(x)) \\
& \quad + \Lambda_v v \partial_x P_m^{aux} - D \partial_{xx}^2 P_m^{aux} = r. \quad (37)
\end{aligned}$$

It is completed with the following boundary and initial conditions issued from (35):

$$P_m^{aux}|_{t=0} = P_m^0 = \Lambda_v^{-1} P^0, \quad P_m^{aux}|_{x=0} = 0, \quad P_m^{aux}|_{A=0} = 0. \quad (38)$$

We recognize in (37) the structure of an integro-differential Volterra equation. For basic facts on this type of equations, we refer to the monograph

[20]. Mathematical difficulty is here inferred by the singularity of the kernel  $g_\gamma$ . Using the time derivative of order  $\gamma$ ,  $\partial_t^\gamma \cdot = \frac{d}{dt}(g_\gamma * \cdot)$ , equation (37) also reads

$$\begin{aligned} & \Lambda_v \partial_t P_m^{aux}(t, x, A) + \Lambda \partial_t^\gamma (\mathcal{T}_{-u(x), t}^A (P_m^{aux} - P_m^0))(t, x, A - tu(x)) \\ & + \Lambda_v v \partial_x P_m^{aux}(t, x, A) - D \partial_{xx}^2 P_m^{aux}(t, x, A) = r(t, x, A) - \Lambda \partial_t^\gamma P_m^0. \end{aligned} \quad (39)$$

During the last decade, mathematical literature about evolutionary pdes involving fractional derivatives  $\partial_t^\gamma$  has considerably developed. We mention in particular [11] which develops global existence results of BUC solutions (even in quasilinear settings) through maximal regularity arguments for parabolic equations in the form

$$\partial_t^\gamma u + \mathcal{A}u = f(u) + h(t), \quad u|_{t=0} = u^0.$$

We also mention references [14, 46].

Occurrence of the (linear) translation operator  $\mathcal{T}_{-u(x), t}^A$  in (37) may be viewed as a slight perturbation of the general picture of [11]. Derivation of *a priori* estimates for the solutions of (37)-(38) is thus sufficient to ensure the existence of weak solutions. Nevertheless, we have to introduce a parabolic (in  $A$ ) perturbation of (37) by:

$$\begin{aligned} & \Lambda_v \partial_t P_m^\varepsilon + \Lambda \partial_t [I_{0,+}^{1-\gamma} \mathcal{T}_{-u_\varepsilon(x), t}^A] P_m^\varepsilon(t, x, A - tu_\varepsilon(x)) \\ & + \Lambda_v v \partial_x P_m^\varepsilon - D \partial_{xx}^2 P_m^\varepsilon - \varepsilon (u'_\varepsilon(x))^2 \partial_{AA}^2 P_m^\varepsilon = r, \end{aligned} \quad (40)$$

where  $\varepsilon > 0$  and  $u_\varepsilon$  is a derivable function such that  $u'_\varepsilon \in L^\infty(\mathbb{R}_+)$  and  $u_\varepsilon(x) \rightarrow u(x)$  a.e. in  $\mathbb{R}_+$  as  $\varepsilon \rightarrow 0$ . The latter equation also reads

$$\begin{aligned} & \Lambda_v \partial_t P_m^\varepsilon(t, x, A) + \Lambda \partial_t^\gamma (\mathcal{T}_{-u_\varepsilon(x), t}^A (P_m^\varepsilon - P_m^0))(t, x, A - tu_\varepsilon(x)) \\ & + \Lambda_v v \partial_x P_m^\varepsilon(t, x, A) - D \partial_{xx}^2 P_m^\varepsilon(t, x, A) - \varepsilon (u'_\varepsilon(x))^2 \partial_{AA}^2 P_m^\varepsilon \\ & = r(t, x, A) - \Lambda \partial_t^\gamma P_m^0. \end{aligned} \quad (41)$$

For the first set of estimates, we note that  $[\mathcal{T}_{-u_\varepsilon(x), t}^A P_m^\varepsilon](t, x, A - tu_\varepsilon(x)) = P_m^\varepsilon(t, x, A)$ . Thus, multiplying (40) by  $P_m^\varepsilon$  and integrating by parts in



$(0, T) \times \mathbb{R}_+^2$ , we get for any  $T_o \leq T$ :

$$\begin{aligned}
& \frac{\Lambda_v}{2} \int_0^{T_o} \frac{d}{dt} \|P_m^\varepsilon\|_{L^2(\mathbb{R}_+^2)}^2 dt \\
& + \frac{\Lambda}{2} \int_0^{T_o} g_\gamma(t) (\|P_m^\varepsilon(T_o - t)\|_{L^2(\mathbb{R}_+^2)}^2 + \|P_m^\varepsilon\|_{L^2(\mathbb{R}_+^2)}^2) dt \\
& + D \int_0^{T_o} \|\partial_x P_m^\varepsilon\|_{L^2(\mathbb{R}_+^2)}^2 dt + \varepsilon \int_0^{T_o} \|\sqrt{u'_\varepsilon} \partial_A P_m^\varepsilon\|_{L^2(\mathbb{R}_+^2)}^2 dt \\
& + \frac{\Lambda_v}{2} \int_0^{T_o} \int_{\mathbb{R}_+^2} v \partial_x ((P_m^\varepsilon)^2) dt dx dA + \frac{\Lambda_v}{2} \int_0^{T_o} \int_{\mathbb{R}_+^2} u_\varepsilon \partial_A ((P_m^\varepsilon)^2) dt dx dA \\
& \leq \int_{(0, T_o) \times \mathbb{R}_+^2} r P_m^\varepsilon dt dx dA.
\end{aligned}$$

We compute

$$\frac{\Lambda_v}{2} \int_{(0, T_o) \times \mathbb{R}_+^2} v \partial_x ((P_m^\varepsilon)^2) dt dx dA = -\frac{\Lambda_v v}{2} \int_{(0, T_o) \times \mathbb{R}_+} |P_m^\varepsilon|_{x=0}|^2 dt dA = 0,$$

and

$$\frac{\Lambda_v}{2} \int_{(0, T_o) \times \mathbb{R}_+^2} u_\varepsilon \partial_A ((P_m^\varepsilon)^2) dt dx dA = -\frac{\Lambda_v}{2} \int_{(0, T_o) \times \mathbb{R}_+} u_\varepsilon |P_m^\varepsilon|_{A=0}|^2 dt dA = 0.$$

The three later relations give

$$\begin{aligned}
& \frac{\Lambda_v}{2} \|P_m^\varepsilon(T_o, \cdot, \cdot)\|_{L^2(\mathbb{R}_+^2)}^2 \\
& + \frac{\Lambda}{2} \int_0^{T_o} g_\gamma(t) (\|P_m^\varepsilon(T_o - t)\|_{L^2(\mathbb{R}_+^2)}^2 + \|P_m^\varepsilon(t)\|_{L^2(\mathbb{R}_+^2)}^2) dt \\
& + D \int_0^{T_o} \|\partial_x P_m^\varepsilon\|_{L^2(\mathbb{R}_+^2)}^2 dt + \varepsilon \int_0^{T_o} \|\sqrt{u'_\varepsilon} \partial_A P_m^\varepsilon\|_{L^2(\mathbb{R}_+^2)}^2 dt \\
& \leq C(\|r\|_{L^2}, \|\Lambda_v P_m^0\|_{L^2}) = C,
\end{aligned}$$

where  $C$  is a generic constant, not depending on  $\varepsilon$  neither on  $\Lambda_v$ . We infer from the latter relation the following estimates:

$$\begin{cases} \|\sqrt{\Lambda_v} P_m^\varepsilon\|_{L^\infty(0, T; L^2(\mathbb{R}_+^2))} \leq C, \\ \|\partial_x P_m^\varepsilon\|_{L^2(0, T; L^2(\mathbb{R}_+^2))} \leq C, \\ \|g_\gamma^{1/2} P_m^\varepsilon\|_{L^2(0, T; L^2(\mathbb{R}_+^2))} \leq C. \end{cases} \quad (42)$$

Note that these estimates remain true if  $\varepsilon = 0$ .

For the second set of estimates, we aim using the following point (see Lemma 5):

$$\begin{aligned} & \int_0^T \langle \partial_t(g_\gamma * v), \partial_t v \rangle dt \\ & \geq \int_0^T \|\bar{g} * \partial_t v\|_{L^2(\mathbb{R}_+^2)} dt + \int_0^T \langle g_\gamma v|_{t=0}, \partial_t v \rangle dt, \end{aligned}$$

which implies

$$\begin{aligned} & \int_0^T \langle \partial_t(g_\gamma * v)(t, x, A - tu(x)), \partial_t v(t, x, A - tu(x)) \rangle dt \\ & \geq \int_0^T \|\bar{g} * \partial_t v(t, x, A - tu(x))\|_{L^2(\mathbb{R}_+^2)} dt \\ & \quad + \int_0^T \langle g_\gamma(t)v(0, x, A), \partial_t v(t, x, A - tu(x)) \rangle dt. \end{aligned}$$

We are going to apply this relation for  $v(t, x, A) = (\mathcal{T}_{-u_\varepsilon(x), t}^A P_m^\varepsilon)(t, x, A) = P_m^\varepsilon(t, x, A + u_\varepsilon(x)t)$ . Its time derivative satisfies  $\partial_t v(t, x, A) = \partial_t P_m^\varepsilon(t, x, A + u_\varepsilon(x)t) + u_\varepsilon(x) \partial_A P_m^\varepsilon(t, x, A + u_\varepsilon(x)t)$  and  $\partial_t v(t, x, A - u_\varepsilon(x)t) = \partial_t P_m^\varepsilon(t, x, A) + u_\varepsilon(x) \partial_A P_m^\varepsilon(t, x, A)$ . We thus multiply equation (41) by

$$\begin{aligned} & \partial_t P_m^\varepsilon(t, x, A) - \partial_t(P_m^0(x, A - u_\varepsilon(x)t)) \\ & + u_\varepsilon(x) \partial_A P_m^\varepsilon(t, x, A) - u_\varepsilon(x) \partial_A(P_m^0(x, A - u_\varepsilon(x)t)). \end{aligned}$$

We get, for any  $T_o \leq T$ ,

$$\begin{aligned}
& \Lambda_v \int_{(0, T_o) \times \mathbb{R}_+^2} (|\partial_t P_m^\varepsilon|^2 + 2u_\varepsilon \partial_t P_m^\varepsilon \partial_A P_m^\varepsilon + |u_\varepsilon \partial_A P_m^\varepsilon|^2) \\
& - \int_{(0, T_o) \times \mathbb{R}_+^2} (\partial_t P_m^\varepsilon + u_\varepsilon \partial_A P_m^\varepsilon) (\partial_t (P_m^0(x, A - u_\varepsilon(x)t)) + u_\varepsilon \partial_A P_m^0) \\
& + \int_{(0, T_o) \times \mathbb{R}_+^2} |\bar{g} * (\partial_t P_m^\varepsilon + u_\varepsilon \partial_A P_m^\varepsilon - \partial_t (P_m^0(x, A - u_\varepsilon(x)t)) - u_\varepsilon \partial_A P_m^0)|^2 \\
& + D \int_{(0, T_o) \times \mathbb{R}_+^2} \partial_x P_m^\varepsilon \partial_{tx}^2 P_m^\varepsilon + D \int_{(0, T_o) \times \mathbb{R}_+^2} u_\varepsilon \partial_x P_m^\varepsilon \partial_{Ax}^2 P_m^\varepsilon \\
& + D \int_{(0, T_o) \times \mathbb{R}_+^2} u'_\varepsilon \partial_x P_m^\varepsilon \partial_A P_m^\varepsilon \\
& - D \int_{(0, T_o) \times \mathbb{R}_+^2} \partial_x P_m^\varepsilon (\partial_{tx}^2 (P_m^0(x, A - u_\varepsilon(x)t)) + u_\varepsilon \partial_{xA}^2 P_m^0 + u'_\varepsilon \partial_A P_m^0) \\
& + \varepsilon \int_{(0, T_o) \times \mathbb{R}_+^2} (u'_\varepsilon)^2 \partial_A P_m^\varepsilon \partial_{tA}^2 P_m^\varepsilon + \varepsilon \int_{(0, T_o) \times \mathbb{R}_+^2} u_\varepsilon (u'_\varepsilon)^2 \partial_A P_m^\varepsilon \partial_{AA}^2 P_m^\varepsilon \\
& - \varepsilon \int_{(0, T_o) \times \mathbb{R}_+^2} (u'_\varepsilon)^2 \partial_A P_m^\varepsilon \partial_A (\partial_t (P_m^0(x, A - u_\varepsilon(x)t)) + u_\varepsilon \partial_A P_m^0) \\
& + \Lambda_v \int_{(0, T_o) \times \mathbb{R}_+^2} v \partial_x P_m^\varepsilon (\partial_t P_m^\varepsilon + u \partial_A P_m^\varepsilon - \partial_t (P_m^0(x, A - u_\varepsilon(x)t)) - u_\varepsilon \partial_A P_m^0) \\
& \leq \int_{(0, T_o) \times \mathbb{R}_+^2} (r - \Lambda \partial_t^\gamma P_m^0) (\partial_t P_m^\varepsilon + u \partial_A P_m^\varepsilon \\
& \quad - \partial_t (P_m^0(x, A - u_\varepsilon(x)t)) - u_\varepsilon \partial_A P_m^0). \tag{43}
\end{aligned}$$

The first term reads  $\Lambda_v \int_{(0, T_o) \times \mathbb{R}_+^2} |\partial_t P_m^\varepsilon + u_\varepsilon \partial_A P_m^\varepsilon|^2$ . We also write

$$\begin{aligned}
D \int_{(0, T_o) \times \mathbb{R}_+^2} \partial_x P_m^\varepsilon \partial_{tx}^2 P_m^\varepsilon &= \frac{D}{2} \int_0^{T_o} \frac{d}{dt} \int_{\mathbb{R}_+^2} |\partial_x P_m^\varepsilon|^2 \\
&= \frac{D}{2} \int_{\mathbb{R}_+^2} |\partial_x P_m^\varepsilon(T_o, \cdot, \cdot)|^2 - \frac{D}{2} \int_{\mathbb{R}_+^2} |\partial_x P_m^0|^2;
\end{aligned}$$

$$D \int_{(0, T_o) \times \mathbb{R}_+^2} \partial_x P_m^\varepsilon \partial_{Ax}^2 P_m^\varepsilon = \frac{D}{2} \int_0^\infty \frac{d}{dA} \int_{(0, T_o) \times \mathbb{R}_+} |\sqrt{u_\varepsilon} \partial_x P_m^\varepsilon|^2 = 0;$$

$$\begin{aligned}
& \varepsilon \int_{(0,T_o) \times \mathbb{R}_+^2} (u'_\varepsilon)^2 \partial_A P_m^\varepsilon \partial_{tA}^2 P_m^\varepsilon = \frac{\varepsilon}{2} \int_0^{T_o} \frac{d}{dt} \int_{\mathbb{R}_+^2} |u'_\varepsilon \partial_A P_m^\varepsilon|^2 \\
& = \frac{\varepsilon}{2} \int_{\mathbb{R}_+^2} |u'_\varepsilon \partial_A P_m^\varepsilon(T_o, \cdot, \cdot)|^2 - \frac{\varepsilon}{2} \int_{\mathbb{R}_+^2} |u'_\varepsilon \partial_A P_m^0|^2; \\
& \varepsilon \int_{(0,T_o) \times \mathbb{R}_+^2} u_\varepsilon (u'_\varepsilon)^2 \partial_A P_m^\varepsilon \partial_{AA}^2 P_m^\varepsilon \\
& = \frac{\varepsilon}{2} \int_0^\infty \frac{d}{dA} \int_{(0,T_o) \times \mathbb{R}_+} |\sqrt{u_\varepsilon} u'_\varepsilon \partial_A P_m^\varepsilon|^2 = 0.
\end{aligned}$$

We estimate the other terms in (43) using (42) and the Cauchy-Schwarz and Young inequalities. We conclude with the Gronwall lemma that the following set of estimates hold true:

$$\begin{cases} \|\sqrt{\Lambda_v}(\partial_t P_m^\varepsilon + u_\varepsilon \partial_A P_m^\varepsilon)\|_{L^2(0,T;L^2(\mathbb{R}_+^2))} \leq C_\varepsilon, \\ \|\partial_x P_m^\varepsilon\|_{L^\infty(0,T;L^2(\mathbb{R}_+^2))} \leq C_\varepsilon, \\ \|\varepsilon^{1/2} \partial_A P_m^\varepsilon\|_{L^\infty(\mathbb{R}_+;L^2((0,T) \times \mathbb{R}_+))} \leq C_\varepsilon, \end{cases} \quad (44)$$

where  $C_\varepsilon$  is a generic constant depending on  $\varepsilon$ . We conclude that there exists a unique (thanks to the linearity) weak solution  $P_m^\varepsilon$  of (40), (38). Its regularity is described by the functional spaces appearing in (42) and (44).

Then, bearing in mind that (42) does not depend on  $\varepsilon$  and letting  $\varepsilon \rightarrow 0$ , we prove that the whole sequence  $(P_m^\varepsilon)$  weakly converges in the space  $L^\infty(0,T;L^2(\mathbb{R}_+^2)) \cap L^2(0,T;H^1(\mathbb{R}_+;L^2(\mathbb{R}_+^2)))$  to the unique solution  $P_m^{aux}$  of the linear problem (37)-(38). Using the equivalence between (37)-(38) and (32)-(33), (35), we get the result announced in the theorem.  $\square$

We note that the solution  $P$  of (32)-(33), (35), which is characterized by

$$P = \Lambda_v P_m^{aux} + \Lambda \mathcal{T}_{u(x),t}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-u(x),t}^A P_m^{aux}$$

with  $P_m^{aux}$  satisfying estimates (42), is such that

$$\begin{cases} \|\sqrt{\Lambda_v} P\|_{L^\infty(0,T;L^2(\mathbb{R}_+^2))} \leq C, \\ \|\partial_x P\|_{L^2(0,T;L^2(\mathbb{R}_+^2))} \leq C, \\ \|g_\gamma^{1/2} P\|_{L^2(0,T;L^2(\mathbb{R}_+^2))} \leq C. \end{cases}$$

In particular, the third estimate with the fact that we work on a finite time interval implies that

$$\|P\|_{L^2(0,T;L^2(\mathbb{R}_+^2))} = \|g_\gamma^{-1/2} (g_\gamma^{1/2} P)\|_{L^2(0,T;L^2(\mathbb{R}_+^2))} \leq CT^{\gamma/2} \leq C.$$

Then, if we associate to any  $\Lambda_v > 0$  the solution  $P^{\Lambda_v}$  of (32)-(33), (35), we construct a sequence  $(P^{\Lambda_v})$  bounded in  $L^2((0, T) \times \mathbb{R}_+^2)$  (with moreover  $(\partial_x P^{\Lambda_v})$  bounded in  $L^2((0, T) \times \mathbb{R}_+^2)$ ). There exists a subsequence, still denoted  $(P^{\Lambda_v})$  for convenience, and a function  $P^* \in L^2((0, T) \times \mathbb{R}_+^2)$  such that

$$\begin{aligned} P^{\Lambda_v} &\rightharpoonup P^* \text{ weakly in } L^2((0, T) \times \mathbb{R}_+^2), \\ \partial_x P^{\Lambda_v} &\rightharpoonup \partial_x P^* \text{ weakly in } L^2((0, T) \times \mathbb{R}_+^2). \end{aligned}$$

One may use the auxiliary sequence  $(P_m^{aux, \Lambda_v})$  to check easily that  $P^*$  is the unique solution of the linear FFPE problem (34)-(35). Thus the whole sequence  $(P^{\Lambda_v})$  converges to  $P^*$ . We have proven the following result.

**Theorem 5** *For any  $\Lambda_v > 0$ , let  $P^{\Lambda_v}$  be the solution of the fMIM problem (32)-(33), (35). As  $\Lambda_v \rightarrow 0$ , the sequence  $(P^{\Lambda_v})$  weakly converges in  $L^2((0, T) \times \mathbb{R}_+^2) \cap L^2(0, T; H_x^1(\mathbb{R}_+; L_A^2(\mathbb{R}_+)))$  to the solution  $P^*$  of the FFPE problem (34)-(35).*

## 6 Observable behavior

In fluid mechanics, many experiments consist in measuring the mean value of path integrals functionals for small scale motions of particles. For instance, Nuclear Magnetic Resonance (NMR) velocimetry measures magnetization signals of the form  $\langle e^{i \int_0^t g(t') x(t') dt'} \rangle$ , where  $x(t)$  represents the projection of water molecules trajectories along the magnetic field gradient direction. Other examples are occupation times that play an important role in many domains of physics, and the age of water in geophysics. The corresponding path integrals have the form  $A = \int_0^t u(x(t')) dt'$ , which also represents the displacement of a tagged fluid particle due to an average flow field  $u(\cdot) \vec{e}_y$  coupled to diffusive motion in the transverse direction.

In the critical case of anomalous diffusion corresponding to equation (1), we aim to characterize the behavior of the “observable” quantity  $O_u$  corresponding to a path integral of integrand  $u$ :

$$O_u = \left\langle e^{-\int_0^t u(x(t')) dt'} \right\rangle.$$

Expressing this quantity with regard to the density  $P$ , we recognize a  $A$ -Laplace transform evaluated in  $p = 1$ :

$$O_u(x, t) = \int_{\mathbb{R}_+} e^{-A} P(x, t, A) dA = \tilde{P}^A(x, t, p = 1) := \tilde{P}(x, t).$$

**Proposition 4** *Quantity  $\tilde{P}$  is governed by the following p.d.e. a.e. in  $\mathbb{R} \times (0, T)$ :*

$$\begin{aligned} \partial_t \tilde{P} - (D\partial_{xx}^2 - \Lambda_v v \partial_x) ((\Lambda_v \text{Id} + \Lambda J^{1-\gamma})^{-1} \tilde{P}) \\ + u \tilde{P} = u P|_{A=0} + \tilde{r}, \end{aligned} \quad (45)$$

where the memory operator  $J^{1-\gamma}$  is defined by

$$J^{1-\gamma} \tilde{P}(x, t) = \left\{ \left( \frac{(\cdot)_t^{-\gamma}}{\Gamma(1-\gamma)} e^{-u(x)\cdot_t} \right) * \tilde{P}(x, \cdot_t) \right\}(x, t),$$

and  $P|_{A=0}$  is a boundary condition for equation (1).

**Remark 9** *Note that  $J^{1-\gamma}$  is a short memory operator provided  $u(x) > 0$  a.e.  $x \in \mathbb{R}$ .*

**Proof.** Applying the  $A$ -Laplace transform computed in  $p = 1$  to equation (1), we get

$$\begin{aligned} \partial_t \tilde{P}(x, t) = (D\partial_{xx}^2 - \Lambda_v v \partial_x) ((\Lambda_v \text{Id} + \Lambda J^{1-\gamma})^{-1} \tilde{P}(x, t)) \\ - u(x) \tilde{P}(x, t) + u(x) P(x, t, A = 0) + \tilde{r}^A(x, t, p = 1), \end{aligned} \quad (46)$$

where operator  $(\Lambda_v \text{Id} + J^{1-\gamma})^{-1}$  is the  $A$ -Laplace transform computed in  $p = 1$  of  $\mathcal{H}_{\Lambda, \Lambda_v, u}^\gamma = (\Lambda_v \text{Id} + \Lambda \mathcal{T}_{\cdot_t u(x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-\cdot_t u(x)}^A)^{-1}$ . As already mentioned in Remark 5, the double Laplace symbol with respect to  $t$  and  $A$  (with Laplace variables  $\lambda$  and  $p$ ) of operator  $\mathcal{T}_{\cdot_t u(x)}^A I_{0,+}^{1-\gamma} \mathcal{T}_{-\cdot_t u(x)}^A$  is  $(\lambda + pu(x))^{\gamma-1}$  for any fixed  $x$ . Choosing  $p = 1$ , we conclude that the  $t$ -Laplace symbol of  $J^{1-\gamma}$  is  $(\lambda + u(x))^{\gamma-1}$ . We recognize a translation of the  $t$ -Laplace transform of a power-law operator. Applying the inverse  $t$ -Laplace transform, we conclude that

$$J^{1-\gamma} \tilde{P}(x, t) = \left\{ \left( \frac{(\cdot)_t^{-\gamma}}{\Gamma(1-\gamma)} e^{-u(x)\cdot_t} \right) * \tilde{P}(x, \cdot_t) \right\}(x, t).$$

□

## References

- [1] N. Agman and S. Rabinovich. Diffusive dynamics on potential energy surfaces: non equilibrium CO binding to heme proteins. *J. Chem. Phys.*, 92(10):7270–7286, 1992.
- [2] E.G. Altmann and T. Tel. Poincaré recurrences and transient chaos in systems with leaks. *Phys. Rev. E*, 79:016204, 2009.
- [3] E. Barkai, R. Metzler, and J. Klafter. From continuous time random walks to the fractional Fokker-Planck equation. *Phys. Rev. E*, 61:132–138, 2000.
- [4] D.A. Benson and M.M. Meerschaert. A simple and efficient random walk solution of multi-rate mobile/immobile mass transport equations. *Adv. Water Resources*, 32:532–539, 2009.
- [5] B. Berkowitz, A. Cortis, M. Dentz, and H. Scher. Modeling non-Fickian transport in geological formations as a continuous time random walk. *Rev. Geophys.*, 44:RG2003, 2006.
- [6] F. Boano, A.I. Packman, A. Cortis, R. Revelli, and L. Ridolfi. A continuous time random walk approach to the stream transport of solutes. *Water Resour. Res.*, 43:W10425, 2007.
- [7] M. Bromly and C. Hinz. Non-Fickian transport in homogeneous unsaturated repacked sand. *Water Resour. Res.*, 40:W07402, 2004.
- [8] R. Brown. A brief account of microscopical observations made in the months of June, July, and August 1827, on the particles contained in the pollen of plants, and on the general existence of active molecule in organic and inorganic bodies. *Philosophical Magazine*, 4:161–173, 1828.
- [9] S. Carmi, L. Turgeman, and E. Barkai. On distributions of functionals of anomalous diffusion paths. *J. Stat. Phys.*, 141:1071–1092, 2010.
- [10] C. Choquet and M.C. Néel. Rigorous derivation of Feynman-Kac equations for arrested dispersion. Technical report, Lab. MIA, Univ. La Rochelle, april 2013.
- [11] P. Clément, S.-O. Londen, and G. Simonett. Quasilinear evolutionary equations and continuous interpolation spaces. *J. Diff. Equ.*, 196:418–447, 2004.

- [12] G.A. Coon and D.L. Bernstein. Some properties of the double Laplace transformation. *Trans. Amer. Math. Soc.*, 74:135–176, 1953.
- [13] A. Cortis, Y. Chen, H. Scher, and B. Berkowitz. Quantitative characterization of pore-scale disorder effects on transport in homogeneous granular media. *Phys. Rev. E*, 70:041108, 2004.
- [14] S.D. Eidelman and A.N. Kochubei. Cauchy problem for fractional diffusion equations. *J. Diff. Equ.*, 199:211–255, 2004.
- [15] A. Einstein. Über die von der molekularkinetischen Theorie der Wärme geforderte Bewegung von in ruhenden Flüssigkeiten suspendierten Teilchen. *Ann. der Physik*, 17:549–560, 1905.
- [16] L. Erdos. Linear Boltzmann equation as the long time dynamics of an electron weakly coupled to a phonon field. *J. Stat. Phys.*, 107:1043–1128, 2002.
- [17] W. Feller. *An Introduction to Probability Theory and its Applications, Vol.II*. Wiley, New York, 1970.
- [18] R. Friedrich, F. Jenko, A. Baule, and S. Eule. Anomalous diffusion of inertial, weakly damped particles. *Phys. Rev. Lett.*, 96:230601, 2006.
- [19] S. Gheorghiu and M.O. Coppens. Heterogeneity explains features of anomalous thermodynamics and statistics. *PNAS*, 101:15852–15856, 2004.
- [20] G. Gripenberg, S.-O. Londen, and O. Staffans. *Volterra integrals and functional equations*. Cambridge University Press, Cambridge, 1990.
- [21] G. Papanicolaou H. Kesten. A limit theorem for stochastic acceleration. *Comm. Math. Phys.*, 78:19–63, 1980.
- [22] P. Hanggi, P. Talkner, and M. Borkovec. Reaction-rate theory, 50 years after Kramers. *Rev. Mod. Phys.*, 62:251–350, 1990.
- [23] S. Havlin and D. Ben Avraham. Diffusion in disordered media. *Adv. Phys.*, 51:187–292, 2002.
- [24] A. Hunt and R. Ewing. *Percolation theory for flow in porous media*. Lecture Notes in Physics, 771. Springer-Verlag, Berlin, 2009.
- [25] M. Kac. On distributions of certain Wiener functionals. *Trans. Am. Math. Soc.*, 65:1–13, 1949.



- [26] I. Kaj, L. Leskela, I. Norros, and V. Schmidt. Scaling limits for random fields with long-range dependence. *Ann. of Prob.*, 35(2):528–550, 2007.
- [27] P. Lévy. *Théorie de l'addition des variables aléatoires*. Gauthier-Villars Paris, 1937.
- [28] S.-O. Londen, H. Petzeltová, and J. Prüss. Global well-posedness and stability of a partial integro-differential equation with applications to viscoelasticity. *J. Evol. Equ.*, 3:169–201, 2002.
- [29] M. Magdziarz. Langevin picture of subdiffusion with infinitely divisible waiting times. *J. Stat. Phys.*, 763–772:016708, 2009.
- [30] M. Magdziarz, A. Weron, and J. Klafter. Equivalence of the fractional Fokker-Planck and subordinated Langevin equations: The case of time-dependent force. *Phys. Rev. E*, 77:036704, 2008.
- [31] A.J. Majda and P.R. Kramer. Simplified models for turbulent diffusion: theory, numerical modelling, and physical phenomena. *Phys. Rep.*, 314:237–574, 1999.
- [32] S.N. Majumdar. Brownian functionals in physics and computer science. *Curr. Sci.*, 89:2076, 2005.
- [33] B.B. Mandelbrot. *The fractal geometry of nature*. Freeman, 1983.
- [34] B.B. Mandelbrot and J.W. Van Ness. Fractional Brownian motions, fractional noises and applications. *SIAM Rev.*, 10:422–437, 1968.
- [35] M.M. Meerschaert and H.P. Scheffler. *Limit distributions for sums of independent random vectors, Heavy tails in theory and practice*. Wiley, New York, 2000.
- [36] R. Metzler and J. Klafter. The restaurant at the end of the random walk. *J. Phys. A: Math. Gen.*, 37:161–208, 2004.
- [37] M.S. Mommer and D. Lebiedz. Modeling subdiffusion using reaction diffusion systems. *SIAM J. Appl. Math.*, 70:112–129, 2009.
- [38] M.C. Neel, A. Zoia, and M. Joelson. Mass transport subject to time-dependent flow with uniform sorption in porous media. *Phys. Rev. E*, 80:056301, 2009.

- [39] J.P. Nolan. *Stable Distributions - Models for Heavy Tailed Data*. Birkhauser, Boston, 2012. In progress, Chapter 1 online at [academic2.american.edu/~jpnolan](http://academic2.american.edu/~jpnolan).
- [40] H. Owhadi. Anomalous slow diffusion from perpetual homogenization. *Ann. Prob.*, 31:1935–1969, 2003.
- [41] J.B. Perrin. Mouvement Brownien et réalité moléculaire. *Annales de chimie et de physique*, 8:5–114, 1909.
- [42] A.S. Pikovsky. Escape exponent for transient chaos scattering in non-hyperbolic Hamiltonian systems. *J. Phys. A*, 25:L477–L481, 1992.
- [43] Y. Pomeau and P. Manneville. Intermittent transition to turbulence in dissipative dynamical systems. *Commun. Math. Phys.*, 74:189–197, 1980.
- [44] S. Ross. *Introduction in probability models (chapter 10)*. Academic Press, Boston MA, 1993.
- [45] B. Rubin. *Fractional integrals and potentials*. Harlow, Longman, 1996.
- [46] K. Sakamoto and M. Yamamoto. Initial value/boundary value problems for fractional diffusion wave equations and applications to some inverse problems. *J. Math. Anal. Appl.*, 382(1):426–447, 2011.
- [47] S.G. Samko, A.A. Kilbas, and O.I. Marichev. *Fractional integrals and derivatives: theory and applications*. Gordon and Breach, New York, 1993.
- [48] H. Scher and E.W. Montroll. Anomalous transit-time dispersion in amorphous solids. *Phys. Rev. B*, 12:2455–2477, 1975.
- [49] R. Schumer, D.A. Benson, M.M. Meerschaert, and B. Bauemer. Fractal mobile/immobile solute transport. *Water Resour. Res.*, 39:1296, 2003.
- [50] M.F. Shlesinger, G.M. Zaslavsky, and J. Klafter. Strange kinetics. *Nature*, 363:31–37, 1993.
- [51] A. Shojiguchi, C.B. Li, T. Tomatsuzaki, and M. Toda. Dynamical foundation and limitations of statistical reaction theory. *Comm. Nonlinear Sci. Numer. Simul.*, 13:857–867, 2008.

- [52] M. von Smoluchowski. Zur kinetischen Theorie der Brownschen Molekularbewegung und der Suspensionen. *Ann. der Physik*, 21:756–780, 1906.
- [53] E.R. Weeks, J.S. Urbach, and H.L. Swinney. Anomalous diffusion in asymmetric random walks with a quasi-geostrophic flow example. *Phys. D*, 97:291–310, 1996.
- [54] K. Weron, A. Stanislavsky, A. Jurlewicz, M.M. Meerschaert, and H.P. Scheffler. Clustered continuous time random walks: diffusion and relaxation consequences. *Proc. Royal Soc. A*, 468(2141):1615–1628, 2012.
- [55] W.R. Young. Arrested shear dispersion and other models of anomalous diffusion. *J. Fluid Mech.*, 193:129–149, 1988.
- [56] G. Zaslavsky. Fractional kinetic equation for Hamiltonian chaos. *Phys. D*, 76:110–122, 1994.
- [57] Y. Zhang, D. A. Benson, and B. Bauemer. Moment analysis for spatiotemporal fractional dispersion. *Water Resour.*, 44:W04424, 2008.

## A Some remarks about the power-law distribution of waiting times

Assumption ( $\mathbf{H}_1^{\text{long}}$ ) giving a power-law distribution of waiting times could appear rather empirical. Yet our aim in the present annex is to emphasize that ( $\mathbf{H}_1^{\text{long}}$ ) has a physical or chemical nature, parameter  $\gamma$  reflecting the heterogeneity of the medium.

For a first explicit example, let us show that assumption ( $\mathbf{H}_1^{\text{long}}$ ) which is the basis of the anomalous behavior observed in (1) may be justified by combining at the micro-scale a very classical behavior (namely here isotropic diffusion) with a simple geometrical heterogeneity.

Assume that semi-infinite  $d_r$ -dimensional recirculation branches cross the main propagation direction  $\vec{e}_x$  of the random walkers described in Section 2. Stagnation periods of the random walk thus occur when a particle enters a recirculation branch. This trapping occurs until the particle goes back (for the first time) to the main propagation axis  $\{y = 0\}$ . We assume  $d_r < 2$ . Fig. 2 gives an illustration of the case  $d_r = 1$ . Case  $1 < d_r < 2$  corresponds to fractal structures ( $d_r = 2d_f/d_w$  where  $d_w$  is the diffusion coefficient on the fractal and  $d_f$  is the fractal dimension of the crossing object).

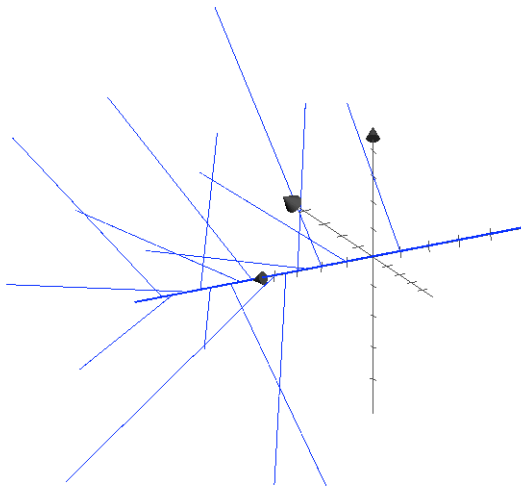


Figure 2: A simple geometry of side branches.

Let  $Q(y, t)$  be the probability that a random walker is at  $y$  at time  $t$  after starting at the origin of a transverse branch, that is  $y = 0$ . Let also  $F(y, t)$  be the corresponding first passage probability. These two quantities are connected through

$$Q(y, t) = \delta_y \delta_t + \int_0^t F(y, t') Q(0, t - t') dt'.$$

Indeed, before returning to  $y$  at time  $t$ , the walker has already reached  $y$  for the first time at  $t' \leq t$ . Using a time Laplace transform we get at the entrance  $y = 0$  of the branch:

$$\hat{F}(0, p) = 1 - \frac{1}{\hat{Q}(0, p)}.$$

Assuming that the displacement is ruled by isotropic diffusion (characterized by a diffusion coefficient  $D_r > 0$ ), it is well-known (*e.g.* [23]) that

$$Q(0, t) = (4\pi D_r t)^{-d_r/2}, \quad t > 0.$$

Thus  $\hat{F}(0, p) = 1 - C(D_r) p^{1-d_r/2}$ ,  $C(D_r) \in \mathbb{R}$ . Since by definition the survival probability  $\Psi$  satisfies  $\Psi'(t) = -F(0, t)$  and  $\Psi(0) = 1$ , it follows that  $\hat{F}(0, p) = -p \hat{\Psi}(p) + 1$ , thus  $\hat{\Psi}(p) = C(D_r) p^{-d_r/2}$  and finally

$$\Psi(t) \sim_{\infty} t^{d_r/2-1}.$$

We thus recover a power-law behavior satisfying assumption  $(\mathbf{H}_1^{\text{long}})$  provided  $0 < d_r < 2$ .

Note that we would obtain the same type of survival probability in case of sorption with diffusion-limited reactions since the form of  $Q$  is similar in this case (see [23, 51] and [1] in a context of protein dynamics).

Note also that assuming  $d_r = 1$  as in Fig. 2 corresponds exactly to the setting considered by Young in [55] (see remark 6 above), yet with a different approach. Moreover, as in [55], we would get a similar result by considering recirculation pockets of finite length  $L$  provided that  $L^2/D_r \gg \mathcal{O}(1)$ .

Let us conclude with more abstract examples. Mandelbrot shows in [33] how power  $\gamma$  in  $(\mathbf{H}_1^{\text{long}})$  is related to sets of fractal dimension and cites many empirical examples. A direct link with the fractal dimension is also stated in [19] for a problem of Knudsen diffusion. We refer to [24] for implication of fractal structures on conductivity. This viewpoint is widely used in percolation theory. The domain might also be peppered with sites at which tracer molecules stick until dislodged, for instance by particularly large thermal fluctuations. The waiting times then depend on the probability distribution density of the energy barrier. Such phenomenon characterizes many physical systems. Assuming that the jumping mechanism among the local minima is described within the framework of Kramer’s reaction rate theory yields to power-law models (see [22]). Power-law tails also characterize weakly chaotic systems, see for instance [43, 42, 2]. We finally cite [37] which shows how the sum of ergodic processes may approach a power-law distribution on any given time interval.

## B A property of the double Laplace transform of causal functions

### B.1 Definition and statement

For the convenience of the reader, we recall the definition of the double Laplace transform, following the lines of [12]. Laplace transform is indeed a very common tool in “physical” literature devoted to the present subject. Let  $G$  be a function, supported by the first quadrant  $\mathbb{R}_+^2$  and Lebesgue measurable in every finite rectangle  $\{0 \leq t_1 \leq T_1\} \times \{0 \leq t_2 \leq T_2\}$ . The double Laplace transform  $\tilde{G}(\lambda, p)$  of  $G$  depends on  $\lambda$  and  $p$ , that are the

Laplace variables conjugate of the physical variables  $t_1$  and  $t_2$ . We set

$$L[G](\lambda, p) = \tilde{G}(\lambda, p) = \int_0^\infty \int_0^\infty G(t_1, t_2) e^{-\lambda t_1 - p t_2} dt_1 dt_2,$$

if the limit of  $\int_0^{T_1} \int_0^{T_2} G(t_1, t_2) e^{-\lambda t_1 - p t_2} dt_1 dt_2$  exists as  $T_1, T_2 \rightarrow \infty$  simultaneously but independently.

**Proposition 5** *Let  $\mathcal{T}_a^2$  denote the translation of amplitude  $a$  with respect to  $t_2$ . Let  $h$  be a causal function of  $t_1$ , with Laplace transform  $\tilde{h}(\lambda)$ . Let  $*_1$  denote convolutions with respect to  $t_1$ . The double Laplace transform  $L[\mathcal{T}_{U \cdot t_1}^2 (h *_1 (\mathcal{T}_{-U \cdot t_1}^2 G))]$  of  $\mathcal{T}_{U \cdot t_1}^2 (h *_1 (\mathcal{T}_{-U \cdot t_1}^2 G))$  is  $\tilde{G}(\lambda, p) \tilde{h}(\lambda + pU)$ .*

**Proof.** We note that in the convolution

$$\begin{aligned} (h *_1 (\mathcal{T}_{-U \cdot t}^2 G(\cdot, \cdot, A)))(t_1, t_2) &= \int_0^{t_1} h(t') G(t_1 - t', A + U(t_1 - t')) dt' \\ &= \int_0^{t_1} h(t') G(t_1 - t', t_2 - Ut') dt' \end{aligned}$$

only the  $t'$  such that  $t_1 < Ut'$  contribute to the integral. Hence,

$$\begin{aligned} &L[\mathcal{T}_{U \cdot t_1}^2 (h *_1 (\mathcal{T}_{-U \cdot t_1}^2 G))](\lambda, p) \\ &= \int_{t_1 > 0} e^{-\lambda t_1} \int_{0 < t' < t_1} h(t') \int_{t_2 > Ut'} G(t_2 - Ut', t_1 - t') e^{-pt_2} dt_2 dt' dt_1, \end{aligned}$$

where

$$\begin{aligned} &\int_{t_2 > Ut'} G(t_2 - Ut', t_1 - t') e^{-pt_2} dt_2 \\ &= e^{-pUt'} \int_{a > 0} G(t_1 - t', a) e^{-pa} da = e^{-pUt'} \tilde{G}(t_1 - t', p). \end{aligned}$$

Hence, we have

$$\begin{aligned} &L[\mathcal{T}_{U \cdot t_1}^2 (h *_1 (\mathcal{T}_{-U \cdot t_1}^2 G))](\lambda, p) \\ &= \int_{t_1 > 0} e^{-\lambda t_1} \int_{0 < t' < t_1} h(t') e^{-pUt'} \tilde{G}(t_1 - t', p) dt' dt_1, \end{aligned}$$

where we recognize the convolution of  $\tilde{G}(\cdot, p)$  with  $h(\cdot) e^{-pU \cdot}$  computed at point  $t_1$ . This latter function has Laplace transform  $\tilde{h}(\lambda + pU)$ . Hence the Proposition.