Approximations of Impulse Response Curves based on the Generalized Moving Gaussian Distribution Function

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Abstract

We derive approximations (in time) for impulse response curves not based on the Gaussian distribution (like the well-known Edgeworth expansions), but based on other suitable distribution functions. In general, we derive the full expansion for such an approximation based on an approximative function, the derivatives and the cumulants of this approximative function and on the cumulants of the impulse response curve itself, see formula (39). Suitable distribution functions exhibit a skewed profile which offers better opportunities for approximations of typical impulse response curves than approximations based on the symmetrical Gaussian distribution. As an example of such a suitable approximative distribution function we study more in detail the Generalized Moving Gaussian distribution

$$Z(t) = \frac{1}{M_0} t^{\nu} \exp\left(-\left(\sqrt{\alpha t} - \sqrt{\beta/t}\right)^2\right), \qquad 0 < t < \infty$$

 $\alpha > 0, \quad \beta > 0, \quad -\infty < \nu < \infty,$
with the normalization
$$M_0 = 2\left(\frac{\beta}{\alpha}\right)^{(\nu+1)/2} \exp\left(2\sqrt{\alpha\beta}\right) K_{\nu+1}\left(2\sqrt{\alpha\beta}\right).$$

A common feature in convection-diffusion problems is that the Laplace transform of an impulse response curve can be described by a single exponential as $a(s) \exp(xb(s))$. We characterize the first four cumulants in terms of the Taylor coefficients of the functions a(s) and b(s). Moreover, it is shown that the temporal cumulants are linear in the spatial variable. This material is applied to several examples and it is shown that the proposed approximative Generalized Moving Gaussian distributions for impulse responses in the field of convection-diffusion type cases perform in general better than the expansions based on the pure Gaussian distribution. Based on this type of approximation other properties based on the original impulse response curve can be found along analytical ways.

1. Introduction

Impulse response curves turn up at many places in the field of (sub)surface hydrology, e.g. as break through curves (BTC) during salt injections, reactive transport with through sorption and sequential firstorder reactions, transport of viruses through the subsoil, or responses due to the management of surface water levels. Also, for hydrogeological time series analysis the measured signal can be considered as a convolution sum of impulse responses due to single events. If one can approximate such curves by means of an analytical expression other properties (e.g. integrals, derivatives) of these signals can be derived in a more formalised way. This approximation is normally based on the matching of the first temporal moments or cumulants. In a number of papers (e.g. Sánchez-Vila and Carrera [11]) the striking resemblance of many impulse responses is observed and explained. Fitting parameters of particular conceptual models is tricky

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since totally different conceptual models can yield almost equal impulse responses as characterized by the first temporal moments or cumulants.

In this paper we elucidate some properties of impulse response functions. In many cases the system which describes the impulse response function can be described by a (large) number of independent random variables (see, e.g., Maas [8]). Under very general conditions the Central Limit Theorem (CLT) can then be applied, which tells us that the impulse response function can be approximated in the limit (for time approaching infinity) by a Gaussian distribution. However, for short or earlier times the impulse response function is skewed and the approximation based on the CLT is not very accurate. There are other approximations (Gram-Charlier and Edgeworth) using the first temporal cumulants which are based on the same Gaussian distribution, but also on higher-order derivatives of this Gaussian distribution. In that way, the approximations become skewed, which resembles more or less reality. The drawback is that the possibility arises that for very early or very late times the approximation becomes negative.

To try to improve these approximations we take as first approximation other distribution functions which are skewed in itself and specify how higher-order approximations can be found using the cumulants along the same lines as the standard Edgeworth approximation for the Gaussian distribution, see for the general results equations Eqs. (38), (39) and (40). The use of other approximating functions has been suggested earlier by Wallace [22]. Another approach has been given by Govindaraju and Das [4], Chapter 12. To apply this theory, the applicant has only to find the cumulants of the impulse response curve and by means of the knowledge of a chosen approximative distribution function (together with derivatives and cumulants) he has to optimize the parameters in this approximative expression in some automated way. As an example for these approximative expressions we study more in detail the (Generalized) Moving Gaussian distribution, see equations Eqs. (42) and (47). In these cases we have just two (α , β) or three (α , β , ν) parameters, respectively. Recently, other authors have used approximations as the Gamma distribution (von Asmuth et al. [20]), and the Generalized Moving Gaussian distribution (for example, Bakker et al. [2]), but these authors do not specify higher-order approximations by means of the derivatives of these approximative functions.

Quite often, the differential equations which describe convection-diffusion processes, give rise to Laplace transforms for the impulse response curve in terms of a single exponential (thus as $a(s) \exp(xb(s))$). Here, it is shown that the temporal cumulants in such cases are linear in the spatial variable x. This property has been derived earlier in a number of cases for specific examples. In Section 3 we characterize the first four cumulants in terms of the Taylor coefficients of the functions a(s) and b(s) which define the Laplace transform of those impulse response functions and use these cumulants thereafter to approximate the corresponding impulse response functions.

Firstly, we motivate our choice later for the (Generalized) Moving Gaussian distribution in Section 2 and show an example of this simple exponential form. Next, we collect some material about moments (Section 3) and give expressions for the moments expressed in terms of the Taylor coefficients of the functions a(s) and b(s). In Section 4 we introduce the notion of cumulants and in Section 5.1 we specify the Gram-Charlier and Edgeworth approximations based on the Gaussian distribution and cumulants. In Subsection 5.2 we derive the theoretical approximation based on suitable distribution functions other than the Gaussian distribution. We also derive correction terms based on derivatives of these approximative functions. As a specific choice we introduce the Generalized Moving Gaussian distribution (Section 5.2.2), for which the Moving Gaussian distribution is a special case (Section 5.2.1). The theory will be illustrated by some examples in Section 6. It is shown that the performance of the approximations based on the Generalized Moving Gaussian distribution is better than these based on the pure Gaussian.

2. Motivation Moving Gaussian distribution

Impulse response functions are measured in situations where models have set up using the concepts of diffusion and dispersion. That means that the corresponding partial differential equation is of the second order with appropriate boundary and initial conditions. To illustrate this, consider the following simple

one-dimensional problem

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - u \frac{\partial c}{\partial x}, \quad t > 0, \quad -\infty < x < \infty,$$

$$c = \frac{G_0}{nA} \delta(t), \quad t = 0, \quad -\infty < x < \infty,$$

$$c = 0, \quad t = 0, \quad x > 0.$$
(1)

where $\delta(x)$ [L⁻¹] is the Dirac-delta function, c [ML⁻³] the concentration, D [L²T⁻¹] the diffusion coefficient, u [LT⁻¹] the velocity, G_0 [M] the amount of released material, n [L³L⁻³] the porosity or volumetric moisture content and A [L²] the cross-sectional area represent. A common method to solve this equation is to apply the Laplace transform technique ($\bar{c}(x,s) = \int_0^\infty c(x,t) \exp(-st) dt$). Then Eq. (1) is transformed into

$$s\overline{c} = D\frac{\partial^2 \overline{c}}{\partial x^2} - u\frac{\partial \overline{c}}{\partial x} + \frac{G_0}{nA}\delta(x), \quad -\infty < x < \infty,$$
(2)

with the usual requirements about the finiteness of the solution at $x = -\infty$ and $x = \infty$. The solution of Eq. (2) reads

$$\overline{c}(x,s) =$$

$$\frac{G_0}{nA} \frac{1}{\sqrt{u^2 + 4Ds}} \exp\left(x \left(\frac{u \mp \sqrt{u^2 + 4Ds}}{2D}\right)\right),$$
(3)

with the –sign for $x \ge 0$, and the +sign for x < 0. The solution c(x,t) is found as $\frac{G_0}{nA}$ times the Gaussian distribution along the real line in the variable $\frac{x-ut}{\sqrt{Dt}}$

$$c(x,t) = \frac{G_0}{nA} \frac{1}{2\sqrt{\pi Dt}} \exp\left(-\left(\frac{x-ut}{2\sqrt{Dt}}\right)^2\right).$$
(4)

The introduction of the Moving Gaussian distribution equation in Subsection 5.2, Eq. (42) is motivated by the exact solution for this example. We remark that in many other cases (see, e.g., van Genuchten and Alves [16]) the Laplace transform of c(x,t) has the form $a(s) \exp(xb(s))$, where $b(s) = -\sqrt{p+qs}$, for some constants p, q, as here above.

3. Moments

The usual approach to study measured breakthrough curves c(x, t) is to calculate the temporal moments. If one assumes that

$$\int_0^\infty c(x,t)dt = M,\tag{5}$$

independent of x, then C(x,t) = c(x,t)/M is really a distribution function and the temporal moments of these curves are defined by

$$n_k(x) = \int_0^\infty t^k C(x, t) dt, \qquad k = 0, 1, \cdots, \infty,$$
(6)

while the central moments are defined as

$$\mu_k(x) = \int_0^\infty (t - n_1)^k C(x, t) dt, \qquad k = 0, 1, \cdots, \infty.$$
(7)

Note that, $n_0(x) = \mu_0(x) = 1$, and $\mu_1(x) = 0$.

The general relationship between the central moments and the moments is given by (see, e.g., Stuart and Ord [15], Eq. (3.7))

$$\mu_k(x) = \sum_{j=0}^k (-1)^j \binom{k}{j} n_{k-j}(x) n_1(x)^j.$$
(8)

Equation (8) implies that the following holds for the second (variance), third and fourth central moments:

$$\mu_{2}(x) = \sigma_{t}^{2}(x) = n_{2}(x) - n_{1}(x)^{2},$$

$$\mu_{3}(x) = g_{t}(x) = n_{3}(x) - 3n_{2}(x)n_{1}(x) + 2n_{1}(x)^{3},$$

$$\mu_{4}(x) = n_{4}(x) - 4n_{3}(x)n_{1}(x) + 6n_{2}(x)n_{1}(x)^{2} - 3n_{1}(x)^{4}.$$
(9)

If one knows the explicit expression of C(x,t) then a useful technique to calculate the moments is to compute the derivatives of the Laplace transform $\overline{C}(x,s)$ of C(x,t)

$$\overline{C}(x,s) = \int_0^\infty C(x,t) \exp(-st) dt,$$
(10)

 \mathbf{as}

$$n_k(x) = (-1)^k \frac{\partial^k \overline{C}(x,s)}{\partial s^k} \Big|_{s=0}, \qquad k = 0, 1, \cdots, \infty.$$
(11)

Formally, this means that

$$\overline{C}(x,s) = \sum_{k=0}^{\infty} n_k(x) \frac{(-s)^k}{k!}.$$
(12)

Next, we suppose that the general form of the function $\overline{C}(x,s)$ has the form

$$\overline{C}(x,s) = a(s)\exp(xb(s)),\tag{13}$$

as is a common form for convection-diffusion equations, and we expand the functions a(s) and b(s) in their power series around s = 0, namely (by the assumption that C(x, t) is a distribution function, see Eq. (5), the term $a_0 = 1$, and $b_0 = 0$)

$$a(s) = 1 + \sum_{i=1}^{5} a_i s^i + O(s^6),$$

$$b(s) = \sum_{i=1}^{5} b_i s^i + O(s^6), \qquad s \to 0.$$
(14)

Now, it is possible to calculate the moments $n_k(x)$ for all k in terms of a_i and b_i . Even for k = 3 this calculation is already a tedious task. By the relations Eq. (8) one will also find expressions for the central moments. For k = 3, 4 the calculations have been carried over using the Formula Manipulation Package of Maple[®]. Here we present the results in terms of a_i and b_i .

$$n_{1}(x) = -b_{1}x - a_{1},$$

$$\sigma_{t}^{2}(x) = 2b_{2}x + 2a_{2} - a_{1}^{2},$$

$$g_{t}(x) = -6b_{3}x - 6a_{3} + 6a_{2}a_{1} - 2a_{1}^{3},$$

$$\mu_{4}(x) = 12b_{2}^{2}x^{2} + (24b_{4} + 24b_{2}a_{2} - 12b_{2}a_{1}^{2})x + 24a_{4} - 24a_{3}a_{1} + 12a_{2}a_{1}^{2} - 3a_{1}^{4},$$

$$\mu_{4}(x) - 3\sigma_{t}^{4}(x) = 24b_{4}x + 24a_{4} - 24a_{3}a_{1} + 24a_{2}a_{1}^{2} - 6a_{1}^{4} - 12a_{2}^{2}.$$
(15)

It turns out that for the first four moments the coefficients in the Taylor expansion Eq. (14) up to the fourth order suffice $(a_4 \text{ and } b_4)$. It is much easier to apply Eq. (15) to find the moments using the Taylor expansion Eq. (14) than to apply Eq. (11) directly. We have specified also $\mu_4(x) - 3\sigma_t^4(x)$, which is the fourth cumulant described in Section 4 below (see Eq. (20)).

4. Cumulants

Another useful tool to characterize the distribution is by means of the theoretically attractive cumulants $\kappa_r(x)$, which are usually defined implicitly as (see Stuart and Ord [15], Eq. (3.31))

$$\hat{C}(x,p) = \int_{-\infty}^{\infty} \exp(ipt)C(x,t)dt$$

$$= \exp\left(\sum_{r=1}^{\infty} \kappa_r(x)\frac{(ip)^r}{r!}\right),$$
(16)

with $i = \sqrt{-1}$ and where $\hat{C}(x, p)$ represents the characteristic function of C(x, t). For distributions C(x, t) with C(x, t) = 0, t < 0, this is equivalent with the definition (see also Eq. (12))

$$\exp\left(\sum_{r=1}^{\infty} \kappa_r(x) \frac{(-s)^r}{r!}\right) =$$

$$\sum_{r=0}^{\infty} n_r(x) \frac{(-s)^r}{r!} = \overline{C}(x,s).$$
(17)

This means that

$$\sum_{r=1}^{\infty} \kappa_r(x) \frac{(-s)^r}{r!} = \log\left(\overline{C}(x,s)\right),\tag{18}$$

and the cumulants can be found as for $r = 1, \dots, \infty$,

$$\kappa_r(x) = (-1)^r \frac{\partial^r \log\left(\overline{C}(x,s)\right)}{\partial s^r} \bigg|_{s=0}.$$
(19)

It depends whether the cumulants are evaluated for distributions with $n_1(x) = 0$ or not. Stuart and Ord [15], (Exercise 3.9, p. 113) gives the relation between $n_r(x)$ and $n_{r-j}(x)$, $\kappa_j(x)$, $j = 1, \dots, r$, from which also an expression can be derived for $\kappa_r(x)$ in terms of $n_j(x)$, $j = 1, \dots, r$. Another representation for $\kappa_r(x)$ in terms of $n_j(x)$ and (B3)).

The first cumulants are given by

$$\kappa_{1}(x) = n_{1}(x),$$

$$\kappa_{2}(x) = \sigma_{t}^{2}(x) = n_{2}(x) - n_{1}(x)^{2},$$

$$\kappa_{3}(x) = g_{t}(x) = n_{3}(x) - 3n_{2}(x)n_{1}(x) + 2n_{1}(x)^{3},$$

$$\kappa_{4}(x) = \mu_{4}(x) - 3\sigma_{t}^{4}(x) =$$

$$n_{4}(x) - 4n_{3}(x)n_{1}(x) - 3n_{2}(x)^{2}$$

$$+ 12n_{2}(x)n_{1}(x)^{2} - 6n_{1}(x)^{4}.$$
(20)

See Eq. (15) for the expression for $\mu_4(x) - 3\sigma_t^4(x)$ in Eq. (20). We remark that the first three cumulants are equal to the central moments and that $\kappa_4(x)$ differs from $\mu_4(x)$ by the term $-3\sigma_t^4(x)$. Some common characteristics of distribution functions may be expressed in moments and cumulants (Abramowitz and

Stegun [1], (26.1.15-18))

skewness; (also) coefficient of skewness : $\gamma_1(x) = \kappa_3(x)/\kappa_2^{3/2}(x) = g_t(x)/\sigma_t^3(x),$

coefficient of skewness (alternative) : $\beta_1(x) = \gamma_1^2(x) = g_t^2(x)/\sigma_t^6(x),$

excess :

$$\gamma_2(x) = \kappa_4(x) / \sigma_t^4(x) = \mu_4(x) / \sigma_t^4(x) - 3$$

kurtosis; (also) coefficient of kurtosis : $\beta_2(x) = \mu_4(x)/\sigma_t^4(x) = \kappa_4(x)/\sigma_t^4(x) + 3.$

From the very definition of the cumulants or from Eq. (18) it can be seen that all cumulants are linear in the independent variable x for cases with $\overline{C}(x,s) = a(s) \exp(xb(s))$. A priori, the behaviour of the central moments w.r.t. x is not obvious, but by the relations in Eq. (15) it is clear that e.g. the skewness $\kappa_3(x) = g_t(x)$ is always linear in x. This has been found earlier in the literature for quite a number of conceptual models (see Schmid [13], Wörman et al. [23], Sánchez-Vila and Carrera [11]). See also Veling [19].

(21)

5. Representation of the distribution functions by moments and cumulants

5.1. Gaussian distribution

With the aid of the moments or the cumulants, the distribution function can be represented either as a so-called Gram-Charlier or an Edgeworth series based on the Gaussian distribution

$$g(t) = \frac{1}{\sqrt{2\pi}} \exp(-t^2/2), \qquad -\infty < t < \infty,$$
 (22)

and, in this section we summarize these representations. We have to remark that in these theoretical approximations it is assumed that the variable t ranges from $-\infty < t < \infty$. A consequence is that for the Gaussian distribution g(t) all cumulants $\lambda_n = 0$, for $n \ge 3$, using Eq. (16), since $\hat{g}(p) = \exp(-p^2/2)$. Under quite general conditions, a distribution function f(x,t) can be represented as a Gram-Charlier series

$$f(x,t) = \frac{1}{\sqrt{2\pi}} \sum_{j=0}^{\infty} c_j(x) \operatorname{He}_j(t) \exp(-t^2/2),$$
(23)

where the functions $\operatorname{He}_{j}(t)$ are the Hermite polynomials, defined by for $j = 0, 1, 2, \cdots$

$$\operatorname{He}_{j}(t) = (-1)^{j} \exp(t^{2}/2) \left(\frac{d}{dt}\right)^{j} \exp(-t^{2}/2).$$
(24)

The very first polynomials are

 $\begin{aligned} &\text{He}_{0}(t) = 1, \\ &\text{He}_{1}(t) = t, \\ &\text{He}_{2}(t) = t^{2} - 1, \\ &\text{He}_{3}(t) = t^{3} - 3t, \\ &\text{He}_{4}(t) = t^{4} - 6t^{2} + 3, \\ &\text{He}_{5}(t) = t^{5} - 10t^{3} + 15t, \\ &\text{He}_{6}(t) = t^{6} - 15t^{4} + 45t^{2} - 15. \end{aligned}$

The coefficients $c_j(x)$ are given by

$$c_j(x) = \frac{1}{j!} \int_{-\infty}^{\infty} f(x,\tau) \operatorname{He}_j(\tau) d\tau.$$
(26)

Under the assumptions $n_0(x) = \mu_0(x) = 1$ (*i.e.*, normalization of the function), which implies that for $t' = (t - n_1(x))/\sqrt{\sigma_t^2(x)}$: $n_1(x) = \mu_1(x) = 0$ and $n_2(x) = \sigma_{t'}^2(x) = 1$, the coefficients $c_j(x)$ can be expressed in terms of the cumulants as

$$c_{0}(x) = 1,$$

$$c_{1}(x) = 0,$$

$$c_{2}(x) = 0,$$

$$c_{3}(x) = \frac{1}{6}\kappa_{3}(x),$$

$$c_{4}(x) = \frac{1}{24}\kappa_{4}(x),$$

$$c_{5}(x) = \frac{1}{120}\kappa_{5}(x),$$

$$c_{6}(x) = \frac{1}{720} \left(\kappa_{6}(x) + 10\kappa_{3}^{2}(x)\right),$$

(27)

(see Eq. (20) and Stuart and Ord [15], Eq. (3.37) for $\kappa_5(x)$ and $\kappa_6(x)$, with $\kappa_1(x) = 0$, $\kappa_2(x) = 1$). These formulas all together result in the following approximation for the distribution function f(x,t)

$$f(x,t) = \frac{1}{\sqrt{2\pi\sigma_t^2(x)}} \exp\left(-T^2/2\right)$$

$$\left\{ 1 + \frac{1}{6}\kappa_3(x)\operatorname{He}_3(T) + \frac{1}{24}\kappa_4(x)\operatorname{He}_4(T) + \frac{1}{120}\kappa_5(x)\operatorname{He}_5(T) + \frac{1}{720}\left(\kappa_6(x) + 10\kappa_3^2(x)\right)\operatorname{He}_6(T) + \cdots \right\},$$
with $T = (t - n_1(x))/\sqrt{\sigma_t^2(x)}.$
(28)

Another approach, the so-called Edgeworth expansion for the same function is written in the concise form (see Stuart and Ord [15], Eq. (6.40))

$$f(x,t) = \frac{1}{\sqrt{2\pi\sigma_t^2(x)}} \times \exp\left(-\sum_{j=3}^{\infty} \kappa_j \mathbf{D}_T^j\right) \exp\left(-T^2/2\right) \Big|_{T = \frac{(t-n_1(x))}{\sqrt{\sigma_t^2(x)}}},$$
(29)

where \mathbf{D}_T is the differential operator w.r.t. T. If we assume that the stochastic variable $T = (\sum_i^n (T_i - n_s)) / (\sqrt{n}\sigma_s)$ is the sum of n independent stochastic variables T_i , all with the same mean n_s , variance σ_s and cumulants $\kappa_{s,j}$, $j \geq 3$, so $n_1(x) = nn_s$, $\sigma_t = \sqrt{n}\sigma_s$ then the cumulants for T become $\kappa_j = n\kappa_{s,j} / (\sqrt{n}\sigma_s)^j = \kappa_{s,j} / (n^{(j/2)-1}\sigma_s^j)$. If one formally expands the exponential in Eq. (29) in powers of $1/\sqrt{n}$ one finds (using Eq. (24))

$$f(x,t) = \frac{1}{\sqrt{2\pi\sigma_t^2(x)}} \exp\left(-T^2/2\right) \times \left\{ 1 + \frac{1}{6}\kappa_3(x) \operatorname{He}_3(T) + \left(\frac{1}{24}\kappa_4(x) \operatorname{He}_4(T) + \frac{1}{72}\kappa_3^2(x) \operatorname{He}_6(T)\right) + \left(\frac{1}{120}\kappa_5(x) \operatorname{He}_5(T) + \frac{1}{144}\kappa_3(x)\kappa_4(x) \operatorname{He}_7(T) + \frac{1}{1296}\kappa_3^3(x) \operatorname{He}_9(T)\right) \right\} + O\left(\frac{1}{n^2}\right) \Big|_{T = \frac{(t-n_1(x))}{\sqrt{\sigma_t^2(x)}}},$$
(30)

where the terms between brackets () are of the same order in n, so, explicitly, $\kappa_4(x)$ is of the same order in n as $\kappa_3^2(x)$. Compare Eq. (30) with Abramowitz and Stegun [1], (26.2.47) & Comment under (26.2.48). In fact, Eqs. (23) and (29) represent the same function and so, the difference between Eqs. (28) and (30) is just the order of the terms (see Stuart and Ord [15], §§ 6.15 - 6.20). In practice, moments and cumulants above the fourth order are difficult to calculate, so neglecting $\kappa_5(x)$, $\kappa_6(x)$, \cdots , and also powers of $\kappa_4(x)$ beyond order 1, $\kappa_3(x)$ beyond order 2 in Eq. (30), one sees that the difference between the Gram-Charlier and the Edgeworth expansion (Eqs. (28) and (30)), both with just three terms, is the extra addition in the third term in (Eq. 30): $\frac{1}{72}\kappa_3^2(x)\text{He}_6(T)$. The grouping of the terms in Eq. (30) is better because it is an asymptotic series. A drawback of both expansions is that for some parameter values they become negative, which is annoying for most applications. For a discussion about the sign of these expansions we refer to Maas [8], (Chapter 2.5).

5.2. Basic approximation functions

In the remainder of this paper we show that it is possible to approximate a given distribution function not only by the Gaussian distribution as given above but also by some other distribution function. We shall apply the technique as presented in the readable account of Blinnikov & Moessner (Blinnikov and Moessner [3], (41)) for one specific distribution. We shall give an account in terms of the Laplace transform, although it is more common to use the Fourier transform. In the sequel we denote the dependence on x in the various functions explicitly, but we remark that all approximations are for fixed x, so this variable x has to be interpreted as a parameter.

Assume the distribution function p(x,t) of a random variable T with $n_1(x) = \kappa_1(x) \neq 0$, variance $\sigma_t^2(x) = \kappa_2(x)$ and cumulants $\kappa_n(x)$, $n \geq 3$. The Laplace transform is $\bar{p}(x,s)$, and we assume that for the basic approximation Z(x,t) for the random variable T^* with cumulants $\lambda_n(x)$, $n \geq 1$, the parameters have been chosen such that $\lambda_1(x) = \kappa_1(x) = n_1(x)$, $\lambda_2(x) = \kappa_2(x) = \sigma_t^2(x)$, and that the Laplace transform of Z is $\bar{Z}(x,s)$. For this case, the distribution functions for the random variables T/σ_t and T^*/σ_t are respectively $q(x,t) = \sigma_t p(x,\sigma_t t)$ and $Z^{\hat{}}(x,t) = \sigma_t Z(x,\sigma_t t)$. The Laplace transforms of these distribution functions are respectively $\bar{q}(x,s) = \bar{p}(x,s/\sigma_t)$ and $\overline{Z^{\hat{}}}(x,s) = \bar{Z}(x,s/\sigma_t)$. Then by Eq. (17)

$$\bar{p}(x, s/\sigma_t) =$$

$$\exp\left(\sum_{r=3}^{\infty} \left(\kappa_r(x) - \lambda_r(x)\right) \frac{(-s)^r}{\sigma_t^r r!}\right) \bar{Z}(x, s/\sigma_t) =$$

$$\exp\left(\sum_{r=1}^{\infty} \sigma_t^{-2} \left(\kappa_{r+2}(x) - \lambda_{r+2}(x)\right) \frac{(-s)^{r+2}}{\sigma_t^r (r+2)!}\right) \times \bar{Z}(x, s/\sigma_t).$$
(31)

We shall expand the exponential into a power series in $w = 1/\sigma_t$. Therefore we shall make use of the following formula of Francesco Faà di Bruno (published in 1855) for the *n*-th derivative of a composite function $f \circ g(w) = f(g(w))$, see, e.g., Lukacs [7] and Blinnikov and Moessner [3], (30):

$$\frac{d^{n}}{dw^{n}}f(g(w)) =$$

$$n! \sum_{\{K\}_{n,r}} f^{(r)}(y)|_{y=g(w)} \prod_{m=1}^{n} \frac{1}{k_{m}!} \left(\frac{1}{m!}g^{(m)}(w)\right)^{k_{m}},$$
(32)

Here we define $\{K\}_{n,r}$ and $\{K\}_n$ as follows

 $\{K\}_{n,r} \text{ the set } \{k_1, k_2, \cdots, k_n\} \text{ of all}$ non-negative integers satisfying $k_1 + 2k_2 + \cdots + nk_n = n \&$ $k_1 + k_2 + \cdots + k_n = r,$ $\{K\}_n \text{ as } \{K\}_{n,r} \text{ without}$ the restriction w.r.t. r.

In the case here, we take $f = \exp$ and $g(w) = \sum_{r=1}^{\infty} \sigma_t^{-2} \left(\kappa_{r+2}(x) - \lambda_{r+2}(x)\right) \frac{(-s)^{r+2}}{(r+2)!} w^r$ in Eq. (32) and we find

$$\bar{p}(x, s/\sigma_t) = \left(\sum_{r=1}^{\infty} \sigma_t^{-2} \left(\kappa_{r+2}(x) - \lambda_{r+2}(x)\right) \frac{(-s)^{r+2}}{(r+2)!}\right) \times \bar{Z}(x, s/\sigma_t) = \left(1 + \sum_{n=1}^{\infty} P_n(s)\sigma_t^{-n}\right) \bar{Z}(x, s/\sigma_t),$$
(33)

where the polynom $P_n(s)$ is defined as

$$P_{n}(s) = \frac{1}{n!} \frac{d^{n}}{dw^{n}} f(g(w))|_{w=0} = \sum_{\{K\}_{n}} \prod_{m=1}^{n} \frac{1}{k_{m}!} \times \left(\sigma_{t}^{-2} \left(\kappa_{m+2}(x) - \lambda_{m+2}(x)\right) \frac{(-s)^{m+2}}{(m+2)!}\right)^{k_{m}},$$
(34)

The polynom $P_n(s)$ can be rewritten as

$$P_{n}(s) = \sum_{\{K\}_{n,r}} (-s)^{n+2r} \prod_{m=1}^{n} \frac{1}{k_{m}!} \times \left(\frac{\sigma_{t}^{-2} \left(\kappa_{m+2}(x) - \lambda_{m+2}(x)\right)}{(m+2)!}\right)^{k_{m}},$$
(35)

Explicitly,

$$P_{1}(s) = \left\{\sigma_{t}^{-2} \left(\kappa_{3}(x) - \lambda_{3}(x)\right)\right\} \frac{(-s)^{3}}{6},$$

$$P_{2}(s) = \left\{\sigma_{t}^{-2} \left(\kappa_{4}(x) - \lambda_{4}(x)\right)\right\} \frac{(-s)^{4}}{24} + \left\{\sigma_{t}^{-2} \left(\kappa_{3}(x) - \lambda_{3}(x)\right)\right\}^{2} \frac{(-s)^{6}}{72},$$

$$P_{3}(s) = \left\{\sigma_{t}^{-2} \left(\kappa_{5}(x) - \lambda_{5}(x)\right)\right\} \frac{(-s)^{5}}{120} + \left\{\sigma_{t}^{-4} \left(\kappa_{3}(x) - \lambda_{3}(x)\right) \times \left(\kappa_{4}(x) - \lambda_{4}(x)\right)\right\} \frac{(-s)^{7}}{144} + \left\{\sigma_{t}^{-2} \left(\kappa_{3}(x) - \lambda_{3}(x)\right)\right\}^{3} \frac{(-s)^{9}}{1296}.$$
(36)

Next, we make use of the property of the Laplace transform that the function

$$s^n \bar{f}(s) - s^{n-1} f(0) - s^{n-2} f^{(1)}(0) - \dots - f^{(n-1)}(0)$$

is the transform of $f^{(n)}(t)$.

So, to proceed we have to require that for our approximative functions Z(x,t), it holds that $Z^{(j)}(x,0) = 0$, $j = 0, \cdots$. That means that we can not allow the so-called Pearson III type approximation

$$Z(t) = \frac{1}{\gamma \Gamma(p)} \left(\frac{t}{\gamma}\right)^{p-1} \exp\left(-\left(\frac{t}{\gamma}\right)\right),$$

$$0 \le t < \infty, \quad \gamma > 0, \quad p > 0,$$

where $n_1 = \lambda_1 = p\gamma, \ \sigma^2 = \lambda_2 = p\gamma^2,$
and $\lambda_n(x) = p\gamma^n \Gamma(n), \ n \ge 1,$
and $\bar{Z}(s) = (1 + \gamma s)^{-p},$
(37)

since the *n*-th derivative of Z(t) becomes unbounded at t = 0 for n > p - 1.

So, for a distribution function p(x,t) with $n_1(x) = \kappa_1(x) \neq 0$, variance $\sigma^2(x) = \kappa_2(x)$ and cumulants $\kappa_n(x)$, $n \geq 3$, the generalized form of such Edgeworth type asymptotic expansion is (by applying the inverse Laplace transform, *i.e.* $\bar{Z}(s/c)$ is the transform of cZ(ct) and $(s/c)^n \bar{Z}(s/c)$ is the transform of $cZ^{(n)}(ct) = Z^{(n)}(t)/c^n$

$$q(x,t) = Z^{\hat{}}(x,t) +$$

$$\sum_{n=1}^{\infty} \sum_{\{K\}_{n,r}} \frac{(-1)^{n+2r}}{\sigma_t^{n+2r}} \frac{d^{n+2r}}{dt^{n+2r}} Z^{\hat{}}(x,t) \times$$

$$\prod_{m=1}^{n} \frac{1}{k_m!} \left(\frac{\kappa_{m+2}(x) - \lambda_{m+2}(x)}{(m+2)!} \right)^{k_m},$$
(38)

And so, in terms of p(x,t) (since $p(x,t) = q(x,t/\sigma_t)/\sigma_t$ and $Z(x,t) = Z(x,t/\sigma_t)/\sigma_t$)

$$p(x,t) = Z(x,t) +$$

$$\sum_{n=1}^{\infty} \sum_{\{K\}_{n,r}} (-1)^{n+2r} \frac{d^{n+2r}}{dt^{n+2r}} Z(x,t) \times$$

$$\prod_{m=1}^{n} \frac{1}{k_m!} \left(\frac{\kappa_{m+2}(x) - \lambda_{m+2}(x)}{(m+2)!} \right)^{k_m},$$
(39)

Writing out for the first terms gives

$$p(x,t) = Z(x,t)$$

$$-\frac{1}{6} (\kappa_3(x) - \lambda_3(x)) \frac{d^3}{dt^3} Z(x,t)$$

$$+\frac{1}{24} (\kappa_4(x) - \lambda_4(x)) \frac{d^4}{dt^4} Z(x,t)$$

$$+\frac{1}{72} (\kappa_3(x) - \lambda_3(x))^2 \frac{d^6}{dt^6} Z(x,t) + \cdots .$$
(40)

An application of the formulas Eqs. (38) and (39) is the Edgeworth asymptotic expansion Eq. (30) from Section 5.1, which uses the Gaussian distribution $Z(t) = \frac{1}{\sqrt{2\pi}} \exp(-t^2/2)$. The stochastic variable T is now scaled as $T^* = (t - n_1(x))/\sigma_t(x)$ with $\lambda_1 = n_1(x)$, $\lambda_2 = \sigma_t^2(x)$, $\lambda_n = 0$, $n \ge 3$, and the corresponding function Z(x,t) becomes

$$Z(x,t) = \frac{1}{\sqrt{2\pi\sigma_t^2(x)}} \exp\left(-\frac{(t-n_1(x))^2}{2\sigma_t^2(x)}\right) = \frac{1}{\sqrt{2\pi\sigma_t^2(x)}} \exp\left(-T^{*2}/2\right),$$
with $T^* = (t-n_1(x))/\sigma_t(x),$
(41)

and the derivatives of Z(x, t) give the product of the Hermite polynomials $\operatorname{He}_j(T^*)$ with Z(x, t). Moreover, Eq. (39) holds for $-\infty < t < \infty$.

In the sequel we shall make some other specific choices for the function Z(t).

5.2.1. Moving Gaussian distribution

In view of the analytical solution of the standard model Eq. (1) it seems appropriate to use a function Z(t) which itself encompasses the essential mathematical behaviour. Therefore, we take for Z(t) the function

$$Z(t) = \sqrt{\frac{\alpha}{\pi}} t^{-1/2} \exp\left(-\left(\sqrt{\alpha t} - \sqrt{\beta/t}\right)^2\right),$$

$$0 < t < \infty, \quad \alpha > 0, \quad \beta > 0.$$
(42)

For the choice $\alpha = u^2/(4D)$ and $\beta = x^2/(4D)$ we find Eq. (4), after we have normalized Eq. (4) by dividing by $n_0 = M = G_0/(nAu)$. We call this distribution the "Moving Gaussian distribution" by the fact that it is a Gaussian distribution which travels to the right as function of the place variable x. In fact Eq. (42) is a generalization of the Pearson Type III distribution Eq. (37) for $\alpha = 1/\gamma$, $\beta = 0$ in Eq. (42), but by the restriction p = 1/2 in Eq. (37), and with $n_1 \neq 0$. The mean n_1 and variance σ^2 become

$$n_1 = \sqrt{\frac{\beta}{\alpha}} + \frac{1}{2\alpha}, \qquad \sigma^2 = \frac{1}{2}\sqrt{\frac{\beta}{\alpha^3}} + \frac{1}{2\alpha^2},\tag{43}$$

but we refrain to translate Eq. (42) such that $n_1 = 0$. The Laplace transform of Eq. (42) is

$$\bar{Z}(s) = \sqrt{\frac{\alpha}{\alpha+s}} \exp\left(2\left(\sqrt{\alpha\beta} - \sqrt{\beta(\alpha+s)}\right)\right).$$
(44)

Relation (44) can be derived using that (see Oberhettinger and Badii [10], Part I, (5.34) and Abramowitz and Stegun [1], (10.2.17), respectively):

$$\int_{0}^{\infty} t^{p-1} \exp\left(-at - b/t\right) dt =$$

$$2\left(\frac{b}{a}\right)^{p/2} K_p\left(2\sqrt{ab}\right),$$

$$a, b > 0, \text{ and } K_{1/2}(z) = \sqrt{\frac{\pi}{2z}} \exp(-z).$$

$$(45)$$

Next, Eq. (45) is applied with p = 1/2 to derive Eq. (44). The cumulants of this distribution are

$$\lambda_n = \frac{1}{2} \alpha^{-n} \Gamma(n) - 2 \alpha^{1/2 - n} \beta^{1/2} \left(-\frac{1}{2} \right)_n, \tag{46}$$

where $(a)_n$ is defined as $(a)_0 = 1$, $(a)_n = a(a+1)(a+2)\cdots(a+n-1)$, $n \ge 1$. See Appendix A for more information w.r.t. the Moving Gaussian distribution.

5.2.2. Generalized Moving Gaussian distribution

Next we introduce a generalized form of Eq. (46) in the sense that we do not fix the exponent of the factor t. So, we study

$$Z(t) = \frac{1}{M_0} t^{\nu} \exp\left(-\left(\sqrt{\alpha t} - \sqrt{\beta/t}\right)^2\right),$$

$$0 < t < \infty, \quad \alpha > 0, \quad \beta > 0, \quad -\infty < \nu < \infty.$$
(47)

Here, M_0 is the normalization constant. Using Eq. (45) we find

$$M_0 = 2\left(\frac{\beta}{\alpha}\right)^{(\nu+1)/2} \exp\left(2\sqrt{\alpha\beta}\right) K_{\nu+1}\left(2\sqrt{\alpha\beta}\right).$$
(48)

The Laplace transform of Eq. (47) is

$$\bar{Z}(s) = 2\left(\frac{\beta}{\alpha+s}\right)^{(\nu+1)/2} \times$$

$$\exp\left(2\sqrt{\alpha\beta}\right) K_{\nu+1} \left(2\sqrt{(\alpha+s)\beta}\right)/M_0 =$$

$$\left(\frac{\alpha}{\alpha+s}\right)^{(\nu+1)/2} \left(\frac{K_{\nu+1} \left(2\sqrt{(\alpha+s)\beta}\right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta}\right)}\right).$$
(49)

The cumulants are more complicated to calculate. Here, we specify the results for the first two (again using the Formula Manipulation Package of $Maple^{(B)}$)

$$\lambda_{1} = n_{1} =$$

$$\frac{\nu + 1}{\alpha} + \frac{\sqrt{\alpha\beta}}{\alpha} \left(\frac{K_{\nu} \left(2\sqrt{\alpha\beta} \right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta} \right)} \right),$$

$$\lambda_{2} = \sigma_{t}^{2} =$$

$$\frac{\nu + 1 + \alpha\beta}{\alpha^{2}} - \frac{\nu\sqrt{\alpha\beta}}{\alpha^{2}} \left(\frac{K_{\nu} \left(2\sqrt{\alpha\beta} \right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta} \right)} \right)$$

$$- \frac{\beta}{\alpha} \left(\frac{K_{\nu} \left(2\sqrt{\alpha\beta} \right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta} \right)} \right)^{2},$$
(50)

We remark that these results for $\nu = -1/2$ reduce to what has been found in Eq. (43) for n = 1, 2. See Appendix B for more information w.r.t. the Generalized Moving Gaussian distribution.

6. Examples

In this section, we apply our general resuls to several boundary value problems.

6.1. Example 1

We modify the first example in the sense that we consider the convection-diffusion equation in a quarterplane $(x, t) = \mathbb{R}^+ \times \mathbb{R}^+$:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - u \frac{\partial c}{\partial x}, \quad t > 0, \quad x > 0,$$

$$c - \frac{D}{u} \frac{\partial c}{\partial x} = \frac{G_0}{nuA} \delta(t), \quad t \ge 0, \quad x = 0,$$

$$c = 0, \quad t = 0, \quad x > 0.$$
(52)

The boundary condition at x = 0 represents a so-called sudden injection in flux, see Kreft and Zuber [6]. The Laplace transform is

$$\overline{c}(x,s) = \frac{G_0}{nuA} \frac{2u}{u + \sqrt{u^2 + 4Ds}} \times \exp\left(x \left(\frac{u - \sqrt{u^2 + 4Ds}}{2D}\right)\right),$$

and the corresponding analytical solution, see Kreft and Zuber [6] and Veling [18]:

$$c(x,t) = \frac{G_0}{nuA} \left[\frac{u}{\sqrt{\pi Dt}} \exp\left(-\left(\frac{x-ut}{2\sqrt{Dt}}\right)^2\right) - \frac{1}{2}\frac{u^2}{D} \exp\left(\frac{ux}{D}\right) \operatorname{erfc}\left(\frac{x+ut}{2\sqrt{Dt}}\right) \right].$$

The normalized solution reads

$$C(x,t) = \frac{u}{\sqrt{\pi Dt}} \exp\left(-\left(\frac{x-ut}{2\sqrt{Dt}}\right)^2\right) - \frac{1}{2}\frac{u^2}{D} \exp\left(\frac{ux}{D}\right) \operatorname{erfc}\left(\frac{x+ut}{2\sqrt{Dt}}\right).$$
(53)

Here, this analytical solution equals a Moving Gaussian distribution except for the second term. The corresponding coefficients a_i and b_i , i = 1, 2, 3, 4, (see Eq. (14)) are found as:

$$a_{1} = -\frac{D}{u^{2}}, \qquad a_{2} = 2\frac{D^{2}}{u^{4}}, a_{3} = -5\frac{D^{3}}{u^{6}}, \qquad a_{4} = 14\frac{D^{4}}{u^{8}}, b_{1} = -\frac{1}{u}, \qquad b_{2} = \frac{D}{u^{3}}, b_{3} = -2\frac{D^{2}}{u^{5}}, \qquad b_{4} = 5\frac{D^{3}}{u^{7}}.$$
(54)

The corresponding moments are now (based on Eq. (15)):

$$\begin{aligned} \kappa_1(x) &= n_1(x) = \frac{x}{u} + \frac{D}{u^2}, \\ \kappa_2(x) &= \sigma_t^2(x) = 2\frac{Dx}{u^3} + 3\frac{D^2}{u^4}, \\ \kappa_3(x) &= g_t(x) = 12\frac{D^2x}{u^5} + 20\frac{D^3}{u^6}, \\ \mu_4(x) &= 12\frac{D^2x^2}{u^6} + 156\frac{D^3x}{u^7} + 237\frac{D^4}{u^8}, \\ \kappa_4(x) &= \mu_4(x) - 3\sigma_t^4(x) = 120\frac{D^3x}{u^7} + 210\frac{D^4}{u^8}. \end{aligned}$$

We compare the different approximations for the analytical solution C(x,t) for the following parameter values

(55)

$$\begin{split} D &= 0.01 \ \mathrm{m}^2 \mathrm{d}^{-1}, \quad u = 0.1 \ \mathrm{m} \mathrm{d}^{-1}, \quad x = 1.4 \ \mathrm{m}, \\ & \text{such that} \quad x u / D = 14 \ [-], \end{split}$$

which implies that the first cumulants are $\kappa_1 = n_1 = 15$ d, $\kappa_2 = \sigma_t^2 = 31$ d², $\kappa_3 = g_t = 188$ d³, $\kappa_4 = 1890$ d⁴. In Fig. E.1 the exact solution is compared with the Gaussian distribution, with the Gram-Charlier approximation Eq. (28) with one and two terms and with the Edgeworth approximation. Only the Edgeworth approximation gives a reasonable good fit.

In Fig. E.2 the exact solution is compared with the first approximation Eq. (42) of the Moving Gaussian distribution Z(x,t) and second approximation, *i.e.* the first two terms in Eq. (40), where we have taken for the values for the parameters α and β the expressions stemming from the first term in Eq. (53): $\alpha = u^2/(4D) = 0.25 \text{ d}^{-1}, \ \beta = x^2/(4D) = 49 \text{ d}, \ \nu = -0.5$. That means that we approximate Eq. (53) just by deleting the second term. The neglection of the second term is Eq. (53) is clearly visible: the approximation is not that good.

In Fig. E.3 the exact solution is compared with the first approximation Eq. (42) of the Moving Gaussian distribution Z(x,t) and second approximation, i.e. with the first two terms in Eq. (40) but now with optimized values for α and β , by equating the expressions for n_1 of the exact solution Eq. (55) to the n_1 of the Moving Gaussian distribution Eq. (43), and the same with the corresponding σ_t^2 values. It follows

$$\alpha = \frac{n_1 + \sqrt{n_1^2 + 4\sigma_t^2}}{4\sigma_t^2},$$

$$\beta = \frac{n_1^3 - 5n_1\sigma_t^2 + (\sigma_t^2 + n_1^2)\sqrt{n_1^2 + 4\sigma_t^2}}{4\sigma_t^2}.$$
(56)

The values for the parameters are $\alpha = 0.2716 \text{ d}^{-1}$, $\beta = 47.0361 \text{ d}$, $\nu = -0.5$. This approximation is already quite good as can also be read off from the differences between the cumulants κ_i and λ_i .

In Fig. E.4 the exact solution is compared with the first approximation Eq. (47) of the Generalized Moving Gaussian distribution Z(x,t) and the third approximation based on the third and fourth cumulants and the third, fourth and sixth derivatives (see Eq. (36) for $P_2(s)$). Here, we have optimized for α , β and ν . The values found are $\alpha = 0.2564 \text{ d}^{-1}$, $\beta = 50.8088 \text{ d}$, $\nu = -1.0116$. It is difficult to see any difference between the exact solution and the approximations. These values correspond very well with the κ_i values.

See Table D.1 for the parameters values and cumulants for the different approximations.

6.2. Example 2

We treat a somewhat more complicated problem, namely the mathematical model for the description of released material into a river with dead zones (see Nordin and Troutman [9], Schmid [13] and Veling [19]). Other applications for this model can be mentioned such as two-species reactive transport equations coupled through sorption and sequential first-order reactions (van Kooten [17]) and transport of viruses through the

subsoil (Sim and Chrysikopoulos [14], Schijven et al. [12]). This model with a different interpretation for the variables and parameters might be applicable in general where one has to deal with one partial differential equation linearly coupled with one ordinary differential equation, which is the case for a broad range of applications in science. For $-\infty < x < \infty$,

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - u \frac{\partial c}{\partial x} - \frac{\varepsilon}{\rho} (c - c_s), \quad t > 0,$$

$$\frac{\partial c_s}{\partial t} = \frac{1}{\rho} (c - c_s), \quad t > 0,$$

$$c = \frac{G_0}{nA} \delta(x), \quad t = 0,$$

$$c_s = 0, \quad t = 0,$$
(57)

where ε [-] is the ratio of the dead zone area to the main stream flow cross section, and ρ [T] is the exchange parameter related to the dead zone residence time. The normalized Laplace transforms $\overline{C}(x,s)$, $\overline{C_s}(x,s)$ are for $-\infty < x < \infty$

$$\overline{C}(x,s) = \frac{u}{\sqrt{u^2 + 4D\left(s + \frac{\varepsilon s}{1 + s\rho}\right)}} \times \exp\left(x\left(u \mp \sqrt{u^2 + 4D\left(s + \frac{\varepsilon s}{1 + s\rho}\right)}\right) / (2D)\right),$$
$$\overline{C_s}(x,s) = \frac{1}{1 + s\rho}\overline{C}(x,s),$$
(58)

with the -sign for $x \ge 0$, the +sign for x < 0. The coefficients a_i , b_i , i = 1, 2, 3, 4 for $\overline{C}(x, s)$ and the moments and cumulants are given in Appendix C.

It is known that the function C(x,t) is given by (see e.g. Hill and Aifantis [5], Walker [21] and van Kooten [17])

$$C(x,t) = w(x,t) \exp\left(-\frac{\varepsilon t}{\rho}\right) \times \int_{0}^{t} w(x,\tau) \exp\left(-\frac{\varepsilon \tau + (t-\tau)}{\rho}\right) \sqrt{\frac{\varepsilon \tau}{\rho^{2}(t-\tau)}} \times I_{1}\left(2\sqrt{\frac{\varepsilon \tau(t-\tau)}{\rho^{2}}}\right) d\tau,$$
(59)

where w(x,t) is given by the normalized function Eq. (4) $(G_0/(nAM) = u)$:

$$w(x,t) = \frac{u}{2\sqrt{\pi Dt}} \exp\left(-\left(\frac{x-ut}{2\sqrt{Dt}}\right)^2\right),\tag{60}$$

and I_1 is the modified Bessel function of the first kind. We compare the different approximations for the analytical solution C(x,t) for the following parameter values

$$\begin{split} D &= 8.64 \ \mathrm{m^2 d^{-1}}, \quad u = 86.4 \ \mathrm{m d^{-1}}, \quad \varepsilon = 0.1 \ [\text{-}], \\ \rho &= 1/43.2 = 0.02315 \ \mathrm{d}. \end{split}$$

For x = 66 m, the cumulants are $\kappa_1 = n_1 = 8.4282\text{E}-01$ d, $\kappa_2 = \sigma_t^2 = 5.6998\text{E}-03$ d², $\kappa_3 = g_t = 2.8999\text{E}-04$ d³, $\kappa_4 = 2.6066\text{E}-05$ d⁴.

First, we compare in Fig. E.5 the exact solution and the approximations based on the Gaussian distribution itself and the Gram-Charlier approximation (28) with one and two additional terms, and with the Edgeworth approximation (30), with three terms. We see that this Edgeworth approximation gives the best fit.

If we now try to approximate the functions C(x,t) by the first two terms in Eq. (40) based on Eq. (42), we have to solve α and β from Eq. (43). See Eq. (56) for the result. In Fig. E.6 we show the results of the numerical evaluation of the first two terms in Eq. (40) in which n_1 , σ_t^2 and $\kappa_3 = g_t$ are given by Eqs. (C.9), (C.10) and (C.11), respectively, and with α and β given by Eq. (56) and λ_3 by the values for α and β (see Eq. (46) for n = 3). For x = 66 these values become $\nu = -1/2$, $\alpha = 74.5231$ d⁻¹, $\beta = 52.0982$ d. See Fig. E.6 for this approximation based on Eq. (42), *i.e.* the first approximation and the first two terms in Eq. (40), i.e. the second approximation.

It is also possible to fit the parameter ν using the knowledge κ_3 . So, we try to fit the parameters ν , α , and β using the knowledge of κ_1 , κ_2 and κ_3 . It turns out that it is not possible to fit the three parameters ν , α and β such that all differences $\kappa_1 - \lambda_1$, $\kappa_2 - \lambda_2$ and $\kappa_3 - \lambda_3$ become reasonable small. We find the values $\nu = -12.2303$, $\alpha = 67.6214 \text{ d}^{-1}$, $\beta = 57.0428 \text{ d}$, and these values imply $\lambda_3 = 1.1896\text{E}-04 \text{ d}^3$, which is less than half the value for κ_3 (2.8999E-04 d³). In Fig. E.7 the exact solution is compared with Eq. (47) and in the third approximation based on Eq. (38) with the first two terms in the sum (using $P_2(s)$ from Eq. (36) with the fourth cumulants and the fourth derivative (see below) but neglecting the term with $(\kappa_3 - \lambda_3)^2 \simeq 2.92\text{E}-08 \text{ d}^6$ and so the sixth derivative). We included the contribution of $P_1(s)$ with the third derivative, due to the poor fit for κ_3 with λ_3 . The factor for the fourth derivative becomes $(\kappa_4 - \lambda_4)/24$. Now, κ_4 is given by Eq. (C.13) and λ_4 together with the fourth derivative of Z(t) has been specified in Appendix B.

It turns out that in this problem it is difficult to find an appropriate value for ν . If one compares the two figures one observes that reasonable fits are possible for different values for ν . If one fixes $\nu = -1/2$, it is also possible to include the correction based on the fourth cumulant. That has been done in Fig. E.8. The corresponding fourth cumulant of the approximation is $\lambda_4 = 3.8851E-06 \text{ d}^4$ (to be compared with $\kappa_4 = 2.6066E-05 \text{ d}^4$). In Fig. E.8 one can easily see the improvement by taking into account the term based on the fourth cumulant and the fourth and sixth derivatives.

In Fig. E.9 we show the difference between the exact solution and four different approximations (Gram-Charlier, Edgeworth, Moving Gaussian ($\nu = -1/2$) and Generalized Moving Gaussian distribution ($\nu = -12.2303$)), with all terms shown before. We see that the Moving Gaussian distributions approximations are performing almost the same and are comparable to the Edgeworth approximation.

In Fig. E.10 we show the difference between the two Moving Gaussian distribution approximations based on $\nu = -1/2$ and $\nu = -12.2303$, which is very small, and the differences between the exact solution and each of the two Moving Gaussian distribution approximations, which is each of the order of 2E-02. The results for $\nu = -1/2$ and $\nu = -12.2303$ hardly differ.

In Fig. E.11 we show in detail the performance of two approximations which performed the best: the Edgeworth approximation and the Generalized Moving Gaussian approximation based on the third and fourth cumulants (and so on the sixth derivative). It is difficult to say which approximation is the best.

See Table D.2 for the parameters values and cumulants for the different approximations.

6.3. Example 3

We use measurements from the field (pers. comm. M. Westhoff) where it was the purpose to determine the discharge of a stream. By throwing instantly an amount of salt (100 gr) into a stream, one can measure the concentration using the electric conductivity (EC) given in micro Siemens/cm at distances downstream (10 m to 20 m) as a function of the time. The discharge of the stream was around 0.001 m³s⁻¹. The measured values are corrected for the background EC. In essence, one measures an impulse response curve. The measurements have been normalized by dividing through the 0-order moment M_0 , Eq. (5), calculated numerically. Subsequently, the first four cumulants have been found numerically. Based on these values in Fig. E.12 the Edgeworth approximation Eq. (30) with three terms has been compared with the Moving Gaussian distribution, with $\nu = -1/2$, $\alpha = 0.7502 \text{ d}^{-1}$, and $\beta = 2.3454 \text{ d}$, based on Eq. (56), and with the Generalized Moving Gaussian distribution with optimized $\nu = -3.1657$, $\alpha = 0.3402 \text{ d}^{-1}$, and $\beta = 5.5578 \text{ d}$. For the Generalized Moving Gaussian distribution we have fitted the parameters ν , α , and β using the knowledge of $\kappa_1 = n_1$, $\kappa_2 = \sigma_t^2$ and $\kappa_3 = g_t$. We see that in this case the Edgeworth approximation is performing about the same as the Generalized Moving Gaussian distribution for values of t in the interval (1, 4), but for higher values of t it shows wiggles, while the Generalized Moving Gaussian distribution does not.

See Table D.3 for the parameters values and cumulants for the different approximations.

7. Conclusions

We presented an approach to approximate a given impulse response curve using his cumulants by a distribution function (named Generalized Moving Gaussian distribution) which represents better the behaviour for shorter times than the usual Gaussian distribution.

We have shown through a number of experiments that the proposed approximative Generalized Moving Gaussian distribution for convection-diffusion type impulse responses performs similarly or better when compared to the expansions based on the pure Gaussian distribution.

For a given impulse response curve it might be worthwile to determine numerically the normalization M_0 and the first three central moments or cumulants, $n_1(x)$, $\sigma_t^2(x)$, and $g_t(x)$. Using these quantities one can try to find the parameters α , β and ν of the Generalized Moving Gaussian distribution. To go beyond the third cumulant is in practical cases not possible due to the inaccuracies of the measurements.

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Appendix A. Moving Gaussian distribution

The Moving Gaussian distribution is defined as

$$Z(t) = \frac{1}{M_0} t^{-1/2} \exp\left(-\left(\sqrt{\alpha t} - \sqrt{\beta/t}\right)^2\right),$$

$$0 < t < \infty, \quad \alpha > 0, \quad \beta > 0,$$
(A.1)

$$M_0 = \sqrt{\frac{\pi}{\alpha}}.\tag{A.2}$$

The cumulants are

$$\lambda_n = \frac{1}{2} \alpha^{-n} \Gamma(n) - 2 \alpha^{1/2 - n} \beta^{1/2} \left(-\frac{1}{2} \right)_n.$$
(A.3)

$$\lambda_1 = n_1 = \frac{1}{2\alpha} + \sqrt{\frac{\beta}{\alpha}}.\tag{A.4}$$

$$\lambda_2 = \sigma_t^2 = \frac{1}{2\alpha^2} + \frac{1}{2\alpha}\sqrt{\frac{\beta}{\alpha}}.$$
(A.5)

$$\lambda_3 = \frac{1}{\alpha^3} + \frac{3}{4\alpha^2} \sqrt{\frac{\beta}{\alpha}}.$$
(A.6)

$$\lambda_4 = rac{3}{lpha^4} + rac{15}{8lpha^3}\sqrt{rac{eta}{lpha}}.$$

The following derivatives have been found using the Formula Manipulation Package of Maple[®].

$$\frac{d^3}{dt^3} Z(t) = Z(t) \times$$
(A.7)
$$(8\alpha^3 t^6 + 12\alpha^2 t^5 - 24\alpha^2 \beta t^4 + 18\alpha t^4$$

$$-72\alpha\beta t^{3} + 15t^{3} + 24\alpha\beta^{2}t^{2} - 90\beta t^{2} + 60\beta^{2}t - 8\beta^{3})/(8t^{6}).$$

$$\begin{aligned} \frac{d^4}{dt^4} Z(t) &= Z(t) \times \\ \left(16\alpha^4 t^8 + 32\alpha^3 t^7 - 64\alpha^3\beta t^6 + 72\alpha^2 t^6 \right) \\ &- 288\alpha^2\beta t^5 + 120\alpha t^5 + 96\alpha^2\beta^2 t^4 \\ &- 720\alpha\beta t^4 + 105t^4 + 480\alpha\beta^2 t^3 - 840\beta t^3 \\ &- 64\alpha\beta^3 t^2 + 840\beta^2 t^2 - 224\beta^3 t + 16\beta^4 \right) / (16t^8). \end{aligned}$$

$$\begin{split} \frac{d^{6}}{dt^{6}}Z(t) &= Z(t) \times \\ \left(64\alpha^{6}t^{12} + 192\alpha^{5}t^{11} - 384\alpha^{5}\beta t^{10} \right. \\ &+ 720\alpha^{4}t^{10} - 2880\alpha^{4}\beta t^{9} + 2400\alpha^{3}t^{9} \\ &+ 960\alpha^{4}\beta^{2}t^{8} - 14400\alpha^{3}\beta t^{8} + 6300\alpha^{2}t^{8} \\ &+ 9600\alpha^{3}\beta^{2}t^{7} - 50400\alpha^{2}\beta t^{7} + 11340\alpha t^{7} \\ &- 1280\alpha^{3}\beta^{3}t^{6} + 50400\alpha^{2}\beta^{2}t^{6} - 113400\alpha\beta t^{6} \\ &+ 10395t^{6} - 13440\alpha^{2}\beta^{3}t^{5} + 151200\alpha\beta^{2}t^{5} \\ &- 124740\beta t^{5} + 960\alpha^{2}\beta^{4}t^{4} - 60480\alpha\beta^{3}t^{4} \\ &+ 207900\beta^{2}t^{4} + 8640\alpha\beta^{4}t^{3} - 110880\beta^{3}t^{3} \\ &- 384\alpha\beta^{5}t^{2} + 23760\beta^{4}t^{2} - 2112\beta^{5}t + 64\beta^{6} \right) / \\ (64t^{12}). \end{split}$$

Appendix B. Generalized Moving Gaussian distribution

The Generalized Moving Gaussian distribution is defined as

$$Z(t) = \frac{1}{M_0} t^{\nu} \exp\left(-\left(\sqrt{\alpha t} - \sqrt{\beta/t}\right)^2\right),$$

$$0 < t < \infty, \ \alpha > 0, \ \beta > 0, \ -\infty < \nu < \infty,$$
(B.1)

(A.8)

(A.9)

$$M_0 = 2\left(\frac{\beta}{\alpha}\right)^{(\nu+1)/2} \exp\left(2\sqrt{\alpha\beta}\right) K_{\nu+1}\left(2\sqrt{\alpha\beta}\right).$$
(B.2)

The following cumulants and derivatives have been found using the Formula Manipulation Package of $Maple^{\textcircled{R}}$.

$$\lambda_{1} = n_{1} =$$

$$\frac{\nu + 1}{\alpha} + \frac{\sqrt{\alpha\beta}}{\alpha} \left(\frac{K_{\nu} \left(2\sqrt{\alpha\beta} \right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta} \right)} \right).$$
(B.3)

$$\lambda_{2} = \sigma_{t}^{2} = \frac{\alpha\beta + \nu + 1}{\alpha^{2}}$$

$$- \frac{\nu\sqrt{\alpha\beta}}{\alpha^{2}} \left(\frac{K_{\nu} \left(2\sqrt{\alpha\beta} \right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta} \right)} \right)$$

$$- \frac{\beta}{\alpha} \left(\frac{K_{\nu} \left(2\sqrt{\alpha\beta} \right)}{K_{\nu+1} \left(2\sqrt{\alpha\beta} \right)} \right)^{2}.$$
(B.4)

$$\lambda_{3} = \frac{2(\nu+1) - \alpha\beta(\nu-1)}{\alpha^{3}} - \frac{(2\alpha\beta - \nu(\nu-1))\sqrt{\alpha\beta}}{\alpha^{3}} \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right) + \frac{3\nu\beta}{\alpha^{2}} \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right)^{2} + \frac{2\beta\sqrt{\alpha\beta}}{\alpha^{2}} \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right)^{3}.$$

$$\lambda_{4} = \frac{-2\alpha^{2}\beta^{2} + \alpha\beta(\nu-1)(\nu-2) + 6(1+\nu)}{\alpha^{4}} + \frac{(2\alpha\beta(4\nu-1) - \nu(\nu-1)(\nu-2))\sqrt{\alpha\beta}}{\alpha^{4}} \times \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right) + \frac{(8\alpha\beta - \nu(7\nu-4))\beta}{\alpha^{3}} \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right)^{2} - \frac{12\nu\beta\sqrt{\alpha\beta}}{\alpha^{3}} \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right)^{3} - \frac{6\beta^{2}}{\alpha^{2}} \left(\frac{K_{\nu}\left(2\sqrt{\alpha\beta}\right)}{K_{\nu+1}\left(2\sqrt{\alpha\beta}\right)}\right)^{4}.$$

$$\begin{split} &\frac{d^3}{dt^3} Z(t) = Z(t) \times \\ &- \left(\alpha^3 t^6 - 3\nu \alpha^2 t^5 - 3\alpha^2 \beta t^4 + 3\nu(\nu - 1)\alpha t^4 \right. \\ &+ 6(\nu - 1)\alpha \beta t^3 - \nu(\nu - 1)(\nu - 2)t^3 \\ &+ 3\alpha \beta^2 t^2 - 3(\nu - 1)(\nu - 2)\beta t^2 \\ &- 3(\nu - 2)\beta^2 t - \beta^3 \right) / t^6. \end{split}$$

$$\begin{split} & \frac{d^4}{dt^4} Z(t) = Z(t) \times \\ & \left(\alpha^4 t^8 - 4\alpha^3 \nu t^7 - 4\alpha^3 \beta t^6 + 6\alpha^2 \nu (\nu - 1) t^6 \right. \\ & \left. + 12\alpha^2 (\nu - 1)\beta t^5 - 4\alpha \nu (\nu - 1) (\nu - 2) t^5 \right. \\ & \left. - 12\alpha\beta (\nu - 1) (\nu - 2) t^4 + \nu (\nu - 1) (\nu - 2) (\nu - 3) t^4 \right. \\ & \left. - 12\alpha\beta^2 (\nu - 2) t^3 + 4\beta (\nu - 1) (\nu - 2) (\nu - 3) t^3 \right. \\ & \left. - 4\alpha\beta^3 t^2 + 6\beta^2 (\nu - 2) (\nu - 3) t^2 \right. \\ & \left. + 4\beta^3 (\nu - 3) t + \beta^4 \right) / t^8. \end{split}$$

(B.5)

(B.6)

(B.7)

(B.8)

$$\begin{split} &\frac{d^6}{dt^6}Z(t)=Z(t)\times\\ &\left(\alpha^6t^{12}-6\alpha^5\nu t^{11}-6\alpha^5\beta t^{10}+15\alpha^4\nu(\nu-1)t^{10}\right.\\ &+30\alpha^4\beta(\nu-1)t^9-20\alpha^3\nu(\nu-1)(\nu-2)t^9\\ &+15\alpha^4\beta^2t^8-60\alpha^3\beta(\nu-1)(\nu-2)t^8\\ &+15\alpha^2\nu(\nu-1)(\nu-2)(\nu-3)t^8\\ &-60\alpha^3\beta^2(\nu-2)t^7+60\alpha^2\beta(\nu-1)(\nu-2)(\nu-3)t^7\\ &-6\alpha\nu(\nu-1)(\nu-2)(\nu-3)(\nu-4)t^7\\ &-20\alpha^3\beta^3t^6+90\alpha^2\beta^2(\nu-2)(\nu-3)t^6\\ &-30\alpha\beta(\nu-1)(\nu-2)(\nu-3)(\nu-4)(\nu-5)t^6\\ &+60\alpha^2\beta^3(\nu-3)t^5-60\alpha\beta^2(\nu-2)(\nu-3)(\nu-4)t^5\\ &+6\beta(\nu-1)(\nu-2)(\nu-3)(\nu-4)(\nu-5)t^5\\ &+15\alpha^2\beta^4t^4-60\alpha\beta^3(\nu-3)(\nu-4)(\nu-5)t^4\\ &-30\alpha\beta^4(\nu-4)t^3+20\beta^3(\nu-3)(\nu-4)(\nu-5)t^3\\ &-6\alpha\beta^5t^2+15\beta^4(\nu-4)(\nu-5)t^2\\ &+6\beta^5(\nu-5)t+\beta^6)/t^{12}. \end{split}$$

We remark that all these results for $\nu = -1/2$ reduce to what has been found for the Moving Gaussian $\operatorname{distribution}.$

Appendix C. Coefficients a_i , b_i and Cumulants for Example 6.2

The coefficients $a_i, b_i, i = 1, 2, 3, 4$ for $\overline{C}(x, s)$ (see Eqs. (13), (14), and (58)) are

$$a_1 = -\frac{2D\left(1+\varepsilon\right)}{u^2},\tag{C.1}$$

$$a_2 = \frac{2D\varepsilon\rho}{u^2} + \frac{6D^2(1+\varepsilon)^2}{u^4},\tag{C.2}$$

$$a_{3} = -\frac{2D\varepsilon\rho^{2}}{u^{2}} - \frac{12D^{2}\varepsilon(1+\varepsilon)\rho}{u^{4}} - \frac{20D^{3}(1+\varepsilon)^{3}}{u^{6}},$$
(C.3)

$$a_4 = \frac{2D\varepsilon\rho^3}{u^2} + \frac{6D^2\varepsilon(2+3\varepsilon)\rho^2}{u^4}$$
(C.4)

$$+\frac{60D^3\varepsilon(1+\varepsilon)^2\rho}{u^6} + \frac{70D^4(1+\varepsilon)^4}{u^8},$$

$$1+\varepsilon$$

$$b_1 = -\frac{1+\varepsilon}{u},$$
(C.5)
$$b_2 = \frac{\varepsilon\rho}{u} + \frac{D(1+\varepsilon)^2}{u^3},$$
(C.6)

$$p_2 = \frac{\varepsilon\rho}{u} + \frac{D(1+\varepsilon)^2}{u^3},\tag{C.6}$$

(B.9)

$$b_{3} = -\frac{\varepsilon\rho^{2}}{u} - \frac{2D\varepsilon(1+\varepsilon)\rho}{u^{3}}$$

$$-\frac{2D^{2}(1+\varepsilon)^{3}}{u^{5}},$$
(C.7)

$$b_4 = \frac{\varepsilon \rho^3}{u} + \frac{D\varepsilon (2+3\varepsilon)\rho^2}{u^3} + \frac{6D^2\varepsilon (1+\varepsilon)^2 \rho}{u^5} + \frac{5D^3 (1+\varepsilon)^4}{u^7},$$
(C.8)

and the moments and cumulants are (based on Eq. (15)):

$$n_1(x) = \frac{(1+\varepsilon)x}{u} + 2\frac{D(1+\varepsilon)}{u^2},\tag{C.9}$$

$$\sigma_t^2(x) = 4 \frac{D\varepsilon\rho}{u^2} + 8 \frac{D^2(1+\varepsilon)^2}{u^4} + 2\left(\frac{\varepsilon\rho}{u} + \frac{D(1+\varepsilon)^2}{u^3}\right)x$$
(C.10)

$$g_t(x) = 12 \frac{D\varepsilon\rho^2}{u^2} + 48 \frac{D^2\varepsilon(1+\varepsilon)\rho}{u^4} + 64 \frac{D^3(1+\varepsilon)^3}{\varepsilon}$$
(C.11)

$$+ \left(6\frac{\varepsilon\rho^2}{u} + 12\frac{D\varepsilon\rho(1+\varepsilon)}{u^3} + 12\frac{D^2(1+\varepsilon)^3}{u^5}\right)x,$$

$$D\varepsilon a^3 = D^2\varepsilon(4+7\varepsilon)a^2$$

$$\mu_{4}(x) = 48 \frac{D\varepsilon \rho^{*}}{u^{2}} + 48 \frac{D^{*}\varepsilon (4 + i\varepsilon)\rho^{*}}{u^{4}}$$

$$+ 960 \frac{D^{3}\varepsilon (1 + \varepsilon)^{2} \rho}{u^{6}} + 960 \frac{D^{4} (1 + \varepsilon)^{4}}{u^{8}}$$

$$+ \left(24 \frac{\varepsilon \rho^{3}}{u} + 24 \frac{D\varepsilon (2 + 5\varepsilon)\rho^{2}}{u^{3}} + 216 \frac{D^{3} (1 + \varepsilon)^{4}}{u^{7}}\right) x$$

$$+ 12 \left(\frac{\varepsilon \rho}{u} + \frac{D(1 + \varepsilon)^{2}}{u^{3}}\right)^{2} x^{2},$$
(C.12)

$$\kappa_{4}(x) = \mu_{4}(x) - 3\sigma_{t}^{4}(x) =$$
(C.13)
$$48 \frac{D\varepsilon\rho^{3}}{u^{2}} + 96 \frac{D^{2}\varepsilon(2+3\varepsilon)\rho^{2}}{u^{4}} + 768 \frac{D^{3}\varepsilon(1+\varepsilon)^{2}\rho}{u^{6}} + 768 \frac{D^{4}(1+\varepsilon)^{4}}{u^{8}} + \left(24 \frac{\varepsilon\rho^{3}}{u} + 24 \frac{D\varepsilon(2+3\varepsilon)\rho^{2}}{u^{3}} + 144 \frac{D^{2}\varepsilon(1+\varepsilon)^{2}\rho}{u^{5}} + 120 \frac{D^{3}(1+\varepsilon)^{4}}{u^{7}}\right) x.$$

Appendix D. Tables

	ν	$\alpha [\mathrm{d}^{-1}]$	β [d]	κ_1 [d]	$\kappa_2 [\mathrm{d}^2]$	$\kappa_3 [\mathrm{d}^3]$	$\kappa_4 [\mathrm{d}^4]$
Exact				15	31	188	1890
				λ_1 [d]	$\lambda_2 [\mathrm{d}^2]$	$\lambda_3 [\mathrm{d}^3]$	$\lambda_4 [\mathrm{d}^4]$
Fig. E.2	-0.5	0.25	49	16	36	232	2448
Fig. E.3	-0.5	0.2716	47.0361	15.0000	31.0000	183.6664	1782.29
Fig. E.4	-1.0116	0.2564	50.8088	15.0000	31.0000	188.0000	1883.95

Table D.1: Values for Example 6.1

Table D.2: Values for Example 6.2

	ν	$\alpha [\mathrm{d}^{-1}]$	β [d]	κ_1 [d]	$\kappa_2 [\mathrm{d}^2]$	$\kappa_3 [\mathrm{d}^3]$	$\kappa_4 [\mathrm{d}^4]$
Exact				8.4282E - 01	5.6998E - 03	$2.8999 \text{E}{-04}$	2.6066E - 05
				λ_1 [d]	$\lambda_2 [\mathrm{d}^2]$	$\lambda_3 [\mathrm{d}^3]$	$\lambda_4 [\mathrm{d}^4]$
Fig. E.6	-0.5	74.5231	52.0982	8.4282E - 01	5.6998E - 03	1.1533E - 04	
Fig. E.7	-12.2303	67.6214	57.0428	8.4282E - 01	5.6998E - 03	1.1896E - 04	4.1856E - 06
Fig. E.8	-0.5	74.5231	52.0982	8.4282E - 01	5.6998E - 03	1.1533E - 04	3.8851E - 06

Table D.3: Values for Example 6.3

	ν	$\alpha [d^{-1}]$	β [d]	κ_1 [d]	$\kappa_2 [\mathrm{d}^2]$	$\kappa_3 [\mathrm{d}^3]$	$\kappa_4 [\mathrm{d}^4]$
Exact				2.4346	2.0668	6.3502	31.4236
				λ_1 [d]	$\lambda_2 [\mathrm{d}^2]$	$\lambda_3 [\mathrm{d}^3]$	$\lambda_4 [\mathrm{d}^4]$
Moving Gaussian	-0.5	0.7502	2.3454	2.4346	2.0668	4.7246	17.3231
Gen. Mov. Gaussian	-3.1657	0.3402	5.5578	2.4346	2.0669	6.3501	35.2669

Appendix E. Figures



Figure E.1: Example 6.1. Exact solution C(x,t), Eq. (53), for x = 1.4 m with parameters $D = 0.01 \text{ m}^2 \text{d}^{-1}$, $u = 0.1 \text{ md}^{-1}$, compared with the Gaussian distribution, with the Gram-Charlier approximation Eq. (28) with one and two terms and with the Edgeworth approximation Eq. (30).



Figure E.2: Example 6.1. Exact solution C(x,t), Eq. (53), for x = 1.4 m with parameters $D = 0.01 \text{ m}^2 \text{d}^{-1}$, $u = 0.1 \text{ m}\text{d}^{-1}$, compared with the first approximation Eq. (42) of the Moving Gaussian distribution Z(x,t) and second approximation, *i.e.* first two terms in Eq. (40). The values for the parameters are $\alpha = u^2/(4D) = 0.25 \text{ d}^{-1}$, $\beta = x^2/(4D) = 49 \text{ d}$, $\nu = -1/2$.



Figure E.3: Example 6.1. Exact solution C(x,t), Eq. (53), for x = 1.4 m with parameters $D = 0.01 \text{ m}^2 \text{d}^{-1}$, $u = 0.1 \text{ md}^{-1}$, compared with the first approximation Eq. (42) of the Moving Gaussian distribution Z(x,t) and second approximation, *i.e.* the first two terms in Eq. (40) with optimized values for α and β . The values for the parameters are $\alpha = 0.2716 \text{ d}^{-1}$, $\beta = 47.0361 \text{ d}$, $\nu = -1/2$.



Approximation normed exact with Generalized Moving Gaussian Approximation with optimized $\alpha = 0.25639$, $\beta = 50.8088$, $\nu = -1.0116$

Figure E.4: Example 6.1. Exact solution C(x,t), Eq. (53), for x = 1.4 m with parameters $D = 0.01 \text{ m}^2 \text{d}^{-1}$, $u = 0.1 \text{ m}^{-1}$, compared with the first approximation Eq. (47) of the Generalized Moving Gaussian distribution Z(x,t) and the third approximation based on the third and fourth cumulants and the third, fourth and sixth derivatives (see Eq. (36) for $P_2(s)$). There is hardly to see any difference between the exact solution and the approximations. The values for the parameters are $\alpha = 0.2564$ d⁻¹, $\beta = 50.8088$ d, $\nu = -1.0112$.



Figure E.5: Example 6.2. Exact solution C(x,t), Eq. (59), for x = 66 m with parameters $D = 8.64 \text{ m}^2\text{d}^{-1}$, $u = 86.4 \text{ m}^{-1}$, $\varepsilon = 0.1$, $\rho = 0.02315$ d, compared with the Gaussian distribution, with the Gram-Charlier approximation Eq. (28) with one and two terms and with the Edgeworth approximation Eq. (30), with three terms.



Figure E.6: Example 6.2. Exact solution C(x,t), Eq. (59), for x = 66 m with parameters $D = 8.64 \text{ m}^2 \text{d}^{-1}$, $u = 86.4 \text{ m}^{-1}$, $\varepsilon = 0.1$, $\rho = 0.02315$ d, compared with the first approximation Eq. (42) of the Moving Gaussian distribution Z(x,t) and second approximation, *i.e.* first two terms in Eq. (40).



Figure E.7: Example 6.2. Exact solution C(x,t), Eq. (59), for x = 66 m with parameters $D = 8.64 \text{ m}^2 \text{d}^{-1}$, $u = 86.4 \text{ m}^{-1}$, $\varepsilon = 0.1$, $\rho = 0.02315$ d, compared with the first approximation Eq. (47) of the Generalized Moving Gaussian distribution Z(x,t), the second approximation based on the third cumulant, the third approximation based on the fourth cumulant and fourth derivative, and the fourth approximation based on the fourth cumulant and fourth derivative (see Eq. (36) for $P_1(s)$ and $P_2(s)$). The third cumulants with corresponding derivatives have been taken into account because of the moderate fit for the κ_3 with λ_3 (see text).



Figure E.8: Example 6.2. Exact solution C(x,t), Eq. (59), for x = 66 m with parameters $D = 8.64 \text{ m}^2\text{d}^{-1}$, $u = 86.4 \text{ m}^{-1}$, $\varepsilon = 0.1$, $\rho = 0.02315$ d, compared with the second approximation, *i.e.* first two terms in Eq. (40) and the third approximation Eq. (40), based on the fourth cumulant and fourth derivative without and with the term with the sixth derivative.



Figure E.9: Example 6.2. Absolute difference between the exact solution and four different approximations (Gram-Charlier, Edgeworth, Moving Gaussian ($\nu = -1/2$) and Generalized Moving Gaussian distribution ($\nu = -12.2303$)), with all terms shown before.



Figure E.10: Example 6.2. Three curves: 1. Difference between the Moving Gaussian ($\nu = -1/2$) and the Generalized Moving Gaussian distribution ($\nu = -12.2303$) based on all terms up to the fourth cumulant. 2. Difference between the exact solution and the approximation based on the Moving Gaussian distribution. 3. Difference between the exact solution and the approximation based on the Generalized Moving Gaussian distribution.



Figure E.11: Example 6.2. Comparison between the Edgeworth approximation and the Generalized Moving Gaussian approximation. Shown is the absolute difference between the approximation and the exact solution.



Approximation normed measurements with (Generalized) Moving Gaussian Approximation with ν = -0.5 and with optimized α = 0.34024, β = 5.5578, ν = -3.1657

Figure E.12: Example 6.3. Comparison between the Edgeworth approximation and the (Generalized) Moving Gaussian approximations for the salt experiment.