Physically Parameterized Fourth-Order Retention and Scaling Analysis in Bi-Flux Turbulent Dispersion for the Planetary Boundary Layer☆

Parametrização Física do Termo de Retenção de Quarta Ordem e Análise de Escala na Dispersão Turbulenta Bi-Fluxo para a Camada Limite Planetária

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Abstract

Classical Fickian diffusion theory often fails to capture the complex structure of turbulent dispersion in the Planetary Boundary Layer (PBL), particularly regarding the description of memory effects and plume retention. This work investigates the Bi-flux dispersion theory, which extends the advection-diffusion equation by incorporating a fourth-order derivative term to model non-Fickian transport phenomena. Unlike previous analytical approaches limited to constant coefficients, this study develops a numerical model using a high-order implicit finite difference scheme that incorporates realistic height-dependent profiles for both wind speed and vertical eddy diffusivity. A novel physical parameterization is proposed for the fourth-order retention coefficient, defined as $K_{z2} \propto u_*|L|^3$, linking the non-Fickian mechanism to the local atmospheric turbulence scales (friction velocity and Monin-Obukhov length). The model was validated against the Copenhagen Experiment dataset. A sensitivity analysis of the partition parameter β revealed an optimal performance at $\beta = 0.99$. The results demonstrate that the physically parameterized Bi-flux model significantly outperforms the classical Fickian formulation ($\beta = 1.0$), achieving a 24.1% reduction in the Normalized Mean Square Error (NMSE) and a 70.4% improvement in the Fractional Standard Deviation (FS). These findings confirm that the inclusion of an atmospheric-scaled fourth-order term effectively preserves the internal variance of the plume, offering a superior predictive capability for pollutant dispersion in unstable boundary layers.

Keywords

Atmospheric Dispersion • Bi-Flux Model • Anomalous Diffusion

Resumo

A teoria clássica de difusão Fickiana frequentemente falha em capturar a estrutura complexa da dispersão turbulenta na Camada Limite Planetária (CLP), particularmente no que diz respeito à descrição de efeitos de memória e retenção da pluma. Este trabalho investiga a teoria de dispersão Bi-fluxo, que estende a equação de advecção-difusão ao incorporar um termo de derivada de quarta ordem para modelar fenômenos de transporte não-Fickianos. Diferentemente de abordagens analíticas anteriores limitadas a coeficientes constantes, este estudo desenvolve um

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modelo numérico utilizando um esquema implícito de diferenças finitas de alta ordem que incorpora perfis realistas dependentes da altura tanto para a velocidade do vento quanto para a difusividade turbulenta vertical. Uma nova parametrização física é proposta para o coeficiente de retenção de quarta ordem, definido como $K_{z2} \propto u_*|L|^3$, conectando o mecanismo não-Fickiano às escalas locais de turbulência atmosférica (velocidade de fricção e comprimento de Monin-Obukhov). O modelo foi validado com o conjunto de dados do Experimento de Copenhague. Uma análise de sensibilidade do parâmetro de partição β revelou um desempenho ótimo em $\beta = 0{,}99$. Os resultados demonstram que o modelo Bi-fluxo fisicamente parametrizado supera significativamente a formulação Fickiana clássica ($\beta = 1{,}0$), alcançando uma redução de 24,1% no Erro Quadrático Médio Normalizado (NMSE) e uma melhoria de 70,4% no Desvio Padrão Fracionário (FS). Essas descobertas confirmam que a inclusão de um termo de quarta ordem com escalonamento atmosférico preserva efetivamente a variância interna da pluma, oferecendo uma capacidade preditiva superior para a dispersão de poluentes em camadas limites instáveis.

Palavras-chave

Dispersão Atmosférica • Modelo bi-fluxo • Difusão Anômala

1 Introduction

The modelling of atmospheric pollutant dispersion constitutes an indispensable tool for air quality management, risk assessment, and urban planning [1, 2], particularly in the face of increasing urbanisation. The theoretical framework for describing contaminant transport in the atmosphere is founded upon the advection-diffusion equation [3, 4, 5]. Its application to turbulent flows, however, introduces the closure problem, which necessitates a parameterisation of the turbulent concentration flux [6, 7]. The most conventional solution to this challenge is K-theory, also known as the Gradient Diffusion Hypothesis (GDH), which posits a turbulent flux proportional to the mean concentration gradient, analogous to Fick's law [6]. Notwithstanding its extensive application, the classical Fickian paradigm exhibits significant limitations, often proving inadequate for representing the complex dynamics of dispersion in realistic scenarios, such as within urban topologies or for the phenomenon of anomalous diffusion [5, 6, 8, 9]. Anomalous diffusion, an inherent characteristic of turbulence within the PBL [5, 9], thereby challenges the premises of traditional diffusion models.

The need to transcend the insufficiencies of Fickian models has driven the development of alternative formulations as a fundamental step in dispersion modeling [5, 10, 11]. In this context, the bi-flux dispersion theory, proposed by Bevilacqua and co-workers, presents itself as an alternative with more physically robust foundations [10, 11]. The central premise of this theory is that the ensemble of diffusing particles may be segmented into two fluxes with distinct dynamics: a primary flux, corresponding to the fraction (β) that adheres to Fick's law, and a secondary flux for the remaining fraction ($1 - \beta$), which is governed by a separate transport law associated with temporary retention phenomena [5, 12, 13]. The mathematical formalisation of this dynamic balance is achieved through the incorporation of a fourth-order differential term into the transport equation [11, 13, 14]. The efficacy of analogous models, which integrate the bi-flux mechanism with other approaches such as fractional derivatives, has been demonstrated in reference works, including that of Palmeira et al. [5]. However, the attainment of analytical solutions in such studies has necessitated the adoption of a constant turbulent diffusivity coefficient, a hypothesis that constrains the representation of vertically heterogeneous turbulence.

This work extends prior research by considering the development and validation of a numerical model for the bi-flux equation, which incorporates a more physically realistic representation of the turbulence structure within the PBL. A key innovation of this study is the substitution of the constant velocity and diffusivity hypothesis with height-dependent profiles for both wind speed, $U(z)$, and vertical turbulent diffusivity, $K_{z1}(z)$. Furthermore, this study conducts an in-depth analysis of the K_{z2} parameter, investigating its order of magnitude relative to the second-order (Fickian) terms and proposing different formulations for its definition within the model. This physical analysis seeks to understand the impact of the fourth-order derivative on plume dynamics, evaluating how different choices for K_{z2} influence the accuracy of the results. The governing equation is solved numerically via an implicit finite difference scheme. As validation, the model is evaluated with the Copenhagen Experiment dataset.

The subsequent sections of this paper detail the methodology, results, and conclusions of the study. Section 2 describes the Copenhagen Experiment, followed by the mathematical formulation and numerical implementation of the model, including the proposed parameterisation for the non-Fickian coefficient. Section 3 presents and discusses the simulation results, comprising a sensitivity analysis of the β parameter and the statistical validation of the model against experimental data. Finally, Section 4 summarises the main conclusions and implications of this work.

2 Materials and Methods

This section outlines the methodology employed in this study. It is organised into two main parts: first, a description of the experimental campaign from which the validation data is sourced; and second, a detailed presentation of the mathematical and numerical framework of the bi-flux dispersion model.

2.1 Case Study Description

The case study is based on the Copenhagen Tracer Experiment, a full-scale measurement campaign conducted between 1978 and 1979 in Denmark. Its primary objective was to investigate the atmospheric dispersion of pollutants from an elevated source within a residential urban environment, an area characterised by a surface roughness length of approximately 0.6 metres. Throughout the experiments, the inert tracer gas, sulphur hexafluoride (SF_6), was released from a 115-metre tower. Concentrations were subsequently measured by samplers positioned in arcs at ground level, at downwind distances of 2 to 6 kilometres [3, 5, 15, 16].

A notable feature of the campaign, particularly for its time, was the collection of a detailed meteorological dataset. This included measurements of the boundary layer height (h) and the Obukhov length (L), which are fundamental parameters for characterising atmospheric turbulence [15, 16]. The experiments were conducted under atmospheric conditions ranging from near-neutral to slightly convective. Table 1 collates the values for boundary layer height, Obukhov length, friction velocity (u_*), emission rate (Q), and normalised concentration (c/Q), which were extracted from Palmeira et al. [5], Gryning and Lyck [15], Gryning et al. [16] and Gryning and Lyck [17], and used in the simulations for this study.

2.2 The Mathematical Model

Modelling dispersion in complex media often requires formulations that transcend classical Fickian theory, which postulates a diffusive flux proportional to the concentration gradient. To account for non-Fickian phenomena, such as the temporary retention of particles, the present work adopts the bi-flux dispersion theory, as developed by Jiang et al. [11] and Bevilacqua et al. [14]. The central postulate of this theory is that the ensemble of diffusing particles is not homogeneous but is instead partitioned into two subsets with distinct transport behaviours: a primary flux, representing the particle fraction (β) that adheres to the conventional Fickian mechanism; and a secondary flux, corresponding to the complementary fraction $(1-\beta)$, which is governed by a distinct transport law associated with phenomena such as entrapment in states of lower mobility. The interaction and balance between these two competing transport regimes are mathematically formalised by the introduction of a fourth-order differential term into the advection-diffusion equation [5, 12, 13, 14]. The governing equation, Eq. (1), herein is therefore analogous to that proposed by Palmeira et al. [5]; however, it incorporates a modification since the eddy diffusivity (K_z) is a function of height ($K_z(z)$), thereby allowing for a more realistic representation of the turbulence structure within the PBL. The advection-diffusion equation with anomalous diffusion is given by

$$U\frac{\partial C}{\partial x} = \beta\frac{\partial}{\partial z}\left(K_{z1}\frac{\partial C}{\partial z}\right) - K_{z2}\beta(1-\beta)\frac{\partial^4 C}{\partial z^4} + Q\delta(x-x_s)\delta(z-z_s) \tag{1}$$

where C represents the mean pollutant concentration. The parameters β and $\beta(1-\beta)$ govern the balance between diffusion and retention, respectively, with K_{z2} being the coefficient associated with the retention term. Initially, for validation purposes, this parameter is assumed to be unity ($K_{z2} = 1$) following Palmeira et al. [5]. However, this study subsequently performs an order of magnitude analysis to determine the critical scale required for K_{z2} to effectively influence the dispersion dynamics, and proposes alternative formulations based on physical turbulence parameters. The emission is characterised by a continuous rate, Q, from a point source located at coordinates (x_s, z_s). The turbulence structure within the PBL is described by fundamental scaling parameters, including the PBL height, h; the friction velocity, u_*; the Monin-Obukhov length, L; and the surface roughness length, z_0. Furthermore, κ represents the von Kármán constant. Finally, the vertical profiles for the mean wind speed, $U(z)$, and for the turbulent diffusivity, denoted in this study as $K_{z1}(z)$, are parameterised in terms of these PBL variables, with their specific formulations being detailed subsequently.

$$\begin{aligned} U(z) = \frac{u_*}{\kappa}\Bigg[\ln\left(\frac{z}{z_0}\right) + \ln\left(\frac{(1+\mu_0^2)(1+\mu_0)^2}{(1+\mu^2)(1+\mu)^2}\right) + 2(\arctan(\mu) - \arctan(\mu_0)) \\ + \frac{2L}{33h}(\mu^3 - \mu_0^3)\Bigg] \end{aligned} \tag{2}$$

with, $\mu = \left(1 - 22\frac{z}{L}\right)^{1/4}$ and $\mu_0 = \left(1 - 22\frac{z_0}{L}\right)^{1/4}$.

Table 1: Micrometeorological and Dispersion Data from the Copenhagen Experiment. Adapted from [17].

Run	**Experiment Date**	L **(m)**	u_* **(m s^{-1})**	h **(m)**	x **(m)**	C/Q **(10^{-4} s m^{-2})**	Q **(g s^{-1})**
1	Sep 20	-46	0.37	1980	1900	6.28	3.2
					3700	2.31	
2	Sep 26	-384	0.74	1920	2100	5.38	3.2
					4200	2.95	
3	Oct 19	-108	0.39	1120	1900	8.20	3.2
					3700	6.22	
					5400	4.30	
4	Nov 03	-173	0.39	390	4000	11.66	2.3
5	Nov 09	-577	0.46	820	2100	6.72	3.2
					4200	5.84	
					5100	4.97	
6	Apr 30	-569	1.07	1300	2000	3.96	3.1
					4200	2.22	
					5900	1.83	
7	Jun 27	-136	0.65	1850	2000	6.70	2.4
					4100	3.25	
					5300	2.23	
8	Jul 06	-72	0.70	810	1900	4.16	3.0
					3600	2.02	
					5300	1.52	
9	Jul 19	-382	0.77	2090	2100	4.58	3.3
					4200	3.11	
					6000	2.59	

$$K_{z1}(z) = \kappa u_* h \frac{z}{h}\left(1-\frac{z}{h}\right)\left(1-22\frac{z}{L}\right)^{1/4} \tag{3}$$

The formulations for the vertical turbulent diffusivity and wind speed profil, Eq. (3) and Eq. (2) were proposed by Ulke [7].

The governing equation is solved numerically via an implicit finite difference scheme. The domain is discretised into a grid with steps Δx and Δz in the downwind and vertical directions, respectively. The vertical derivative terms are discretised using second-order accurate central difference approximations. Specifically, the second and fourth-order derivatives at a grid point z_i are approximated by the formulas:

$$\left.\frac{\partial^2 C}{\partial z^2}\right|_{z_i} \approx \frac{C_{i+1} - 2C_i + C_{i-1}}{(\Delta z)^2}$$

$$\left.\frac{\partial^4 C}{\partial z^4}\right|_{z_i} \approx \frac{C_{i+2} - 4C_{i+1} + 6C_i - 4C_{i-1} + C_{i-2}}{(\Delta z)^4}$$

The use of the five-point stencil for the fourth derivative transforms the partial differential equation into a system of linear algebraic equations at each downwind step, x_j. The resulting coefficient matrix for the unknown concentration profile at x_{j+1} is penta-diagonal, which results from the bi-flux term. This system is solved using a specialised algorithm for banded matrices, specifically the `solve_banded` function from the SciPy library in Python, which is optimised for such structures [18]. To consider impermeability at the boundaries for both transport mechanisms, conditions of zero Fickian diffusive flux ($\frac{\partial C}{\partial z} = 0$) and zero retention flux ($\frac{\partial^3 C}{\partial z^3} = 0$) are imposed at the ground ($z = 0$) and the top of the PBL ($z = h$).

2.3 Physical Parameterization of the Fourth-Order Coefficient

Unlike the second-order eddy diffusivity K_{z1}, which has been extensively studied and possesses well-established vertical profiles Ulke [7], the fourth-order coefficient K_{z2} lacks a universal parameterization in the literature. A constant value for K_{z2} is physically inconsistent for atmospheric applications, as it fails to account for the temporal and spatial variability of turbulent scales.

To derive a physically consistent formulation, a dimensional analysis combined with an order-of-magnitude assessment of the governing equation was performed. As will be detailed in the Results section, comparing the magnitude of the classical diffusive term with the bi-flux retention term revealed the required scaling for K_{z2} to effectively influence the dispersion dynamics. In the context of the MOST, turbulence within the PBL is governed by velocity and length scales. The friction velocity, u_* ($[LT^{-1}]$), represents the shear-driven turbulent intensity, while the Monin-Obukhov length, L ($[L]$), characterizes the stability-driven stratification limits.

Combining these scales to satisfy the dimensional requirement yields the following closure hypothesis:

$$K_{z2} \propto u_* \cdot |L|^3 \tag{4}$$

This parameterization implies that the "memory" or "retention" of the plume is proportional to the mechanical energy available (u_*) and the cubic volume of the stability eddies ($|L|^3$). In unstable conditions (negative L), this term dynamically adjusts the non-Fickian contribution, growing larger as conditions shift toward near-neutrality (large $|L|$). However, it is crucial to note a theoretical limitation of this approach: as the atmospheric boundary layer approaches strictly neutral conditions, $|L| \to \infty$. In such a scenario, K_{z2} would increase excessively, causing the fourth-order term to unphysically dominate the transport equation. Therefore, this specific formulation assumes that the boundary layer remains in an unstable regime where $|L|$ is bounded.

This proposed formulation will be evaluated in the Results section to verify its capability to minimize the discrepancy between Fickian predictions and experimental observations.

3 Results

3.1 Numerical Validation against Reference Solution

Before applying the height-dependent profiles proposed by Ulke[7], a preliminary validation of the numerical scheme was conducted. The objective of this step was to verify the accuracy of the finite difference implementation by reproducing the results reported by Palmeira et al. [5].

Since the reference study assumes a simplified scenario with constant wind speed and constant vertical eddy diffusivity, the present model was first adapted to match these conditions for comparison purposes. Specifically, for this validation case, the profiles $U(z)$ and $K_{z1}(z)$ were replaced by mean values averaged over the boundary layer height, as defined in Palmeira et al. [5].

By adopting these constant parameters and setting $K_{z2} = 1$, the model results were compared against the reference data. Figure 1 presents the scatter diagram comparing the concentrations predicted by the proposed numerical model against the analytical solution. A visual inspection reveals a precise superposition between the numerical results, blue circles, and the reference analytical values, red crosses). This near-perfect alignment demonstrates that the finite difference discretization, particularly regarding the fourth-order derivative term, introduces negligible numerical errors.

This quantitative agreement is further corroborated by the data in Table 2. The relative difference between the numerical and analytical solutions is consistently below 0.2% across all examined arcs. The maximum deviation observed was approximately 0.16% (Case 9), with the vast majority of points exhibiting differences in the range of 0.02% to 0.07%. Such marginal discrepancies are attributable to truncation errors inherent to the grid resolution and confirm the robustness of the implemented solver. Consequently, the numerical model is deemed verified and suitable for the subsequent application of height-dependent profiles and the investigation of the K_{z2} parameter.

3.2 Order of Magnitude Analysis of the Retention Term

To investigate the effective contribution of the fourth-order term, a scale analysis was performed to compare the magnitude of the Fickian diffusion term against the bi-flux retention term throughout the dispersion domain. Dimensional groups were defined based on the characteristic scales of the PBL (height $H \approx 2000$ m) and the turbulence intensity. The dimensionless magnitude of the diffusive term (Π_1) and the retention term (Π_2) are defined as:

$$\Pi_1 \sim \frac{\beta K_{z1} L}{U H^2} \quad \text{and} \quad \Pi_2 \sim \frac{\beta(1-\beta) K_{z2} L}{U H^4} \tag{5}$$

where L represents the downwind distance scale. The ratio between these two mechanisms, defined as $\Psi = \Pi_2/\Pi_1$, indicates the relative weight of the retention phenomenon in the transport equation.

The results reveal a stark discrepancy between the transport mechanisms. As shown in Fig. 2, the magnitude of the retention term is approximately 10 orders of magnitude smaller than the diffusive term ($\Psi \approx 10^{-10}$). This

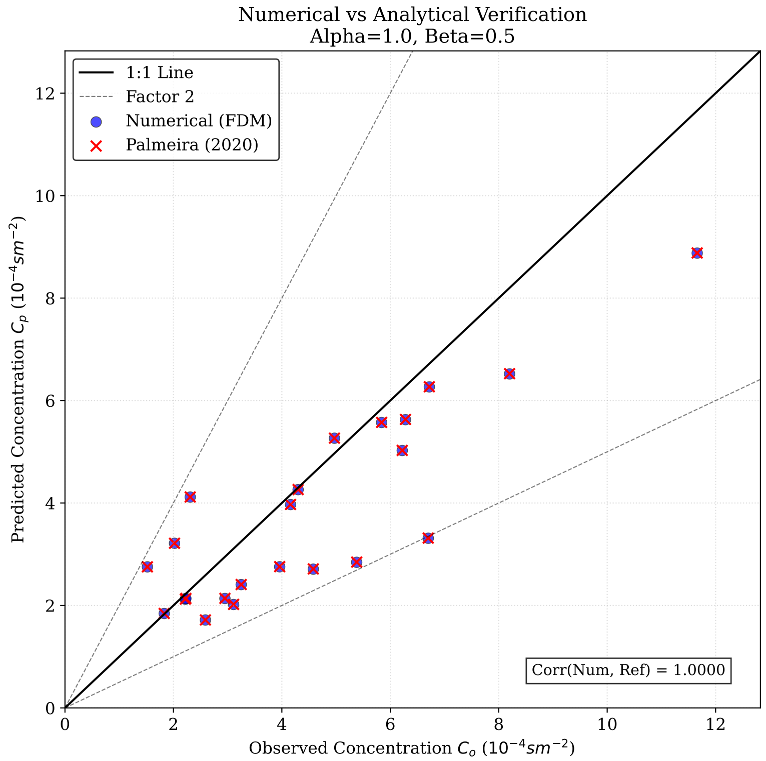

Figure 1: Scatter plot comparing Numerical (FDM) results against the Analytical solution.

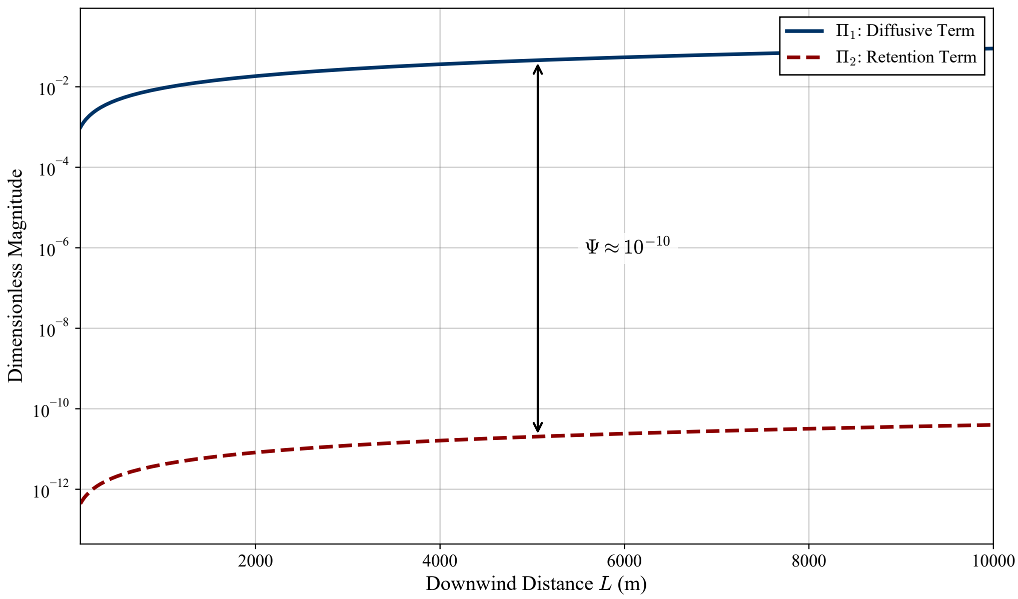

Figure 2: Scale analysis comparing the dimensionless magnitude of the Fickian diffusion term (Π_1) and the Retention term (Π_2) for $K_{z2} = 1$.

behavior is physically explained by the geometric scaling of the PBL: the fourth-order derivative implies a scaling factor of H^{-4}, which drastically penalizes the magnitude of the term when H is large (on the order of 10^3 m).

It is crucial to emphasize that the original bi-flux equation was initially formulated to model anomalous molecular diffusion, where characteristic length scales are microscopic. In the present context, however, the model is applied to turbulent diffusion in the atmosphere, governed by macroscopic spatial scales. This fundamental shift from molecular to turbulent transport phenomena is the primary reason for this massive discrepancy in the order of

Table 2: Pointwise comparison between the numerical solution (FDM) and the analytical solution for the Copenhagen experiment arcs ($\alpha = 1.0, \beta = 0.5$).

Case ID	x (m)	Observed (C_o/Q)	Numerical (C_p/Q)	Analytical (C_a/Q)	Diff. (%)
1	1900	6.28	5.627	5.631	0.07
	3700	2.31	4.116	4.118	0.04
2	2100	5.38	2.843	2.847	0.15
	4200	2.95	2.136	2.138	0.08
3	1900	8.20	6.520	6.525	0.07
	3700	6.22	5.025	5.027	0.04
	5400	4.30	4.261	4.262	0.03
4	4000	11.66	8.878	8.881	0.03
5	2100	6.72	6.267	6.269	0.04
	4200	5.84	5.572	5.573	0.03
	5100	4.97	5.265	5.266	0.03
6	2000	3.96	2.755	2.758	0.10
	4200	2.22	2.122	2.123	0.06
	5900	1.83	1.843	1.844	0.04
7	2000	6.70	3.312	3.315	0.10
	4100	3.25	2.407	2.409	0.05
	5300	2.23	2.136	2.137	0.04
8	1900	4.16	3.970	3.971	0.04
	3600	2.02	3.213	3.214	0.03
	5300	1.52	2.753	2.754	0.02
9	2100	4.58	2.709	2.713	0.16
	4200	3.11	2.019	2.021	0.08
	6000	2.59	1.716	1.717	0.06

magnitude.

Therefore, it is concluded that adopting a constant unitary value for K_{z2} renders the bi-flux term mathematically negligible, effectively reducing the model to a classical Fickian formulation. For the bi-flux theory to be physically relevant in atmospheric dispersion problems, K_{z2} must be parameterized to scale with the turbulent properties of the PBL, specifically compensating for the order of H^2 relative to the eddy diffusivity K_{z1}. Preliminary estimates based on Eq. (5) suggest that, for typical PBL heights ($H \approx 2000$ m), the coefficient K_{z2} requires values in the order of 10^5 to 10^8 m^4 s^{-1} to yield a non-negligible Ψ, a magnitude that physically represents the scale of fourth-order turbulent correlations.

3.3 Sensitivity Analysis and Parameter Optimization

Following the mathematical verification, the physical performance of the model was evaluated using the Copenhagen Experiment dataset. In this stage, the longitudinal wind speed and vertical eddy diffusivity were modeled using Eq. (2) and Eq. (3), respectively. A parametric sweep was conducted varying the retention coefficient K_{z2} from 10^0 to 10^9 m^4 s^{-1} and the partition parameter β from 0.1 to 1.0.

The optimization landscape is presented in Fig. 3, which illustrates the NMSE behavior across the parameter space. For clarity, this figure focuses on the transition region (10^4 to 10^8) where the model sensitivity is most pronounced. The analysis reveals two distinct regions of optimal performance, highlighting a trade-off between error minimization and predictive robustness. Specifically, a Scenario of Minimum Error is observed at $\beta = 0.95$ and $K_{z2} = 10^5$, yielding the lowest global error, whereas a Scenario of Maximum Robustness is achieved at $\beta = 0.99$ and $K_{z2} = 10^7$, providing the best correlation and reliability metrics.

A detailed inspection of the statistical metrics, summarized in Table 3, clarifies the dual impact of the bi-flux approach. Comparing the Minimum Error Scenario ($\beta = 0.95$) against the classical Fickian baseline ($\beta = 1.0$), it is observed that both models perform exceptionally well regarding mass balance conservation. The classical model already exhibits a negligible Fractional Bias (FB ≈ 0.006); however, the introduction of the fourth-order term further refined this metric to an even smaller value (4.30×10^{-4}). While both values indicate practically zero systematic bias,

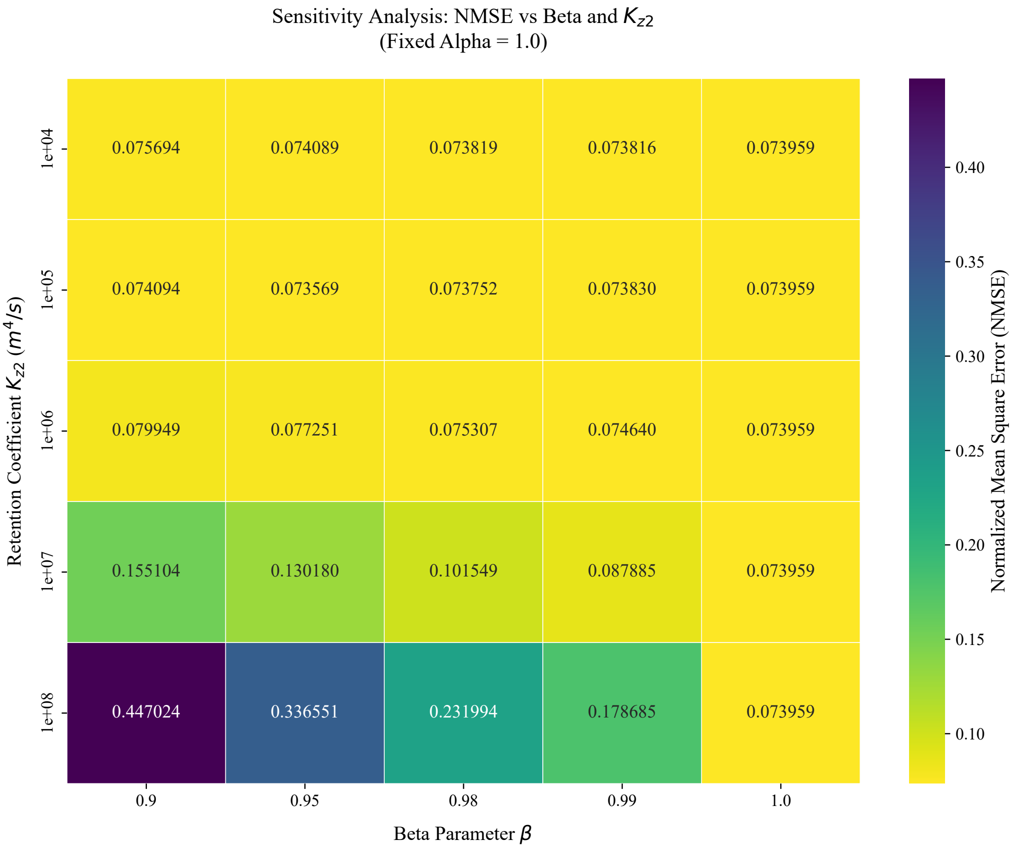

Figure 3: Heatmap of NMSE performance evaluating the interplay between β and K_{z2}.

Table 3: Comparison of statistical indices between the Fickian model and the optimized Bi-flux configurations.

Metric	$\beta = 1.0$ (Fickian)	$\beta = 0.95$ ($K_{z2} = 10^5$)	$\beta = 0.99$ ($K_{z2} = 10^7$)	**Ideal**
NMSE	0.073959	0.073569	0.0879	0.0
COR	0.862427	0.862273	0.8690	1.0
FAT2	0.956522	0.956522	1.000	1.0
FB	0.006680	0.000430	0.0841	0.0
FS	0.206149	0.208033	0.3002	0.0

this subtle refinement contributed to achieving the lowest overall NMSE, all while keeping the Correlation (COR) and Factor of 2 (FAT2) indices stable.

Conversely, the Maximum Robustness Scenario is achieved with a higher retention magnitude ($K_{z2} = 10^7$) and a lighter weighting ($\beta = 0.99$). Despite a marginal increase in the mean squared error, this configuration excelled in predictive safety. Notably, it was the only case to achieve a FAT2 of 1.000, ensuring that 100% of the estimated concentrations fall within a factor of two of the observed data. Furthermore, this scenario yielded the highest correlation coefficient (COR = 0.8690), indicating an improved capability to capture the trend of the experimental data compared to the baseline model.

3.4 Evaluation of the Physically Parameterized Bi-flux Model

The sensitivity analysis conducted in the previous section demonstrated that the primary issue with the retention parameter K_{z2} lies in its order of magnitude when applied to atmospheric turbulent dispersion, rather than the mere use of constant values. The parametric sweep was essential primarily to provide a baseline notion of the required order of magnitude for this parameter. It is highly undesirable to treat K_{z2} as a free mathematical hyperparameter in the dispersion problem; instead, its definition must be fundamentally grounded in the physics of the PBL. Consequently, the physical parameterization was proposed to ensure that the dynamically calculated parameter yields the specific magnitude that best described the results in the preceding section.

The proposed formulation, $K_{z2} = u_* |L|^3$, scales the retention term with the friction velocity (u_*) and the Monin-Obukhov length (L). This approach ensures that the non-Fickian memory effects are dynamically adjusted based on the mechanical turbulence intensity and the thermal stability of the atmosphere. To determine the optimal contribution of the fourth-order term, the model was evaluated for a range of the partition parameter β varying from 0.85 to 1.0. The best overall performance, characterized by the minimization of the global error and maximization of the correlation, was found at $\beta = 0.99$. This specific configuration, which balances the Fickian diffusion with the retention mechanism, was then compared against the classical Fickian solution ($\beta = 1.0$).

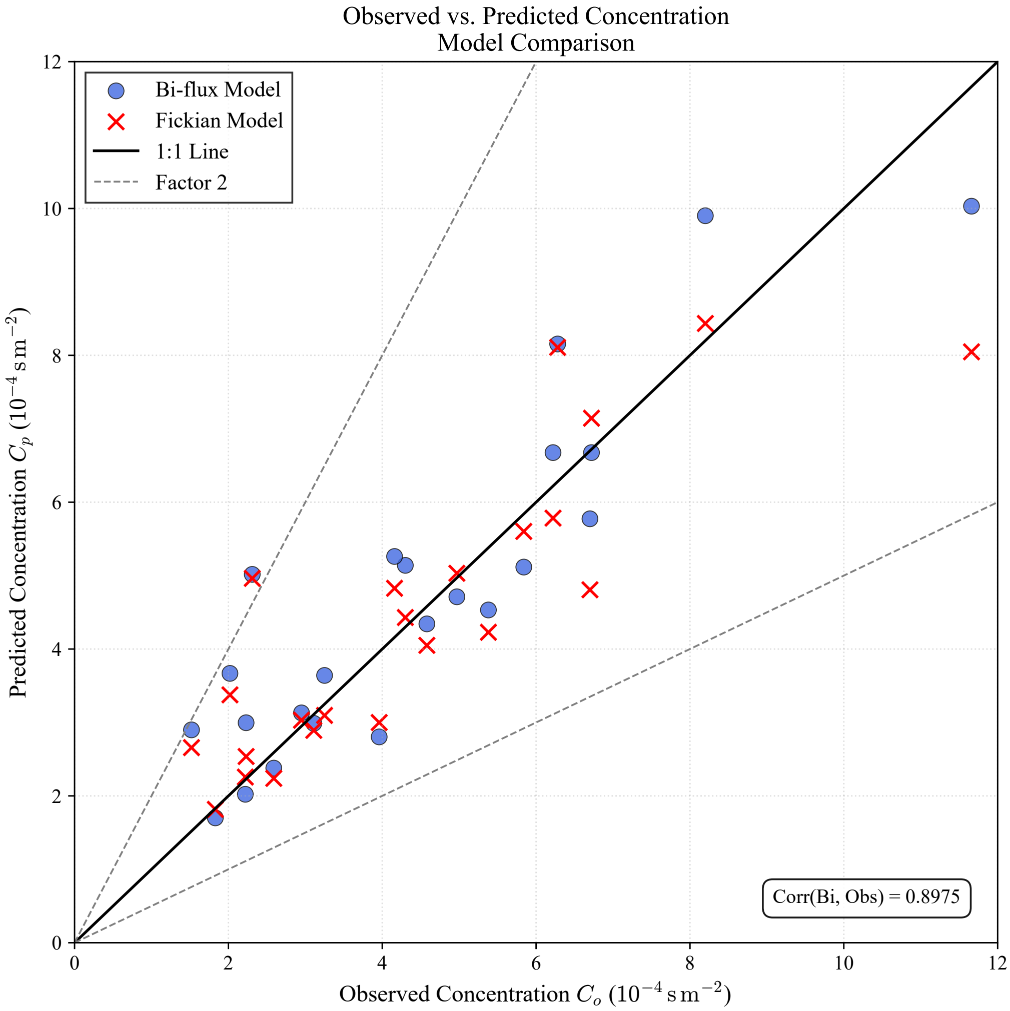

Figure 4: Scatter plot comparison of observed vs. predicted concentrations for the Fickian model ($\beta = 1.0$) and the Parameterized Bi-flux model ($\beta = 0.99, K_{z2} = u_* |L|^3$).

Figure 4 presents the scatter diagram of observed versus predicted concentrations for both models. While scatter plots offer a visual representation of the data distribution, visually diagnosing specific overestimation or underestimation tendencies can be imprecise and potentially misleading. A rigorous assessment of whether the model underpredicts peak concentrations or excessively smooths the plume must be grounded in objective statistical metrics rather than visual inspection, strictly requiring the analysis of the FS and FB.

The quantitative impact of this parameterization is summarized in Table 4. The introduction of the atmospheric scaling for K_{z2} resulted in a substantial improvement in global performance metrics.

The NMSE decreased from 0.0740 to 0.0562, representing a net accuracy gain of 24.1%. Furthermore, the Correlation Coefficient increased from 0.8622 to 0.8975 (an improvement of 4.1%), indicating that the parameterized Bi-flux model is more effective at reproducing the trend of the experimental data across different downwind distances.

Table 4: Statistical performance comparison between the Classical Fickian model and the Parameterized Bi-flux model using Copenhagen experimental data.

Statistical Metric	**Fickian Model**	**Bi-flux Model**	**Improvement**
	($\beta = 1.0$)	($\beta = 0.99, K_{z2} \propto u_*\|L\|^3$)	
NMSE	0.0740	0.0562	**24.1%**
COR	0.8622	0.8975	**4.1%**
FAT2	0.957	0.957	-
FB	0.0063	-0.0617	-
FS	0.4067	0.1202	**70.4%**

However, the most physically significant result is effectively verified in the FS. The Fickian model yielded a relatively high FS value (0.4067), which objectively implies a tendency to under-predict the internal variability of the concentration field. This is consistent with the theoretical limitation of second-order diffusion, which assumes instantaneous smoothing of gradients. In contrast, the Bi-flux model achieved an FS of 0.1202, achieving a remarkable 70.4% improvement. This reduction confirms that the fourth-order term introduces a physical "retention" or "inertia" to the dispersion process. By limiting the artificial expansion of the plume, the model successfully preserves the internal structure of the concentration field, allowing for a realistic representation of plume fluctuations.

Regarding the Factor of Two (FAT2), both models performed exceptionally well (0.957), confirming the absolute robustness of the numerical solver. As for the mean bias, the FB shifted slightly from a near-zero positive value (0.0063) to a small negative value (-0.0617). This statistical evaluation of the FB demonstrates a slight conservative tendency (overestimation) in the Bi-flux model—contradicting any visual assumptions of general underestimation—yet it remains well within the highly acceptable range for air quality models ($|FB| < 0.3$).

4 Conclusion

This study investigated the application of the bi-flux theory to atmospheric dispersion modeling, specifically evaluating the impact of the fourth-order diffusion term on the simulation of pollutant transport in the planetary boundary layer. By implementing a high-order finite difference scheme and validating it against analytical solutions and the Copenhagen experiment dataset, several key conclusions can be drawn regarding the physical and numerical behavior of non-Fickian dispersion.

First, the numerical verification confirmed that the proposed fourth-order solver is robust and accurate, reproducing analytical solutions with a relative error below 0.2% for constant coefficients. However, the order of magnitude analysis revealed that simply adopting a unitary constant for the retention parameter K_{z2} renders the bi-flux term physically negligible due to the geometric scaling of the PBL height (H^{-4}). This finding highlights the necessity of a physical parameterization for the fourth-order coefficient to ensure its relevance in atmospheric scales.

The sensitivity analysis demonstrated that the model performance is highly dependent on the interplay between the partition parameter β and the magnitude of K_{z2}. The optimization landscape revealed a trade-off between error minimization and predictive robustness, with distinct regions of optimal performance.

The most significant contribution of this work lies in the successful implementation of an atmospheric-dependent parameterization for the retention term, defined as $K_{z2} = u_*|L|^3$. This formulation dynamically links the non-Fickian memory effects to the local turbulence scales, specifically, the friction velocity and the thermal stability. When evaluated with the optimal partition parameter $\beta = 0.99$, this physically grounded approach yielded superior results compared to the classical Fickian model ($\beta = 1.0$).

Quantitatively, the parameterized bi-flux model achieved a 24.1% reduction in the NMSE and a 4.1% increase in the COR. More importantly, the FS was reduced by 70.4%, dropping from 0.407 to 0.120. This improvement in FS indicates that the fourth-order term effectively mitigates the excessive smoothing characteristic of second-order diffusion, preserving the internal variance and structure of the plume.

These results suggest that while atmospheric transport is predominantly governed by Fickian diffusion, the inclusion of a physically scaled fourth-order term acts as a crucial corrective mechanism for the turbulent energy dissipation scales. The bi-flux formulation, therefore, offers a more realistic representation of plume fluctuations in the unstable boundary layer, proving to be a valuable refinement for high-fidelity air quality modeling.

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