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Particle dispersion in atmospheric modelling: A comprehensive review

Bohuai Xiao[1,3], Chengjin Tang[2]

[1]School of YKPao, Shanghai, 201600, China
[2]School of Cranbrook, Bloomfield Hills, 48304, USA

[3]s15068@ykpaoschool.cn

Abstract. Atmospheric dispersion modeling, traditionally inclined towards gaseous dispersion, has undergone significant evolution in capturing the intricacies of particle dispersion in urban and open environments. This comprehensive review explores the nuances of particle-gas interactions, highlighting discrepancies in correlations between their concentrations, influenced by factors such as turbulence and multiple emission sources. The research accentuates the intriguing dynamics between PM2.5 and PM10 concentrations, suggesting the viability of models based on passive scalars for such particles in open environments. However, a marked challenge emerges in modeling particle number concentration, necessitating the integration of aerosol dynamics modules. Emphasizing the diversity of model types, this paper elucidates the specific requirements across varying spatial scales, identifying gaps in understanding particle dispersion and aerosol dynamics. The review critically assesses the performance of notable models, highlighting the paramount importance of quality data sources and underscoring the need for more dedicated focus on particle dynamics beyond mass predictions. Through a synthesis of existing literature and model evaluations, this review seeks to guide future research endeavors, fostering advancements in atmospheric dispersion modeling.

Keywords: Atmospheric Dispersion Modelling, Gaussian Plume, Langrangian, Computational Fluid Dynamics.

1. Introduction

The intricate realm of dispersion modeling navigates through complex mathematical equations that encapsulate the atmospheric dynamics, processes occurring within the plume, and the dispersion of particles and gases [1]. Historically, the bulk of attention has been dedicated to modeling gaseous dispersion with substantial reviews being committed to street canyon dispersion and comparisons across distinct models leveraging test meteorological data. However, studies diving deep into the juxtaposition of particle concentrations with gases are limited in number [2, 3].

Research in open environments has often portrayed an inconsistency in correlations between concentrations of gases and particles. For instance, a paper unveiled a weak correlation between outdoor PM10 and NO2 concentrations within urban confines, a contrast to findings by another who found a stronger association between SF6 and PM10[4, 5]. Interestingly, in most of these studies, wind flow emanated perpendicularly from the road, ensuring minimal interference. Yet, notable disparities have been observed in how gases and particles disperse locally, especially in complex settings influenced heavily by turbulence and multiple emission sources [6, 7].

While urban terrains predominantly see traffic emissions as a major particle source, correlations between PM2.5 and PM10 concentrations are intriguing. A paper delineated an R2 value of 0.95 for these particles, although the PM10/PM2.5 ratio showcased significant variations [8]. This, in combination with other nuanced findings from other papers and subsequent studies, suggests that models fixated on passive scalars such as inert gases might efficiently gauge PM2.5 and PM10 concentrations in open environments. Here, transient events leading to short-term variations get averaged out over extended periods.

This review aims to illuminate the diversity of model types, delve into specificities required for varied spatial scales ranging from local to regional, and pinpoint deficiencies related to particle dispersion and aerosol dynamics across these scales [6, 9]. While an exhaustive appraisal of all models remains out of scope, this review will encapsulate the most pertinent models, emphasizing crucial parameters and inputs as detailed in Tables 1 and 2. Furthermore, the ability of many models to claim competence in modeling particle dispersion is scrutinized. Without a dedicated focus on particle dynamics, results often remain confined to particle mass predictions, primarily PM2.5 and PM10, neglecting particle number concentration. Validations for many such models are conspicuously absent, but where possible, this review will highlight their performance in terms of gas dispersion validation, leveraging insights from multiple studies that portray substantial correlations between non-reactive gases and particles in broader airsheds [10-12].

Table 1. Basic parameters for models not containing aerosol dynamics modules [13].

Name	Model Type [a]	Scale [b]	Grid size	Resolution	Source Types [c]	Pollutants [d]	Output Frequency	Atmospheric Stability [e]	Turbulence [f]
AURORA	B	L	1 x 1 km	NA	L	CO, NO_2, SO_2, PM_{10}	1h, 24h, 1yr	NA	Limited AMB
CPB	B	L		NA	L	NO_2, and inert gases		NA	NA
CALINE4	GP	L	H:100-500 m	1m	L	CO, NO_2, TSP	1h, 8h	P	VIT, AMB
HIWAY2	GP	L	10-100 m but up to 10km depending on scaling factor	1m	L	Non-reactive gases	1h	P	AMB
CAR-FMI	GP	L	Up to 10 km	H: adjustable V: not defined	L	CO, NO, NO_2, NO_x, $PM_{2.5}$	1h, 8h, 24h, 1yr	BL	VIT, AMB
AEROPOL	GP	L	H: Up to 100 km V: Up to 2km	H: 10-1000 m V: 100 m	P, V	G, P	1h	P	AMB
ADMS	3D quasi GP	L, R	3000 grid cells up to 50 km	H: no limits V: no limits	P,A,L	G, P	10 mins to 1yr	BL	VIT
GRAL	L	L	100 m-20km	H: no limits V: no limits	P, L	G, P	10 min to 1h	BL	Local (*k-L* model) VIT3DWF
GATOR	E	L, R, G	Up to Global	Depends on scale of area	P, L, A, V	G, P	1h- 1yr	BL	AMB
OSPM	GP/Box	L	NA	NA	L	NO_x, NO_2, O_3, CO PM	1h	NA	VIT, Empirical Wind Turbulence
STAR-CD	CFD	L	<1km	H: <1m+ V: <1m+	P, L, A, V	G, P	1 min	BL	VIT
ARIA	CFD	L	Depends on scaling factor	H: <1m+ V: <1m+	P, L, A, V	G, P	Real time	P	VIT, Local (*k-L* model) VIT3DWF

Table 1. (continued)

PBM	Box	R	H: <50km V: variable <2km	NA	P, L,A	G		NA	NA
CALPUFF	Multi-layer non-steady GPuff	R	<200km	H: no limits V: no limits	P, L, A, V	G, P	>1h	BL	AMB
SCREEN3	GP	R	<50km	H: no limits V: no limits	P,A,V	G, P	1h in simple terrain, <24h in complex	T	Y
TAPM	E/L	R	<1000 x 1000km	H: 0.3 - 30km V: >10m	P,A,V	G, P	1h, 8h, 1yr	BL	*k-ε*
AERMOD	Bi Gaussian steady state GP	L, R	<50km	H: no limits V: no limits	P,A,V	G, P	1h, 24h, 1yr	BL	AMB
SPRAY	L	L, R	<1-100km	H: 1m – 4km V: 1m – 4km	P,L,V	G, P	1 min +	BL	
MISCAM	CFD	L	<300m	H:1m(60cells) V: 1m(20cells)	P,L,V	G, P	1 min +	BL	AMB
MICRO-CALGRID	CFD	L	<10km	H: 1 m V: 1 m	P,L,V	G, P	1 min +	BL	VIT, AMB

Note: NA= Not applicable.
[a] Model Types: B = Box, GP = Gaussian Plume, L = Lagrangian, E = Eulerian, CFD = Computational Fluid Dynamics, GPuff = Gaussian Puff.
[b] Scale: L= Local, R = Regional.
[c] Source Types: L = Line, P = Point, A = Area, V = Volume.
[d] Pollutants: G Gases, P = Particles.
[e] Atmospheric Stability: P = Pasquill, BL = Boundary Layer Scaling, T = Turner.
[f] Tubulence: VIT = Vehicle Induced Turbulence, AMB = Turbulence of Ambient Air, VIT3DWF = Vertical Inhomogeneous Turbulence and Inhomogeneous 3D Wind Field.

Table 2. Processes included in the dispersion models not containing an Aerosol Dynamics package [13].

Name	Street Canyon	Building Wake Effects [a]	Topography	Intersections	Plume Rise	Chemistry
AURORA	O	X	Simple	X	X	X
CPB	O	O	Simple	X	X	X
CALINE4	X	X	Simple	O	X	DPM
HIWAY2	X	X	Simple	X	X	X
CAR-FMI	X	X	Simple	X	X	DPM
AEROPOL	X	X	Simple	X	O	O
ADMS	O	O	Complex	O	O	O
GRAL		X	Complex	X	O	X
GATOR	X	X	Simple	X	X	O
OSPM	O	O	Simple	X		O (NO – NO_2 – O_3 chemistry)
ARIA	O	O	Complex	O	O	O
PBM	X	X	X	X	X	O
CALPUFF	X	S-S H-S	Complex	X	X	X
SCREEN3	O	S-S H-S	Simple and Complex	X	X	X

Table 2. (continued)

TAPM	X	S-S H-S	Complex	X	O (Simplified)	O GRS
AERMOD	X	Evaluation version	Simple and Complex	X	X	O
MISCAM	O	O	Simple	X	X	X
MICRO-CALGRID	O	O	Simple and Complex	X	O	O

Note: X Not included, O included
[a] Building Wake Effects: S-S = Schulman-Scire, H-S = Huber-Snyder

2. Overview of Models for Dispersion Within a Street Environment

Air pollution within urban settings, particularly dense street canyons, presents unique challenges. The microclimate, architectural variances, and myriad of emission sources in such environments necessitate specialized modeling techniquestable [6, 9, 14]. It has been highlighted that there is this need through a detailed examination of urban dispersion models[2, 6]. Some findings underscored the vitality of specific models tailored for intricate urban dynamics. Here, the paper will discuss the particular models that does not explicitly focus on aerosol dynamics, but provide valuable insights into the dispersion of pollutants within urban confines.

2.1. Box Models

At the heart of box modeling lies the visualization of an urban segment, typically a street canyon, as a confined ‘box’. Within this construct, pollutant concentrations and dynamics are evaluated to render a comprehensive picture of air quality. The AURORA model, a brainchild of VITO in Belgium, stands as a cornerstone in this realm. With its integrated urban framework, AURORA has the capability to predict both gaseous and particulate concentrations [14]. This model, while offering an overarching view of pollutant state within street canyons, may occasionally miss the intricate real-time dispersion nuances [14]. On the other hand, the CPB model by GEOMET takes a specialized approach [15]. Designed with a keen eye on urban canyons that adhere to certain height-to-width ratios, it is adept at calculating average concentrations of inert gases, showcasing its prowess in such tailored environments [15]. However, as urban terrains become more complex and diverse, the model might find its precision wavering.

2.2. Gaussian Models

Building upon the foundational Gaussian plume concept, Gaussian models offer an amalgamation of user-friendliness with operational efficiency. Models like CALINE4, stemming from the efforts of the California Department of Transportation, and HIWAY2, an initiative of the US EPA, stand as testimonies to this approach [16, 17]. While both models shine in their simplicity, CALINE4 goes a step further [18]. By incorporating elements of both thermal and vehicle-induced turbulence, its predictions in urban settings gain a layer of accuracy missing in other similar models [19, 20]. Progressing further, the CAR-FMI model, a creation of the Finnish Meteorological Institute, endeavors to perfect the Gaussian approach. By integrating turbulence data from a spectrum of sources, CAR-FMI often astounds with its precision, especially in specific urban settings [21]. Yet, like all models, it’s not without its Achilles’ heel, with low wind scenarios presenting a challenge.

2.3. Lagrangian (and Eulerian) Models

Traversing a different trajectory, Lagrangian models present a nuanced perspective on urban pollution dispersion. One cannot discuss this category without mentioning the GRAL model, a product of the rigorous research at the Institute for Internal Combustion Engines and Thermodynamics in Graz, Austria. Tailored meticulously for understanding the dispersion of inert compounds amidst fluctuating wind terrains, GRAL's predictions often resonate with on-ground realities, making it an invaluable asset for researchers [3, 22, 23].

2.4. Computational Fluid Dynamic Models

When it comes to precision, adaptability, and depth, computational fluid dynamic models often steal the limelight. The ARIA Local model exemplifies this category's prowess with its unmatched real-time dispersion analytics. Its malleability, in accommodating a wide array of variables, renders it indispensable for rapid-response scenarios [24, 25]. Parallelly, the MISKAM model, with its laser-focus on densely constructed urban zones, often surfaces in academic circles for its uncanny accuracy in such settings. Taking inspiration from MISKAM, the MICRO-CALGRID model broadens the horizon further [26]. Its holistic approach, encompassing everything from traffic-induced emissions to complex chemical reactions, encapsulates the multifaceted reality of urban pollution dispersion.

3. Overview of urban and regional scale dispersion models

3.1. Box Models

Box models are a simple yet effective means to represent atmospheric processes on a basic level. They essentially consist of a volume (the "box") within which various atmospheric constituents and processes are homogenized, meaning the contents are well-mixed and uniform across the box [13, 27]. This section will provide a detailed exploration of the Photochemical Box Model, a representative of this type of modeling. For example, the US EPA developed PBM. The PBM expands upon the rudimentary nature of box models to simulate photochemical smog specifically at an urban scale. Although it upholds the fundamental box model concept of having a fixed horizontal area (usually spanning 10-50km), the PBM diverges from other box models in that its vertical boundary is variable [27]. This flexibility in boundary height, which oscillates between 0.1 and 2km, resonates with observed diurnal variations [28]. Owing to its design and the processes it can simulate, the PBM is adept at handling situations characterized by low wind speeds and fluctuating conditions, especially when sunlight is a key player [27]. Representations of the urban environment can be crafted via single cells or multiple interconnected cells. These cells are tasked with tracking hourly variations in specific pollutants, namely hydrocarbons and ozone [27, 28]. Though there are a few assumptions made by this model. Whether they stem from point, line, or area sources, emissions are assumed to be uniformly spread across the box's surface [27]. Also, the air and the constituents within the box are believed to be well-mixed, ensuring a consistent concentration of pollutants throughout the box [28].

3.2. Gaussian Models

Atmospheric dispersion models play a pivotal role in decoding the intricacies of pollutant movement within our atmosphere. As modern industries and cities have expanded, so too has the need to comprehend and predict the trajectories of various contaminants in the air [29]. Over the decades, this understanding has culminated in the development of several sophisticated models, each honed to cater to specific scenarios and nuances [1, 29]. The AEROPOL model, for instance, stands out as a primarily steady-state dispersion tool [6]. It's meticulously crafted for environments within a 100km radius of a pollution source, particularly when the topography leans towards being flat [2, 29]. A salient feature of AEROPOL is its dexterity in factoring in obstructions, such as towering buildings, which can profoundly influence localized air currents and resultant dispersion patterns [29]. However, no model is without its quirks. AEROPOL's Achilles' heel is its steadfast adherence to a neutral atmospheric stability assumption [2, 6]. This means that in dynamic atmospheres, where stability varies, the model might present a skewed picture, potentially glossing over intricate details [29].

Pivoting from AEROPOL's approach, CALPUFF introduces a more dynamic perspective [30]. Functioning as a non-steady-state puff dispersion model, CALPUFF doesn't just see pollution as a continuous stream; instead, it visualizes them as discrete 'puffs' or 'clouds', meandering and evolving with time [2, 30]. This characteristic grants CALPUFF a unique vantage point, enabling it to capture the nuances of an ever-shifting meteorological canvas [6, 30]. From towering industrial chimneys to sprawling cityscapes, CALPUFF's versatility is evident in its capability to model emissions from an array of sources—be they point, line, area, or volume. However, with great detail comes the challenge of complexity [1, 6, 30]. In densely populated urban areas, where air currents twist and turn unpredictably due to myriad factors, CALPUFF might find itself ensnared in the web of intricate dispersion pathways, slightly muddling its predictions [30].

A collaboration between stalwarts, the American Meteorological Society (AMS) and the US Environmental Protection Agency (EPA), birthed AERMOD, a model that straddles multiple terrains and scenarios. AERMOD is nothing short of a chameleon; it effortlessly adapts, whether tasked with modeling surface-level pollutant sources or those lofted high into the atmosphere [31, 32]. Predominantly leaning towards gas-phase dispersion, AERMOD is a force to reckon with when charting the course of gaseous pollutants [32]. Yet, even this collaborative genius is not without its idiosyncrasies [2]. Grounded in certain assumptions, AERMOD, on occasions, might slightly deviate from ground truths, especially when nature throws a curveball [6, 31].

Further east, the UK boasts its flagship model: the UK-ADMS. Functioning as a primary regulatory tool, ADMS (Atmospheric Dispersion Modeling System) is the embodiment of versatility [33]. Whether it's the bustling lanes of London or the industrial heartlands of Manchester, ADMS navigates the challenges with aplomb, predicting the fates of both gases and particulate matter [2, 33, 34]. Its robustness, however, has been under the scanner in some circles [2, 6, 33]. Comparative studies occasionally highlight ADMS's potential hesitancy in predicting concentrations, especially when juxtaposed against its international counterparts, suggesting a potential underestimation in certain circumstances [33, 34].

Lastly, but by no means the least, stands SCREEN3. Deceptively simple, this regulatory screening model doesn't aim for the minutiae but goes for the big picture [10, 28]. Envisioned as a tool to provide a bird's-eye view or worst-case scenarios, SCREEN3 is versatile, accommodating an array of source types from point to volume [10, 31, 34]. Adding a touch of realism is its consideration of building downwash effects, accounting for the dramatic impacts towering structures can have on dispersion patterns. But, SCREEN3, like its peers, carries the baggage of its foundation—the Gaussian dispersion equations [2, 6]. Historically revered for their simplicity, these equations come with inherent assumptions which, when thrust into the chaotic realms of complex scenarios, might falter [35].

In essence, the realm of atmospheric dispersion modeling is vast and diverse. Each model, a product of rigorous research and development, offers a unique lens to view the atmospheric dance of pollutants [1, 34]. Choosing the right tool requires not just understanding the model but also the landscape it's meant to decode.

3.3. *Eulerian and Lagranigian Models*

Eulerian and Lagrangian models offer two distinct methods for atmospheric dispersion modeling, each catering to specific scenarios and requirements.

TAPM, or The Air Pollution Model, adopts an Eulerian grid-based approach for broader regional dispersion modeling while introducing a Lagrangian particle mode for precise near-source concentrations [36, 37]. In TAPM, the atmosphere is depicted as an incompressible non-hydrostatic fluid [38, 39]. The dynamics of horizontal wind components are derived from momentum equations, and the model intricately treats cloud processes [37, 40, 41]. It encapsulates boundary layer parameterization using similarity scaling and employs a k-e methodology for turbulence. On the surface, conditions are shaped by variations in surface temperature and moisture for distinct soil and land types [36, 41]. TAPM also emphasizes dry deposition using a resistance method [23, 39]. This model regards scalars similarly to heat, especially in terms of roughness and stability, with surface resistance modified based on the surface type[23, 40, 41]. However, wet deposition in TAPM is restricted to highly soluble gases and particles, determining partitioning based on the liquid-rain water volume fraction. Despite minor discrepancies in predicting concentrations during certain atmospheric conditions, overall, TAPM

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exhibits reliability, with an excellent record in predicting NO2 and other concentrations even without meteorological data inputs [38, 39, 41].

ARIA Regional, on the other hand, is designed to analyze gas and particle dispersion originating from diverse sources such as industries, transportation, and large areas, spanning up to 1000 km [36]. ARIA's strength lies in its adaptability; it can analyze multi or single constituent isothermal and non-isothermal gas flows based on their thermodynamic properties [37]. Its meteorological component employs a turbulence and deposition processor, capable of calculating wind flows across various terrains, ranging from microscale to synoptic scale [23, 36, 37]. ARIA stands out for its dual approach: the Eulerian-based FARM model and the Lagrangian-focused SPRAY model [37, 40]. While FARM concentrates on reactive emissions, including photochemical gases over long distances, SPRAY zeroes in on non-reactive emissions, especially over complex terrains [13]. SPRAY, a Lagrangian particle model, excels in modeling dispersion for multiple sources across various scales, with recent updates enhancing its capability further [38, 40]. However, despite its strengths, SPRAY does face challenges in accurately calculating aspects like daytime turbulence, affected by thermal convection [37]. In practice, the model showed generally reasonable agreement with measured pollutants like NOx and SO2, barring a few exceptions.

In summary, both TAPM and ARIA Regional offer unique insights and tools for atmospheric dispersion modeling [37]. While TAPM is holistic and excels in regional scenarios with a strong emphasis on photochemistry and accurate predictions, ARIA Regional's strength lies in its flexibility and dual modeling approach, catering to both reactive and non-reactive emissions across vast terrains [34]. Both models, however, face their own set of challenges and discrepancies, underlining the inherent complexities of atmospheric dispersion modeling [23].

4. Conclusion

Dispersion modeling packages play an indispensable role in understanding the spread of particles in the atmospheric realm. In an effort to elucidate the capabilities and constraints of these tools, this paper offered an exhaustive review spanning various modeling approaches: Box models, Gaussian models, and even intricate ones like Computational Fluid Dynamics (CFD) and Lagrangian/Eulerian models[24, 28, 34]. The scope also covered models emphasizing aerosol dynamics. One of the most striking revelations of this review was the pronounced variability and inherent limitations of the surveyed models [15, 34]. Each model, based on its distinct mathematical structure and treatment of aerosol processes, is suitable for specific scenarios and unsuitable for others.

Several critical factors emerged as determinants in the choice of a model. The complexity of the environment under study, the model's scale, the nature of the particle source, the computational resources at disposal, and the desired accuracy and timeline for outcomes are all pivotal in this decision-making process. However, it's imperative to understand that even the most advanced models are mere approximations of the real-world dynamics, largely owing to unpredictable elements such as wind variations and inconsistencies in emission strengths. This understanding throws light on another significant insight: the constraints faced due to limited computing power, time, and uncertainties in parameters like emission factors [42]. It became clear that these constraints and uncertainties necessitate an astute assessment to ensure the resultant concentrations are within acceptable margins of error and resonate with the real-world timeline.

A significant chunk of the discussion revolved around the considerations for particle dispersion. The appropriateness of models for particle dispersion is tightly tethered to the type of concentration in focus. For scenarios necessitating a deep dive into particle number concentrations, especially in proximal sources in urban locales, a comprehensive aerosol dynamics model becomes indispensable [26]. However, in broader, regional contexts, the influence of aerosol dynamics on particle mass concentrations appears to be on the minimal side. This finding underpins another crucial observation: most dispersion models, especially those not diving deep into intricate chemistry and particle dynamics, are primed for predicting mass concentrations. They majorly lean on the principle of mass conservation at every timestep [31]. This observation cemented the belief that, when it comes to predicting average daily and yearly particle mass concentrations in uncomplicated and regional domains, gas-phase dispersion models hold their ground.

Yet, a paramount note of caution surfaced: no model can be universally crowned as the "best." The review refrained from a hierarchical ranking, underscoring that the performance of a model is context-dependent. A model's efficacy can oscillate based on the specific scenario or dataset at hand. Hence, any comparison between models should be rooted in the context, giving users the discretion to cherry-pick based on their specific requirements. Despite the comprehensive nature of the review, certain gaps were hard to overlook. There's a palpable dearth of studies focusing simultaneously on particle number concentration and gaseous pollutant concentrations. Another conspicuous absence is that of validation studies that juxtapose various model performances against a set validation dataset, potentially because many aerosol dynamics models haven't ventured into the commercial domain yet. In conclusion, while this paper strived to be exhaustive, it doesn't encapsulate every nuance. Yet, it stands as a pivotal resource, especially at a juncture when understanding particle dispersion in the atmosphere is gaining unprecedented importance.

5. Acronyms

1. AERMOD – American Meteorological Society (AMS) /Environmental Protection Agency (EPA) Regulatory Model
2. AEROPOL – AERO-Pollution
3. ARIA Local – Atmospheric Resource Impact Assessment – Local
4. ARIA Regional – Atmospheric Resource Impact Assessment – Regional
5. AURORA – Air Quality Modelling in Urban Regions using an Optimal Resolution Approach
6. CALINE4 – California Line Source Dispersion Model
7. CALPUFF – California Puff Model
8. CAR-FMI – Contaminants in the Air from a Road – Finnish Meteorological Society
9. CPB – Canyon Plume Box
10. FARM – Flexible Air quality Regional Model
11. GRAL – Graz Lagrangian Model
12. HIWAY2 – Microscale California Photochemical Grid Model
13. MICRO-CALGRID – Microscale California Photochemical Grid Model
14. MISKAM – Microscale Flow and Dispersion Model
15. PBM – Photochemical Box Model
16. SCREEN3 – Screening Version of ISC3 model
17. SPRAY – A Lagrangian Pollution Dispersion Model, without exact words that it stands for
18. TAPM – The Air Pollution Model
19. UK-ADMS – UK – Atmospheric Dispersion Model System

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