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An Introduction to Atmospheric Pollutant Dispersion Modelling †

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Abstract: Modelling the dispersion of atmospheric pollutants plays an important role in regulatory and epidemiological settings. Although the majority of modelling concepts were developed in the 1980s, a significant amount of optimisation and refinement of dispersion models has occurred since this time. In addition, some completely novel models such as computational fluid dynamics have emerged. Furthermore, next generation models are continually improving the accuracies of the results obtained. This review provides a non-technical outline of the mechanisms of atmospheric pollutant dispersion modelling and discusses common model types and their applications.

Keywords: Gaussian model; Eulerian model; computational fluid dynamic (CFD) model; Lagrangian model

1. Introduction

With the well-established link between various forms of air pollution and detrimental health conditions including respiratory conditions [1–3], cardiovascular disease [4–6], cancer [7,8] and other systemic conditions [9,10], the importance of maintaining air quality has never been more accentuated. Particularly in light of the continuing decrease in ambient air quality in regions such as East Asia [11], the modelling of atmospheric pollutants plays a vitally important role in guiding regulatory decisions relating to existing and future air quality [12–14]. In addition to providing the ability to predict (i.e., forecast) pollutant levels at a given timepoint [15], pollutant modelling allows for specific pollution events to be traced back to their most likely origin [16]. Amongst the numerous potential uses, this is particularly important for regulatory decision making or planning [12,17,18], epidemiological studies [19,20] and forensic purposes (i.e., identification of the polluter(s) responsible for an observed reduction in air quality) [16,21,22]. Air quality monitoring also plays in important role in allowing industries to demonstrate their compliance with national air quality standards [23].

The choice of pollutant dispersion model plays a key factor in the accuracy of the results obtained [24]. The available modelling techniques were reviewed by Daly and Zannetti [25] over a decade ago, and more recently by Barratt [26] and Colls and Tiwary [27]. In addition, several recent reviews have focussed on the modelling techniques specifically associated with traffic-derived atmospheric pollution [28,29]. However, the technical jargon associated with such reviews may render them unintelligible for the layperson. This review aims to provide a simple introduction to atmospheric pollutant dispersion modelling in terminology accessible to the uninitiated and outline the currently available models. Particular emphasis is given to models and applications reported over the past five years.

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2. The Basics of Dispersion Modelling

2.1. Data Input

The basic inputs of a pollutant dispersion model include the emission source(s) and pollutant emission levels, meteorological conditions and any changes, topography and any chemical processes (if applicable). A range of possible inputs is given in Table 1.

Table 1. Some possible data inputs for a dispersion model.

Emission Characteristics	Source Characteristics	Location Characteristics	Meteorological Characteristics
Pollutants	Source types (e.g., point, line, area, volume)	Location (e.g., urban vs. rural)	Temperature
Pollutant characteristics	Source dimensions (if applicable)	Terrain (simple vs. complex)	Wind speed
Distribution of source(s)	Volume emission rates	Surface roughness (z_0)	Wind direction
Emission rates	Temperature	Interfaces of land & water (if any)	Atmospheric stability/turbulence
	Moisture content	Existing (background) pollutant levels	Solar radiation (particularly important for photochemical modelling)
	Presence of buildings or other infrastructure		Cloud cover
			Moisture

2.2. Data Processing—The "Black Box"

For many, the model comprises a "black box" wherein the necessary data is entered, the start button is pressed and the outputs consequently analysed. Indeed, with the rising complexity of the models available, it would be impractical for most users to spend the time necessary to gain a complete understanding of the operations of the model they are using.

At the most basic level, atmospheric models comprise one or more mathematical formulae that take into account the input parameters to calculate the concentrations of one or more pollutants at specific locations at any point downwind or downtime. Clearly, the most accurate results would be gained from modelling the trajectory of every pollutant molecule over the simulation period. However, this would require an inordinate amount of processing power. Rather, models must simulate pollutants as a number of discrete components, typically taking either a fixed grid (Eulerian) or trajectory approach. With the fixed grid approach, the area in question is divided into a grid; the air quality within each grid is calculated at each time point based on its previous air quality and that of adjacent grids, taking into account the prevailing meteorological conditions [23]. In the simpler trajectory approach, the emissions are chunked into either a single block or a number of "puffs", each comprising a potentially variable (albeit known) amount of pollutant [30]. The directional and temporal spread of each puff is then simulated.

In order to do this, processing power is divided among a number of modules, each connected to the core "dispersion" module. Each module simulates a specific aspect within the simulation, such as the identity and concentrations of any pollutants present, any chemical reactions, effects of buildings or terrain, effects of meteorology, plume rise, and deposition of pollutants. Other modules may be added onto a model. For example, the module PRIME (Plume RIse Model Enhancements) is included in many regulatory dispersion models (e.g., ISC, AERMOD, CALPUFF, TAPM, AUSPLUME), allowing for the prediction of turbulent flow and mixing induced by buildings.

Some models ("reactive models") also allow for chemical reactions between components to be simulated. This allows for more realistic prediction of the true atmospheric quality, albeit at a higher processor cost. Concentrations of compounds such as CO and SO_2

are often forecast using non-reactive models due to their relative inertness, while the more chemically reactive species NO, NO_2 and O_3 necessitate the use of reactive models [23].

2.3. Data Output

Although outputs will depend on the specific application to which the model is applied, the most important output is typically the predicted concentrations of specific pollutants at given point(s) surrounding the emission source, at specified points in time.

Before being released to the general public, the outputs of a new pollutant dispersion model will be calibrated against the true pollutant levels across a number of sites, obtained from air quality monitoring stations. Particularly with the rise of cheaper air quality monitoring stations which could be implemented more widely [31,32], the validation of dispersion models, both pre- and post-release, is expected to only increase in the future.

2.4. Data Analysis

From the data output, an assessment of likely environmental or health effects can then be made. Despite its seeming simplicity, accurate interpretation of the model output is of the utmost concern. If the model results are not interpreted correctly, then there is little point in running the model in the first place.

2.5. Simulation Timeframe

Models can either be short-term (hours to days) or long-term (months to years) [23,33]. Short-term modelling is typically used for predicting pollutant levels under "worst case" scenarios. On the other hand, long-term modelling is often used for epidemiological and atmospheric deposition studies [23].

3. Box Models

3.1. Introduction

Box modelling is one of the earliest and simplest forms of pollutant dispersion modelling. Traditionally, box models found particular use in situations requiring the simulation of chemical interactions between pollutants, as the simplified spatial and temporal dispersion allowed for a greater focus on the chemical aspects.

In a box model, the airshed is assumed to be a simple box of set dimensions, with all emissions released into the box. Once released, the emissions are assumed to be evenly distributed throughout the box. As expected, the accuracy of such a model is quite limited, as shown in comparative studies [34]. The main advantage of the box model is its simplicity, thus requiring very little processing power and allowing for very fast simulation runtimes. In addition, very little input data are required.

3.2. Examples of Simple Box Models

EKMA

The model EKMA (Empirical Kinematic Modelling Approach) was used as an early method of assessing the likelihood of photochemical smog formation in urban settings [35]. In this model, the concentrations of VOCs and NO_x were assumed to remain constant from their values measured in the early morning. EKMA is in fact a type of Lagrangian simulation, albeit limited to a box model system [36,37]. Despite its age, EKMA is still occasionally used for the study of ozone-NO_x-VOCs relationships in simple settings [38–40].

3.3. Uses

Given their overt simplicity, box models are not commonly used in contemporary regulatory settings except for preliminary assessment purposes [41]. However, they do retain a place in pollutant dispersion modelling, particularly in small anthropogenic enclosed spaces. For example, Lin, et al. [42] applied a box model to investigate the disappearance of formaldehyde from indoor air spaces via photodegradation. Given the relatively small air spaces indoors coupled with the frequent lack of ventilation to the outdoor environment,

the use of a box model is quite appropriate in such circumstances. Nevertheless, more complicated models have also been applied to the indoor environment [43].

Modified versions of the box model, such as a two-box model, have been utilised in modelling photochemical pollutant levels in street canyons (i.e., a street enclosed by tall buildings on each side) [44,45], amongst other uses [46]. Many other models designed specifically for street canyons are based off the box model, albeit typically modelling each street as an individual box. Examples include the STREET and STREET-BOX models [47,48].

4. Eulerian Models

4.1. Introduction

Eulerian models take a strictly mathematical approach to pollution modelling. The area of study is divided into a number of grid cells, both horizontally and vertically, and the average pollutant concentration within each cell is calculated at each time point. Eulerian dispersion modelling was introduced by Reynolds, et al. [49]. Although initially used for modelling time periods of only a few days per simulation, more recent versions may be used for longer periods of time.

As Eulerian models are based on the average grid concentrations rather than following an entire plume, they easily account for removal of the constituent particles through deposition or chemical reactions [50].

4.2. Examples

4.2.1. TAPM

The Air Pollution Model (TAPM), developed by CSIRO [51], is unusual for a dispersion model in that it can use either a Eulerian grid or Lagrangian particle model to calculate dispersion [52]. The latter is considered to be more accurate at locations close to the emission source. Another remarkable aspect is its ability to extract meteorological conditions from synoptic charts (past, present or forecast). Surface measurements can also be incorporated.

TAPM functions particularly well in complex situations, such as locations with a sea breeze or complex terrain [53]. The incorporated prognostic meteorological model has also been used to provide meteorological input data for other dispersion models [54,55]. As expected for a mathematical-based simulation, TAPM is quite computationally intensive.

Recent applications of TAPM include use in a complex, mountainous terrain [53], modelling of heavy metal deposition around a copper smelter [56] and evaluation of health risks resulting from VOC emissions from municipal waste [57].

4.2.2. Variable K-Theory Model

A Eulerian Variable K-Theory model has been found to provide the highest accuracy compared to box, Gaussian plume and Lagrangian models, when simulating NO_2 and SO_2 concentrations across 17 sites [34].

5. Gaussian Models

5.1. Introduction

Based off the assumption that plume spread is due to the diffusion of the constituent pollutants, Gaussian models take the pollutant concentrations to follow a normal (Gaussian) distribution in both the horizontal and vertical aspects [23], as determined through experimental measurements of plume spread [58]. These models have been in regulatory use in the USA for almost 60 years [23]. Gaussian plume models assume the pollutants are emitted at a continuous rate, modelling the pollutants as a single, continuous plume (Figure 1). Gaussian plumes expand in two dimension over time (y and z). Gaussian plume models require the following assumptions: the emission and meteorological conditions must remain constant, no chemical transformations occur, and wind speeds always equal or exceed 1 m s^{-1} [23].

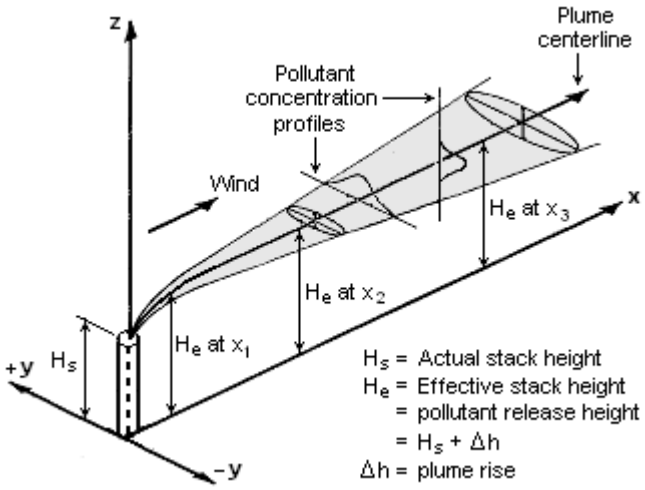

Figure 1. A representation of a Gaussian plume model. Image by Milton Beychok; reproduced under Creative Commons 3.0 licence.

Inputs include factors such as the pollutant release rate, release height, wind speed (at the reference height; often the height of emissions release), mixing/inversion height, and the horizontal and vertical dispersion factors. In addition, rising or sinking of the plume may be modelled. When the plume reaches the ground or the upper boundary layer of air, it is assumed to reflect quantitatively from these surfaces. Over time, this may lead to the false appearance of pollutants accumulating at ground level, which can be accounted for in the model [23]. Further detail surrounding the mathematical calculations underlying Gaussian models is presented by Godish, et al. [23], hence will not be discussed in detail here.

5.2. Gaussian Plume Models

5.2.1. AEOLIUSF

AEOLIUSF (Assessing the Environmental Of Locations In Urban Streets Full version) is designed for modelling dispersion in urban street canyons [26,59]. Compared to other street models, AEOLIUS/AEOLIUSF is not as commonly used in contemporary settings. Its accuracy is middle-of-the-range, being higher than STREET models but lower than SEUS models [24].

5.2.2. AERMOD

AERMOD superseded the ISC model as the preferred Gaussian plume model of the US EPA [Environmental Protection Agency [EPA] [60]. From the meteorological conditions, terrain and upper atmospheric conditions entered, a single wind field is calculated and used in the simulation, while the terrain, elevation, surface roughness and land use is used to calculate factors such as the turbulence, stability class and Monin–Obukhov length (a continuous measure of near-surface atmospheric stability) [25]. AERMOD is suitable for ground or elevated sources, and both simple and complex terrain [60]. AERMOD is unsuitable for modelling when the wind speed is zero [61].

Contemporary applications of AERMOD include assessing complex industrial emissions [62], modelling emissions from cement factories [63,64] and gas-fired power plants [65], and predicting near-road pollutant levels [66]. In particular, pollutant sources surrounded by complex terrain are often modelled via AERMOD [67].

5.2.3. AUSPLUME

This model is often used in Australia and New Zealand. Its level of detail is believed to be a little lower than AERMOD; however, AUSPLUME can be used in modelling situations where the wind speed is zero [61].

Recent applications of this model include mapping the dispersion of radon released from a Romanian uranium mine [68] and modelling odour dispersion from rubber factories in Malaysia [69].

5.2.4. CALINE3

CALINE is a modified Gaussian plume model, where the emission source is a line rather than a point [70]. Its main use is in modelling pollutant dispersion from roads; the road geometry can be varied rather than being restricted to a straight line. CALINE3 remains an EPA-recommended Gaussian line model [60], while the updated CALINE4 model is also used in some contemporary applications [71]. CALINE3 is designed for relatively simple terrain and forms the basis of models such as CAL3QHC and CAL3QHCR [60].

Ref [71] used CALINE4, combined with ISCST3, to predict NO_2 and PM_{10} concentrations along a road line source.

5.2.5. CAL3QHC and CAL3QHCR

Both models are based off CALINE3, but are specifically designed for determining the build-up of CO hotspots resulting from traffic stagnation, particularly at intersections [60]. CAL3QHCR is the "refined" version of CAL3QHC and consequently requires a greater data input, in particular localised meteorological data [60].

In recent years, CAL3QHC has been applied to the epidemiological study of the congenital effects of vehicular pollutants [72] and assessing the spread of CO, NO_x and VOCs from intersections in India [73].

5.2.6. CTDMPLUS

As suggested by its name, Complex Terrain Dispersion Model Plus Algorithms for Unstable Situations (CTDMPLUS) is designed for use in complex terrain situations [74]. However, it can be used in all stability conditions (including stable and neutral conditions) [60]. From a review of the literature, it does not appear to be commonly used in contemporary settings.

5.2.7. ISC

The ISC (Industrial Source Complex) model, as reported and evaluated by Bowers, et al. [75], was previously the approved Gaussian plume model of the US Environmental Protection Agency (EPA) [25]. Recent applications of the ISC model include modelling VOCs downwind of a petrochemical manufacturing plant [76] and monitoring a number of pollutants (CO, VOC, NO_x and PM_{10}) in Italian agricultural land [77,78]. Some authors also combine the ISC model with AEROMOD for improved accuracy of the results [79,80].

5.2.8. OCD

The Offshore and Coastal Dispersion (OCD) model is a straight-line Gaussian model, designed for predicting the dispersion of pollutants over marine or coastal regions [60]. Changes as the plume crosses the coastline are incorporated [60]. This model has been used across a range of environments, including the Gulf of Mexico [81].

6. Lagrangian Models

6.1. Introduction

Lagrangian models simulate a number of "puffs" of pollutants emitted from the source, usually at regular intervals. The most common is a "Gaussian puff" model, where each puff is assumed to follow a Gaussian distribution as it moves downwind and expands. The puffs are 3D elements that expand in all dimensions (x, y and z) over time, concurrently moving downwind from the emission source. A model may comprise hundreds to hundreds of thousands of these theoretical puffs [25]. As each puff is treated independently, they can have varying rates of dispersion and move in various directions, allowing for more

realistic modelling of local conditions within the simulation. Another related model of this type is the Lagrangian random walk model, where the plume is discretised as numerous independent tracer particles. The particles are transported by the mean wind field with local turbulence accounted for using a stochastic 'random walk' algorithm. In particular, Lagrangian models show improved accuracy in models with complex topography or flow patterns (e.g., recirculation of the pollutants) and temporal variation in emissions or meteorology.

Lagrangian models are often used for modelling across longer distances and time-frames (up to several years long) [25]. In contrast, Gaussian plume models are typically restricted to predictions up to 50 km from the point source [23]. The field of Lagrangian modelling was introduced by Rodhe [82], Rodhe [83] and has gathered momentum rapidly since that time.

Time steps of between 1 and 180 s may be used [14,84]. As with Gaussian plume modelling, the assumption of complete pollutant reflection from the ground and upper atmospheric boundary layer may result in misleading conclusions unless this is taken into account [85,86].

6.2. Examples

6.2.1. AFTOX

The AFTOX (Air Force Toxic) chemical dispersion model was created by Kunkel [87]. It assumes that four Gaussian puffs are released from the source every minute. As it does not account for decay or settling of the pollutants, AFTOX often provides higher pollutant concentrations and a lower accuracy compared to other model types [34]. AFTOX is restricted to neutrally buoyant gases, but is particularly useful for modelling liquid spills which subsequently evaporate [88].

AFTOX was recently used as the basis for modelling investigating the relationship between raindrop size and the scavenging efficiency of aerosol particles [89].

6.2.2. CALPUFF

CALPUFF is the approved long-range (>80 km) atmospheric emissions model of the US EPA [25]. However, CALPUFF also finds use in short-range simulations with complex surface topography [90], being widely used as a regulatory model in Australian and New Zealand. CALPUFF is versatile at both short-range and long-range simulations [25]. With more recent software such as VISTAS version 6, CALPUFF can be run at timescales of less than one hour [91].

Applications include the modelling of odour dispersion [92,93], dispersion of various pollutants produced by industrial facilities [62] and in the study of shipping emissions in a Western Australian port [94]. One application of particular note was the modelling of atmospheric mercury released from a coal-fired power plant in Mexico [95]. Numerous studies have also utilised CALPUFF in modelling dispersion over complex terrain [90,96].

6.2.3. Hybrid Eulerian–Lagrangian Dispersion Models (HDMs)

Hybrid models combining the Eulerian and Lagrangian methods, as outlined by Andrén [97], remain relatively common, particularly for simulating dispersion close to point-line sources [14]. The emissions are initially modelled as puffs using the Lagrangian method, then after travelling a specified distance or expanding to a specified level, the puffs are assumed to approximate a volume emission. The Eulerian method then takes over to calculate the long-range pollutant dispersal [14]. The major advantage of this method is the reduction in required processing power compared to using a Lagrangian model at all scales [14].

Hybrid models have been used in complex environments such as complex urban [98] and mixed industrial-residential environments [99], and in combination with CFD modelling to predict PM_{10} levels [100] and other pollutants [101] in complex scenarios.

7. Computational Fluid Dynamics Models

7.1. Introduction

Computational fluid dynamics (CFD) has been a well-established modelling technique in engineering disciplines for many years. However, it is only relatively recently that it was turned toward the application of modelling atmospheric pollutants. CFD is based off Navier–Stokes equations, which are 3-dimensional, unsteady, non-linear, partial-differential equations that can exactly model the flow of atmospheric gases. The number of unknowns exceeds the number of discretised equations, hence different techniques are used to model unknown turbulence terms to find a solution. A Langrangian or Eulerian framework is incorporated into the model in order to calculate the transport and dispersion of contaminants through the atmosphere. Depending on the fidelity and resolution of the simulation, CFD models can require very large amounts of computing power.

There are three major classes of CFD models: Reynolds-Averaged Navier Stokes (RANS), Large Eddy Simulation (LES) and Direct Numerical Simulation (DNS).

7.2. Examples and Uses

Brown, et al. [102] compared the Quick Urban and Industrial Complex (QUIC) dispersion model, based off empirical parameterizations of the flow around and between buildings in order to model wind flow in an urban environment. Although the results of the model provided a similar accuracy to standard CFD modelling in this instance, the advantage of CFD models is that wind flow through novel building configurations and/or combinations can be simulated, rather than relying on empirical data.

Mocho, et al. [43] used a CFD model to investigate the movement of formaldehyde in an indoor setting. As expected, the accuracy of the results was improved over a simple box model.

CFD has been used for modelling the near-field dispersion of pollutants, when plumes of varying buoyancies were present [103]. Other uses include the modelling of PM_{10} movement [100] and the movement of reactive chemical components [104].

8. Street Network Models

8.1. Introduction

The street network model is currently the least utilised contemporary modelling technique [105]. This method, designed for the analysis of vehicle emissions in built-up urban environments, typically treats each street as a line source of emissions, with the quantity of emissions calculated from the traffic volume along that street.

8.2. Examples

SIRANE

The SIRANE model, developed by Soulhac, et al. [106], is specifically designed for modelling pollutant dispersal from traffic in urban regions. To date, it is the main street network model reported in the literature [105]. Each street is modelled as a box, with transfer of pollutants occurring along the box (i.e., along the street), between boxes (at street intersections) and between boxes and the atmospheric boundary layer [106]. Atmospheric conditions may change hourly, but are assumed to be constant in between these timepoints. The model has been validated against wind tunnel data [107] and against a year of NO_2 emissions data [105]. However, work by Wang, et al. [108] has suggested that the output of SIRANE shows a poor correlation with near-road NO_2 concentrations, but better correlation with the average NO_2 values. Nevertheless, SIRANE has been used in several epidemiological studies, particularly in France [109–111].

Derivative street pollution models have been created based off SIRANE, including MUNICH (Model of Urban Network of Intersecting Canyons and Highways), which utilises a grid modelling method [112].

9. Other Models

Specialised models are available for modelling pollutant dispersion over long ranges, in complex terrain, and for photochemically reactive pollutants [23]. One example is the Operational Street Pollution Model (OSPM) for modelling the chemistry of photochemical smog formation [108,113]. Other common photochemical models include CMAQ (Community Multiscale Air Quality), CAMx (Comprehensive Air quality Model with extensions), UAM (Urban Airshed Model®) and CALGRID [25]. Of particular note are CAMx, an open-source model [114], and UAM, the most widely used photochemical air quality model [25].

Statistical models are available for the short-term forecasting of air quality, based off recent and current air quality measurements [115]. Such models do not seek to establish cause and effect, rather solely aiming to link patterns in emission trends to the air quality [25]. In a similar fashion, machine-learning algorithms have also been trialled for the prediction of O_3, NO_2 and SO_2 concentrations [116].

Due in part to the short lifespan and unique properties of odorous compounds, air quality models specifically designed for predicting the dispersion of such compounds are available [25], such as ModOdor [117].

10. Conclusions

Atmospheric pollutant dispersion models have played an enormous role in setting and regulating atmospheric emission levels and have likely played a vital role in the improvement in air quality observed across many westernised countries over the past few decades. With new applications and models reported on a weekly basis, the emissions modeller is faced with a baffling array of models to choose from. However, a basic understanding of the mechanisms behind each model and the strengths and limitations of each should assist in guiding this choice. Particularly with advances in computer processing power and simulation abilities, the results produced by atmospheric pollutant dispersion models are more detailed and accurate than ever before. It is hoped that regulatory bodies will be able to utilise this accuracy in such a way that the world's air quality continues to improve over the coming years.

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