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Particulate Matter Sampling Techniques and Data Modelling Methods

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http://dx.doi.org/10.5772/65054

Abstract

Particulate matter with 10 μ m or less in diameter (PM₁₀) is known to have adverse effects on human health and the environment. For countries committed to reducing PM₁₀ emissions, it is essential to have models that accurately estimate and predict PM₁₀ concentrations for reporting and monitoring purposes. In this chapter, a broad overview of recent empirical statistical and machine learning techniques for modelling PM₁₀ is presented. This includes the instrumentation used to measure particulate matter, data preprocessing, the selection of explanatory variables and modelling methods. Key features of some PM₁₀ prediction models developed in the last 10 years are described, and current work modelling and predicting PM₁₀ trends in New Zealand—a remote country of islands in the South Pacific Ocean—are examined. In conclusion, the issues and challenges faced when modelling PM₁₀ are discussed and suggestions for future avenues of investigation, which could improve the precision of PM₁₀ prediction and estimation models are presented.

Keywords: particulate matter, modelling, regression, artificial neural networks, instrumentation and measurement

1. Introduction

Particle pollution—also known as particulate matter or particulates—is a complex but stable gaseous suspension of liquid droplets and solid particles in the earth's atmosphere. Particle pollution is known to have many environmental effects from poor visibility to more serious consequences such as acid rain, which pollutes soil and water. The science of air quality is



© 2016 The Author(s). Licensee InTech. This chapter is distributed under the terms of the Creative Commons Attribution License (http://creativecommons.org/licenses/by/3.0), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited. complex, and many aspects of the problem are not understood fully. Particles are commonly classified according to their size as either coarse or fine. Fine particles have a diameter of 2.5 μ m (PM_{2.5}) or less, and coarse particles are 10 μ m or less (PM₁₀). Particulate matter that has a diameter over 100 μ m tends not to stay airborne long enough to be measured. Fine particles are commonly generated through combustion or by secondary gas to particle reactions. These fine particles are typically rich in carbon, nitrates, sulphates and ammonium ions. Coarse particles are commonly the product of mechanical processes but also include naturally occurring wind-blown particles. A common example of coarse particulate matter is dust containing calcium, iron, silicon and other materials from the earth's crust.

Sources of particulate matter are often classified according to whether they originate from natural or anthropogenic sources. Natural sources include particles suspended in the atmosphere by volcanic eruptions, bush fires and pollen dispersal. Mechanistic processes cause natural particles such as dust and sea-salt particles to be suspended in the atmosphere. Biological sources of particulate matter are also natural sources; these consist largely of fungal spores ($\leq 1 \mu$ m) and plant debris (normally < 2 μ m) but also include microorganisms, viruses, pollen ($\leq 10 \mu$ m) and fragments of living things (e.g. skin cells). Anthropogenic sources of biological particles include sources from farming, horticulture, waste disposal and sewage. Another anthropogenic source is emissions from combustion of fuels, for example, vehicle exhaust. In Europe, anthropogenic sources have been identified as the main contributor to PM₁₀ due to urbanisation, high population density and areas of intensive industry. In New Zealand, the main contributors are also anthropogenic but are emissions from winter household heating (i.e. the wide use of wood-burning fires) and industry.

 PM_{10} are so minute that they can be inhaled, penetrate the lungs and cause serious health problems. One event which illustrates the effect of particle pollution on human health is the 1952 'Great Smog' in London. Particle pollution from coal burning hung over the city for four days due to cold temperatures and lack of wind. Approximately 4000 deaths were linked to this single event [1]. As a result of events such as the Great Smog and obvious signs of climate change, many countries are now committed to international and national clean air legislation and air quality standards. These agreements require regular reporting of air quality including PM_{10} concentrations.

The economic costs of particulate pollution on a country can be significant. In the European Union in 2015, the cost of air pollution-related deaths was reported to be over US\$1.4 trillion. In Israel, it is estimated that 2500 people a year die as a result of exposure to air pollutants [2]. In New Zealand (population ~ 4.4 million), it was reported that, despite relatively low air pollution when compared with other members of the Organisation for Economic Cooperation and Development, during 2012 a total of 1370 deaths, 830 hospital admissions and 2.55 million restricted activity days were linked to PM_{10} pollution [3]. Even low levels of PM_{10} have been found to significantly affect human health.

In order to make informed decisions, as individuals or as policymakers, it is critical that particulate matter is measured and modelled appropriately.

2. PM₁₀ modelling

Models can be designed to estimate, predict or project. Discontinuities in data represent a real obstacle for time series analysis and prediction. Thus, estimating PM₁₀ is important in situations where small periods of ground-truth data, acquired from sensors, are missing. Prediction models allow us to determine that something will happen in the future based on past data, generally with some level of probability, and are based on the assumption that future changes will not have a significant influence. In this sense, a prediction is most influenced by the initial conditions-the current situation from which we predict a change. Predicting short-range PM₁₀ is important in order to identify days in which PM₁₀ levels spike so that people with medical conditions which make them vulnerable to air pollution, such as asthmatics, can avoid exposure. It also allows for initiatives such as free public transport days to reduce commuter traffic volumes and thus reduce PM₁₀ concentrations on a predicted high day. Models that allow for long-range projections are also important in order to assess the impact of different air quality management scenarios. A projection determines with a certain probability what could happen if certain assumed conditions prevailed in the future. Most PM₁₀ models are designed to predict short range hourly, mean daily or maximum daily PM₁₀ concentrations one day ahead.

A wide variety of techniques, ranging from simple to complex, have been used to predict PM_{10} concentrations. Mechanistic models are complex three-dimensional physiochemical models requiring theoretical information to simulate, using mathematical equations, the processes of particulate matter transportation and transformation (e.g. the air pollution model (TAPM) [4]). Such models are complex and time-consuming to implement and often prove inaccurate. Mechanistic models require a wide variety of input variables for which ground-truth data are not available. These missing data are either estimated or the model is simplified and all begin with meteorological forecasting, introducing both errors and uncertainties to a model.

Statistical models aim to discover relationships between PM_{10} concentrations and other explanatory variables. Statistical models work on a number of assumptions. Machine learning algorithms, on the other hand, are largely free of such assumptions and learn from the data they are presented with, finding patterns and relationships that are not necessarily obvious in the data. Machine learning approaches also tend to be good at modelling highly non-linear functions and can be trained to accurately generalise when presented with new, unseen data. As a result, machine learning methods have on the whole proven to be better at predicting PM_{10} concentrations than statistical models. This chapter focuses on statistical and machine learning approaches to PM_{10} modelling and prediction.

The vast majority of models in the last decade have been developed using a data-driven approach and have their origins in statistical modelling and machine learning. These models use ground-level sensor data and make no attempt to model the physical or chemical processes involved in PM₁₀ generation, transportation and removal. They are reliant on measurements of pollutants and meteorological variables which are accurate only within a small area around

the monitoring stations. Thus, any model is limited by coverage, reliability and distribution of monitoring stations.

There are several steps in building an empirical PM_{10} model (**Figure 1**). The first is data acquisition from various types of particulate matter sensor. The next step is cleaning and preparing the raw data for analysis, including handling missing data, suspected errors and outliers. The next step, variable selection, is central to the performance of most models [5]. The aim of variable selection is to simplify the model by reducing the dimensions and removing any variables that do not significantly contribute to the model. The model is then built based on this subset of variables. Once a model is established, it is tested, after validation where required, by exposing the model to new data and measuring how well it predicts.





2.1. Particulate matter sampling techniques

The most common instruments for measuring particulate matter measure either its concentration or size distribution. The most accurate measurements are obtained from instruments that use a gravimetric (weighing) method. Air is drawn through a preweighed filter, and particles collect in the filter. The filter is then removed and reweighed. This approach has the added advantage that particles collected in the filter can be analysed chemically [6]. This method involves careful pre- and post-conditioning of the filter. Filter choice is also important as substrates are sensitive to environmental factors such as relative humidity. PTFE-bonded glass fibre has been found to be the most stable type of filter [7]. Accurate weighing is essential, and precise weighing protocols must be followed for results to be comparable [7]. This method is the most widely adopted by regulatory bodies including the EPA and the EU. However, it is not the most pragmatic method for PM₁₀ modelling purposes because it is not real time and provides only average data for the period the filter was deployed. A manual process and consequently high operating costs limit the applications of this method. However, gravimetric measurements may be useful to provide a quick snapshot of PM₁₀ at a site in order to determine locations for more intensive monitoring [8].

The TEOMTM sensor is the most commonly used instrument based on the microbalance method. TEOMTM uses a filter which is mounted on the end of a hollow tapered tube made of quartz. Particles collect on the filter and cause the oscillation frequency of the quartz tube to vary. PM_{10} measurements can be logged in near real time. A study which examined the

measurements on PM₁₀ in New Zealand using microbalance measurement instruments found that the measurements were not equivalent to those from gravimetric methods [9].

Real-time monitoring of PM_{10} concentrations can be achieved using optical instruments. These instruments measure either light scattering, light absorption or light extinction caused by particulate matter. The most common instrument is an optical particle counter (OPC) which uses a light source, normally a laser diode, to illuminate particles and a photodetector to measure light scattered by those particles. Measurements may be periodically verified and calibrated using data from gravimetric instrumentation. OPC instruments have lower purchase and operating costs than gravimetric meters, but their lower precision and sensitivity mean that they are not considered appropriate for compliance monitoring [8]. However, the low cost of OPC instruments and real-time monitoring capability make OPCs suitable for particulate matter research.

Regardless of the data collection methods used, PM_{10} models are reliant on accurate and complete time series data from geographically localised monitoring stations.

2.2. Explanatory variables

Suspended PM_{10} regardless of location is dependent on many factors such as meteorological properties of the atmosphere, topo-geographical features, emission sources and the physical and chemical properties of the particles (size, shape and hygroscopicity). Many natural environmental factors influence PM_{10} concentrations from the time of year, to the weather, to extreme events such as volcanic eruptions and earthquakes. The effect of extreme events in nature on PM_{10} concentrations is well documented: high PM_{10} levels have been reported during heatwaves in Greece [10], as a result of forest fires [11], and in the aftermath of the Christchurch earthquakes in New Zealand [12]. Relatively low PM_{10} concentrations are observed during the monsoon season in India [13]. Of the myriad complex interrelated potential explanatory variables, only a small number have been used in the modelling of PM_{10} concentrations.



Figure 2. Particulate matter and the atmospheric boundary layer.

One key factor commonly used to explain and evaluate trends in PM_{10} data is the impact of meteorological conditions. The atmospheric boundary layer (ABL) is the lowest part of the earth's atmosphere (**Figure 2**). The thickness of the ABL can vary from 100 to 3000 m and extends from the ground to the point where cumulus clouds form. In the ABL wind, temperature and moisture fluctuate rapidly, and turbulence causes vertical and horizontal mixing. Suspended in the ABL, particles may undergo physical and chemical transformations triggered by factors such as the amount of water vapour, the air temperature, the intensity of solar radiation and the presence or absence of other atmospheric reactants. It is these physical processes, which help to explain why meteorological variables have such an influence on PM_{10} concentrations.

Having accurate and complete input data is critical to the success of any PM₁₀ prediction model. As a result, most models make use of data that are readily recorded using weather station sensors. In cases where data are incomplete, the instance is often removed rather than imputed because of errors which may be introduced by estimation processes. The outputs of numerical weather forecast models can also be used as input variables in PM₁₀ models. However, this is not common because of the uncertainties such variables introduce to PM₁₀ predictions [14, 15].

Wind speed and temperature are the meteorological explanatory variables most frequently used in PM_{10} prediction models (**Table 1**). Wind variables have been found to be useful proxies for physical transportation factors; wind is critical to the horizontal dispersion of PM_{10} in the ABL. Wind direction controls the path that the PM_{10} will follow, while wind speed determines the distance it is carried and the degree to which PM_{10} is diluted due to plume stretching. The effect of wind speed and direction on PM_{10} varies with the geographical characteristics of a location. Low wind speed can be associated with high PM_{10} [16, 17]; this is common in hilly or mountainous regions. Conversely, in coastal or desert regions, high wind speeds result in high PM_{10} concentrations due to salt or dust suspension. In Europe, PM_{10} concentrations are significantly influenced by long-range transport contributions, which are independent of local emissions, so both wind direction and speed have a significant impact [18]. In Invercargill, New Zealand, where there are no close neighbours and thus little long-range transboundary PM_{10} , wind speed explains most of the variability in PM_{10} concentrations [19].

Cold temperatures increase the likelihood of an inversion layer forming in many locations. An inversion exists where a layer of cool air at the earth's surface is covered by a higher layer of warmer air. An inversion prevents the upward movement of air from the layers below and traps PM_{10} near the ground. As a result, cold temperatures tend to coincide with high concentrations of PM_{10} . However, in some locations days with high temperatures, no clouds and stable atmospheric conditions result in high PM_{10} [17]. In other locations when the difference between daily maximum and minimum temperatures is large and the height of the ABL mixing layer is low, high PM_{10} concentrations are observed [20].

 PM_{10} levels can be reduced by rain, snow, fog and ice. Rain scavenging, a phenomenon in which below-cloud particles are captured and removed from the atmosphere by raindrops, is considered to be one of the major factors controlling the removal of PM_{10} from the air. The degree to which PM_{10} is removed is dependent on rainfall duration and intensity [21]. While rainfall is a primary factor in PM_{10} concentrations, it has not been used widely in models. This is in part due to the fact that in some countries, there is no rain for long periods of time or little rainfall in summer. The lack of rain data means that it is not often included in PM₁₀ models [14].

Study reference		[16]	[26]	[25]	[34]	[33]	[35]	[35]	[39]	[14]	[23]
Country of study (ISO 3166-1 alpha 3)		GRC	GRC	PRT	CHL	MYS	AUT	CZE	TUR	SAU	MYS
Predicted variable											
PM ₁₀	Daily Hourly	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Explanatory variables											
	$PM_{10} lag$	Y		Y	Y	Y	Y	Y		Y	Y
Co-pollutants	CO ₂			Y		Y				Y	Y
	SO ₂			Y		Y				Y	Y
	NO			Y		Y					
	NO ₂			Y		Y				Y	Y
	O ₃					Y					Y
Meteorological data	Temperature	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
	Temperature lag						Y	Y			
	Wind direction	Y						Y	Y	Y	
	Wind direction lag										
	Wind speed	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
	Wind speed lag										
	Precipitation	Y			Y		Y				
	Solar radiation	Y									Y
	Sunshine hours										
	Air pressure				Y				Y		
	Dew point		Y								
	Humidity (%)	Y		Y	Y	Y			Y	Y	Y
	Cloud cover							Y			
	Date/time	Y	Y				Y	Y			
	Seasonal effects	Y						Y			
	Spatial variables										

Table 1. Explanatory variables used in recent MLR models for predicting PM₁₀ concentrations.

Relative humidity has been used more frequently in models than rainfall. The relationship between PM_{10} concentration and relative humidity also depends on other meteorological conditions. For example, if humidity is high and there is also intense rainfall (such as during a monsoon season), then humidity has a negative correlation with PM_{10} due to rain scavenging. If high humidity is not accompanied by rainfall but is accompanied by high temperatures,

predicted values were calculated to investigate the variations in PM_{10} that were unexplained by the regression model. These residuals were added to the overall mean of the temperaturecorrected PM_{10} data and a simple moving average filter applied to smooth the data. The modelled trend showed peak emissions in 2001 and 2002 with a subsequent steady decline. This trend did not match with those reported by local authorities in their three yearly emission inventories in which a steady decline was reported [58]. However, it is difficult to compare the two. In the inventory, constant emissions are assumed and then modified according to meteorological conditions and are only undertaken every three years. In [58], the method has been modified to allow for emissions that are not constant, and the model is based on hourly observations making it difficult to assess the success of the method.

In 2010, a study was undertaken to identify the influence of weather factors on occurrences of high PM_{10} concentrations—those in which the NES limits were breached—in Blenheim [60]. Blenheim is a small coastal town (population ~ 30,000) in the South Island of New Zealand. The town is on a flat area surrounded by hills on three sides. Blenheim has a dry climate with hot summers and cold winters. A boosted regression tree using a Gaussian link function was used to identify the meteorological variables which best explained the observed variance in PM_{10} concentrations. Mean daily wind speed and average temperature between 8 pm and midnight were found to best explain the variance; these variables were then used as input to a normal regression tree. It was discovered that low wind speed and low temperatures explained the majority of the NES exceedances. A similar result obtained for Invercargill using CART found that low wind speeds and low temperatures in the evening hours also accounted for most of the variation in PM_{10} levels [19]. In both studies, the model was used to account for trends in PM_{10} rather than to predict or estimate PM_{10} .

Much of the PM_{10} modelling undertaken in New Zealand until recently has been for areas in the South Island. This may be due to the fact that there is constant and historic time series data available from a well-maintained network of South Island PM_{10} monitoring stations or that frequent and higher exceedances of PM_{10} limits have been recorded for South Island regions than for regions in the North Island.

An in-depth study of Christchurch's daily mean PM_{10} employing statistical modelling approaches was undertaken using GLM, GAM, generalised additive mixed model with autocorrelated errors (GAMM + AR) and QR [61]. All of the models evaluated used a natural log transform of the PM_{10} response variable as a number of the explanatory meteorological variables impacted PM_{10} concentrations in a negative exponential form. It was concluded that simple linear regression modelling was not a suitable approach as the data violated all of the assumptions. A total of 41 meteorological variables were considered from which a subset of 20 in addition to lag PM_{10} were chosen by forward and backward stepwise selection. Models were built using the response PM_{10} data both without imputation and with missing values imputed by linear interpolation. The GAMM+AR model was found to be the best prediction model and able to explain around 70% of the variability in daily average PM_{10} concentrations [61].

There have been very few models developed using ANNs to estimate or predict PM_{10} concentrations in New Zealand. Gardner and Dorling [48] compared the performance of

different models such as linear regression, feedforward ANNs and CART approaches for modelling mean hourly PM_{10} in Christchurch, New Zealand. As with studies in other parts of the world, ANNs were found to be the best-performing modelling method. In another more recent study, ANNs were combined with a k-means clustering method to group and rank explanatory variables. The data used were from Auckland—New Zealand's most populated city with a population of over 1.4 million. It was found that the inclusion of cluster rankings, derived from k-means cluster analysis, as an input parameter to the ANN model showed a statistically significant improvement in the performance of the ANN model and that the model was also better at predicting high concentrations [62, 63].

Near-ground maximum PM_{10} concentrations for two sites in Timaru, a small rural town, were estimated using a feedforward backpropagation ANN with a hyperbolic tangent sigmoid function [41]. The response and explanatory variables were normalised. Additionally, due to the correlation between the seasonal changes and PM_{10} concentration, the PM_{10} data were divided into high season (winter/autumn) and low season (spring/summer) classes prior to creating the model. The inputs included one-day lagged meteorological variables and one-day lagged PM_{10} , in addition to meteorological variables for the day of estimation. Levenberg-Marquardt optimisation and Bayesian regularisation training were evaluated, and it was found that Bayesian regularisation was the best approach for tuning the weights and bias values for the network. This approach gave good estimations of daily mean PM_{10} concentration for both sites.

Some research has been conducted using TAPM, a deterministic global atmospheric pollutant model, [4] which includes fundamental fluid dynamics and scalar transport equations to predict meteorology and pollutant concentration [64-66]. Localised models of PM₁₀ concentrations for two South Island towns, Alexandra (population ~ 5000) and Mosgiel (population ~ 10,000), were developed. Alexandra has a borderline oceanic semiarid climate - the country's coldest, driest and warmest-due to its geographic location as New Zealand's most inland town. Mosgiel is separated from Dunedin city by hills and is situated on a plain. It has a temperate climate with a significant annual average rainfall of 738 mm. TAPM was found to correctly predict daily PM₁₀ concentration breaches and non-breaches of the NES 66% of the time in Alexandra and 71% of the time in Mosgiel [65]. Another study has looked at TAPM for simulating PM₁₀ dispersion for a single winter in Masterton and also obtained good predictions of PM₁₀ [65]. Yearlong PM₁₀ was modelled using TAPM for Christchurch city. TAPM was reported to provide an acceptable simulation of ground-level weather and PM₁₀ dispersion (with a 4 µg/m³ difference in annually averaged concentration of modelled and measured PM_{10}), but the model tended to overestimate wind speed during still nights resulting in low PM₁₀ estimates for those periods [66].

4. Summary

Although there are now several models available for predicting PM_{10} , it is difficult to compare them. The complex nature of ambient particulate matter composition and the physical and

chemical transformations that particulate matter can undergo between emission source and sampling location seems to mean that PM_{10} concentrations are largely explained by location-specific variables and events. Meteorological variables used in these localised models tend to be restricted to those which are routinely collected by local authorities.

It is also difficult to compare models because of the variation in PM₁₀ instrumentation and measurement approaches used between different studies. In the future, improved sensor technology and lower costs associated with such monitoring could allow for more comprehensive coverage of areas—improving the inputs available for modelling. Ability to sense at different atmospheric levels should also enhance the data and in turn any empirical models. The use of geo-topological features such as elevation and land use could be considered as inputs for modelling as they reflect site-specific conditions and are readily available, but few models utilise these variables. Inclusion of air quality data, such as AOD measurements, from satellite-based remote sensing should also enhance models. Such data have the potential to provide a means of imputing missing values, to verify and enhance the accuracy of sensorbased ground-level observations and to provide additional inputs to models.

While general trends in PM_{10} concentrations can be explained and similarities can be seen between countries and factors contributing to PM_{10} , no empirical comparison can be made between models developed for specific locations. An attempt to develop a single general model for an area found that the general model performed poorly compared with site-specific models [32]. Some of these site-specific issues are removed when a deterministic physiochemical modelling approach is used, but accuracy of such models is currently limited as many of the actual mechanisms involved in pollution generation, dispersal, dilution and removal are not fully understood. However, it is possible that in the future, with better understanding, deterministic models could prove to be the way forwards.

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