

Particulate matter components and health: a literature review on exposure assessment

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Abstract: A number of studies have provided evidence on the association between particulate matter (PM) and adverse health effects. However, PM is a mix of heterogeneous composition that varies greatly by season and region. Epidemiological studies rely primarily on spatial variation of the air pollutants to estimate the associations between exposure to air pollutants and health effects. Since PM components exhibit a great deal of spatial variability, additional monitoring campaign and more refined exposure models are still needed. To our knowledge, the exposure assessment methods for PM components in epidemiological studies haven't been reviewed. Hence, in this review, we summarize several exposure assessment methods which have been applied in estimating the associations between PM components and adverse health effects in epidemiological studies. Many concerns have been accumulated on the potential health effects of PM components with very little evidence focusing on the exposure assessment of PM components in living and work place. Thus, it is quite important to develop exposure models that cover both outdoor and indoor environments for PM components.

Keywords: Exposure assessment; PM components; short term; long term

Received: 04 September 2017; Accepted: 30 January 2018; Published: 30 March 2018.

doi: 10.21037/jphe.2018.03.03

View this article at: <http://dx.doi.org/10.21037/jphe.2018.03.03>

Introduction

Ambient particulate matter (PM) is a mix of heterogeneous components varies greatly by season and by region (1,2). It consists of various components, which includes organic carbon (OC), elemental carbon (EC), nitrate, sulfate and trace elements (e.g., iron, vanadium, nickel). These chemical components are derived from various anthropogenic and natural sources, such as soil or road dust, vehicle exhaust, biomass combustion, sea salt, forest fires, and may have great spatiotemporal variations associated with particular regional and local sources (3).

Since PM is a complex mixture, the toxicology of each

chemical component may vary by time and locations. Multicity studies of PM show that associations between PM and morbidity and mortality vary across areas (4-6), with this variation attributed in part to chemical composition differences. Thus, it is still uncertain that whether certain PM components cause greater public health concern than others. Both National Academy of Sciences (NAS) and U.S. Environmental Protection Agency (EPA) have given prominence to the significance of investigating chemical components and characteristics of PM which contribute to their toxicity (7,8). Identifying most harmful PM components to human health could target control regulations more effectively so that it may reduce the

Table 1 Techniques and procedures used for particulate matter (PM) chemical speciation

Chemical component	Technique
PM mass (Quartz)	Gravimetry
EC, OC	Thermal/optical transmission
NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Cl ⁻ , Br ⁻ , Na ⁺ and K ⁺	Ion chromatography
Al, Ba, Be, Ca, Cd, Cr, Cu, Fe, Mg, Mn, Ni	USEPA IO-3 method with Inductively
Pb, V and Zn	Coupled plasma atomic emission spectroscopy
As, Hg and Se	USEPA IO-3 method with flow injection analysis—atomic absorption technique

burden of disease due to PM at a lower cost.

PM chemical components are usually measured by fixed monitoring stations which are collected and managed by government agencies on the base of regulatory purposes. For instance, in US, mainly two monitoring networks measure PM chemical components, which are the U.S. EPA Chemical Speciation Network (CSN) and the Interagency Monitoring of Protected Visual Environment (IMPROVE) sponsored by the U.S. EPA and other agencies (9-11). There are around 250 monitoring stations measuring PM components, for each single urban area less than 3 monitors on average, in contrast to 2,000 monitors for PM_{2.5} mass across the whole country.

In Hong Kong, PM chemical speciation network was established by the Hong Kong Environmental Protection Department to measure twenty-six PM chemical components. Six general monitoring stations and one road side monitoring station were distributed across the whole area. The frequency of collecting samples was every six days. Thus, it is difficult to estimate the spatial and temporal variability based on the limited numbers of monitors.

PM components usually include OC, EC, ammonium ion (NH₄⁺), nitrate (NO₃⁻), sulfate (SO₄²⁻), bromide ion (Br⁻), chloride ion (Cl⁻), sodium ion (Na⁺), potassium ion (K⁺), aluminium (Al), arsenic (As), beryllium (Be), barium (Ba), calcium (Ca), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), magnesium (Mg), manganese (Mn), nickel (Ni), lead (Pb), selenium (Se), vanadium (V) and zinc (Zn). The common speciation methods and procedures for PM chemical components are summarized in *Table 1*. Due to the complexity of speciation methods and procedures,

PM chemical components are commonly measured every 6 days for each monitoring stations (12).

The exposure assessment methods for PM components in epidemiological studies haven't been reviewed. In this review, we summarize exposure assessment methods which have been applied in estimating the associations between PM components and adverse health effects in epidemiological studies.

Exposure assessment methods for short term exposure to PM components

Evidence have been accumulated on the association between PM components and adverse health effects on short term studies, like time series studies and case crossover studies (13,14). These studies rely on the day-to-day variations of the concentrations of PM components. Usually, associations with health effects were estimated using the exposure of the same day on which the outcome occurs or several days before the outcome occurs. Since the components were measured in every 3–6 days in general, it is very likely that during certain days there is no corresponding data for the PM components. Temporal imputation should be utilized to impute the missing data, while it is still challenging since the missing data might be substantially different from the observed data.

Exposure assessment methods for long term exposure to PM components

Table 2 summarized the exposure assessment methods for long term exposure to PM components. Several studies estimated the individual exposure level according to the monitoring concentrations close to their geocoded addresses. One study attempted to assess the association between PM_{2.5} components and cardiopulmonary mortality in California (15,16), by assigning an concentration based on the nearest monitor according to the geocoded address. In this study, PM_{2.5}, Al, ammonium, Br, Ca, Cl, Cu, EC, Fe, Pb, Mn, Ni, OC, K, Si, Na, sulfate, Ti, nitrate, V and Zn were included. Particularly, different buffer distances were examined in this study. In addition, a study in California (17) explored the association between low birth weights and PM_{2.5} constituents including EC and 35 metals, which applied the same exposure assessment method. In Florida, one study also tried to assess the risk of preeclampsia during pregnancy from exposure to PM components using the nearest monitoring station concentration as the surrogate

Table 2 Exposure assessment methods for associations between long-term PM components and adverse health effects in cohort studies

Approach	Description	Study area	Pollutants studied	Reference
Distance based model	Long term average concentration within certain distance to the nearest monitoring station	California, USA	PM _{2.5} , Al, ammonium, Br, Ca, Cl, Cu, EC, Fe, Pb, Mn, Ni, OC, K, Si, Na, sulfate, Ti, nitrate, V, Zn	(15,16)
		Florida, USA	EC and 35 metals	(17)
		Connecticut, USA	BC, Zn, Pb, Cu, Br, Si, Fe, Ba, Ti, Mn, Al, Ca, K, V, Ni, Na, Cl, S	(18)
Interpolation model	Average of the monitoring concentrations within the same area	Northeastern and mid-Atlantic, USA	Al, NH ₄ , As, Cd, Ca, Cl, EC, Pb, Hg, Ni, nitrite, OCM, Si, Na, sulfate, Ti, V, Zn	(19)
	Weighted average concentrations within the same area	California, USA	PM ₁₀ OC, PM ₁₀ EC, PM _{2.5} OC, PM _{2.5} EC, Ammonium nitrate PM _{2.5} , Ammonium sulfate PM _{2.5} , Biomass burning PM _{2.5} , Diesel PM _{2.5} , Gasoline PM _{2.5} , Geological PM _{2.5} , Meat cooking PM _{2.5} , Residual Oil PM _{2.5} , Sea salt PM _{2.5}	(20-22)
	Interpolation method for which the interpolated values are model by Gaussian process	California, USA	EC, OC, K, Cr, Fe, Ti, As, Mn, Cu, Ni, Pb	(23)
Land use regression model	Models were developed from emission sources, traffic intensity, population density, land use etc.	19 European area (ESCAPE)	Cu, Fe, K, Ni, S, Si, V, Zn	(24-28)
		3 cities in USA, The Multi-Ethnic Study of Atherosclerosis (MESA)	PM _{10-2.5} Mass, Cu, Zn, Si, P, endotoxin	(29)
Dispersion model	incorporate information of emissions, meteorology and atmospheric chemistry to predict the concentrations of PM _{2.5} composition	Texas, USA	nitrite, sulfate, OC and EC	(30)
	evaluate PM composition, particle size and source information, which incorporated information of wet and dry deposition, emissions and transport	California, USA	O ₃ , NO, NO ₂ , CO, PM _{2.5} mass EC, OC, nitrate	(23,31)
Satellite-based methods	combine satellite-derived aerosol optical depth and composition information from the global chemical transport model	Global	inorganic aerosol (SO ₄ ²⁻ , NH ₄ ⁺ and NO ₃ ⁻), BC and organic mass	(32)

PM, particulate matter; OC, organic carbon; EC, elemental carbon.

for each individual. Another study attempted to explore the association between the source of fine particulate matter and risk of preterm birth in Connecticut (18). BC, Zn, Pb, Cu, Br, Si, Fe, Ba, Ti, Mn, Al, Ca, K, V, Ni, Na, Cl, S were also included in the study. Exposure

estimates were assigned to each woman based on the closest monitor of the women's residence at time of birth. Source contributions to PM_{2.5} mass was estimated using Positive Matrix Factorization (33). There are several advantages for Proximity method. First, this method is time-efficient

for long term exposure assessment. In addition, models are derived from the monitoring station data, which have less uncertainty. Moreover, it is cost-effective to conduct compared to detailed exposure assessment which are often quite expensive. The distance based method is useful in preliminary studies when prior evidence is limited. However, this method also has limitations. First, it is very likely to lead to spatial misclassifications since only crude exposure is provided for each individual. This could lead to serious problem, when it comes to a large population and the monitoring stations are scarcely distributed. Second, the distance based method only takes the nearest fixed site monitors into account, fail to account the traffic related air pollution while commuting or indoor air pollution at schools or homes. Third, exposure misclassification tends to be smaller when restricted to small buffer radius, however, thus may lead to reduced statistical power. Fewer incidences may occur within smaller buffer which may cause imprecise estimation to some degree. Another potential issue is that urban areas tend to have more monitors than suburban or rural countries. Ambient monitoring stations are needed for more locations.

A number of studies use unweighted or distance-weighted average concentrations of several close monitors as the exposure concentrations for each individual. One study attempted to explore whether low birth weight was affected by airborne $PM_{2.5}$ chemical components (Al, NH_4 , As, Cd, Ca, Cl, EC, Pb, Hg, Ni, nitrite, OCM, Si, Na, sulfate, Ti, V and Zn) in the northeastern and mid-Atlantic regions of United States (19). If one county has several monitors, the average concentration was calculated. However, different air pollutants were measured in different frequency. $PM_{2.5}$ components were measured every 3-6 days, while gaseous pollutants were measured daily. In order to avoid this problem, daily measured air pollutants and apparent temperature were incorporated to estimate weekly exposure. A study was conducted on women in Los Angeles, which tried to examine the risk of preterm birth when the women were exposed to high level of traffic-related particles, including PM_{10} OC, PM_{10} EC, $PM_{2.5}$ OC, $PM_{2.5}$ EC, Ammonium nitrate $PM_{2.5}$, Ammonium sulfate $PM_{2.5}$, Biomass burning $PM_{2.5}$, Diesel $PM_{2.5}$, Gasoline $PM_{2.5}$, Geological $PM_{2.5}$, Meat cooking $PM_{2.5}$, Residual Oil $PM_{2.5}$ and Sea salt $PM_{2.5}$ (20-22). For women who lived near 2 or 3 stations within 5 kilometers, the exposure values were calculated by distance-weighted method. The aforementioned model is a bit more accurate than the distance based model which incorporate weighted and

unweighted concepts in the model. It is quite useful where the monitoring network is dense. Nevertheless, when the network is sparse, it may cause bias. Another limitation is that geographical patterns or topography have not been taken into account. In addition, outliers in monitoring data can affect the prediction. The third limitation is that some studies did not consider the addresses alterations during the study period. For birth cohort study, some studies only take into account the birth addresses not the actual or prior addresses for the mothers. The fourth limitation is that this method does not deal with the spatial heterogeneity within a large single county, since they use a single monitoring station or average concentration as a surrogate for the whole county. In such circumstance, misclassification may occur for participants who reside far away from the monitoring stations. In addition, there is a challenge that the levels of some constituents are below the minimum detection limit which may cause exposure misclassification.

The Multi-Ethnic Study of Atherosclerosis, also known as the MESA study, utilized another approach to measure exposure to PM. They used partial least squares (PLS) and universal Kriging were used to build the national spatial exposure models (9). The aim of this study was to explore the relationship between low birth weight and exposure to particles by chemical composition (EC, OC, Si, S), size fraction and source. PLS, a kind of dimension deduction methods, was used as covariates in universal Kriging models. In addition, cross-validation was used to select the number of PLS components and assess the model's prediction accuracy. Similarly, another study (23) conducted in California used empirical Bayesian Kriging methods to predict the concentrations of chemical composition (EC, OC, K, Cr, Fe, Ti, As, Mn, Cu, Ni, Pb), which could automatically calculate the parameters through a process of simulations comparing to other Kriging methods. The Kriging method was more accurate than the two aforementioned methods. The model will perform well with good accessibility of monitoring data, often in areas with dense monitoring stations compared to intricate spatial variability. On the contrary, when air pollution monitoring network is sparse, it might easily lead to the spatial misclassification, because large errors are exhibited which cannot represent the actual level of air pollutants. In addition, the ordinary Kriging methods assume that there is no global trend in the air pollution monitoring data, i.e., air pollutants spatial variability is only relied on the distance between monitoring stations.

Land use regression has been applied to predict the

spatial variation of the annual mean concentration for PM_{10} , $PM_{2.5}$, NO_x , NO_2 , VOCs and PAHs. Recently, ESCAPE has measured the PM constituents (Cu, Fe, K, Ni, S, Si, V, Zn) in 20 areas across Europe (34). Land use regression for PM components has been developed based on these measurements for these study areas. Predictors are extracted from Geographic Information Systems (GIS), including emission sources, traffic intensity, population density, land use etc. Several studies have been used this exposure assessment to evaluate the long term PM components and health effects (24-29). The health endpoints including all natural cause, cardiovascular mortality, lung cancer mortality, lung cancer function, respiratory health in birth cohort. In the USA, the Multi-Ethnic Study of Atherosclerosis (MESA) (35) also developed the LUR model to predict the spatial variability ($PM_{10-2.5}$ Mass, Cu, Zn, Si, P, endotoxin) for individuals within the cohorts, including fixed-site ambient monitors placed in density populated areas as well as rotating monitors. Both land use regression and universal Kriging were utilized to evaluate chemical components (36). Land use regression performs well with long term traffic-related air pollution and do not require detailed information on emission sources. However, land use regression fails to separate impacts of pollutants clearly, i.e. there may be large overlaps between the predictors for different PM components, since many of them share the same source. Moreover, both studies required additional monitoring campaigns based on the original measures. Such additional campaigns are time consuming and expensive, since it depends on a great number of monitors to develop the spatial-temporal models. In addition, land use regression does not provide much information on seasonal variability. If the topography and land use are quite different, the transferability is relatively low. One of the concerns in many land use regression is that the exposure measurement in land use regression model is developed at recent times while the cohort study might start in the past. So many studies back extrapolated the exposure concentrations to the baseline as well as the follow up period. Recent studies (37-40) have shown that nitrite dioxide (NO_2) could remain the same up to 10 years, which indicates that LUR model derived from current traffic related NO_2 could predict historical exposure well. These findings may be applied to traffic-related components, for example Zn, Fe and Cu.

The chemical transport model was developed from the air quality model which could predict source appointment to visibility reduction in California (41). The CTM model was developed to evaluate PM composition, particle size

and source information, which incorporated information of wet and dry deposition, emissions and transport (30,31). Recently, a study conducted in Los Angeles assesses the PM components (O_3 , NO , NO_2 , CO , $PM_{2.5}$ mass EC, OC, nitrate) and low birth weight by using the CTM modelled data (23). CMAQ (Community Multiscale Air Quality) model incorporates information about emissions, meteorology and atmospheric chemistry to predict the spatial concentrations of $PM_{2.5}$ composition (30,42). A study conducted in Texas applied the Bayesian Spatial-temporal model using modelled CMAQ data to evaluate the associations between PM components and the risk of congenital anomalies (30). Four main components were included in this study nitrite, sulfate, OC and EC. The dispersion model can be applied to both short term and long term air pollution modelling. For limitations, the above-mentioned methods do not consider the effects of buildings during the exposure assessment. It often requires computing demands, which might be costly to run the model at detailed spatial scale. In addition, this method also demands detailed information of emission sources (for instance, road traffic and vehicle counts). If such information is not available, the model may not develop that well.

Recently, a Canadian team developed a method which combined satellite-derived aerosol optical depth and composition information from the global chemical transport model (32). The study evaluated secondary inorganic aerosol (SO_4^{2-} , NH_4^+ and NO_3^-), BC and organic mass. The method which developed by Canadian team is the first assessment of long term exposure to fine PM constituents all over the world. One of the limitations is that trace metals were not evaluated, which serves as one of the important fine PM constituents that should be added when their assessment capability improves. There are still opportunities remaining to improve the modelled estimates. Finer satellite resolution and simulation would provide better intra-urban gradients.

Discussion

We reviewed different PM exposure assessment methods applied in epidemiological studies of adverse health effects of short-term and long-term exposure to PM components. For short term epidemiological study, the exposure largely relies on the day-to-day temporal variation of the pollutant concentrations. For long term epidemiological study, they include PM composition measurements by ambient fixed-site monitors (e.g., unweighted or distance-

weighted methods), or incorporation of both measuring and modeling data (e.g., Kriging, land use regression, CTM). While these methods enable the assessment of adverse health associations of PM components, several challenges remain. Firstly, the large-scale routinely collected PM chemical composition data only started in the early 2000s, and it may vary by country/location. Thus, monitor-specific data on PM components remain sparse or incomplete in certain areas, resulting in lack of spatial variation and also greater exposure assessment error during land use model development and validation. Secondly and most importantly, ambient PM composition measurements at fixed-site monitors may not reflect personal exposures to ambient PM or its composition due to spatial variability of ambient PM concentrations, the house-to-house and temporal variations in indoor infiltration (i.e., attenuation) of ambient PM, and personal variation in the time spent in different outdoor and indoor places. For example, a panel study of four French metropolitan areas has showed little association between ambient monitors and personal measurements (43). As a result, the attempts to estimate long-term effects using average exposure in epidemiological studies, might lead to bias and wrong confidence intervals in health effect estimates, as well as reduce the power of such studies to build up the proper association between exposure and health effects.

Despite the challenges, there lie opportunities for future development and evaluation of appropriate exposure metrics that can estimate personal exposure well. For example, new research has emerged to explore the use of Exposure Model for Individual (EMI) to predict exposure metrics for actual individuals in the study using ambient air pollutants monitoring concentrations, questionnaire information (e.g., the characteristics of different buildings), meteorology, and information of time and location (44). Another study conducted in Saint Louis intended to explore the association between heart rate ability for elderly people during trips on public transportation and other activities (45). Portable monitors were utilized to measure the air pollution ($PM_{2.5}$ mass, BC, fine particle counts, coarse PM) exposure for each participant. So far, the personal monitoring technique has not been applied in the cohort studies, since it is relatively expensive to measure $PM_{2.5}$ components exposure for each individual. In addition, new research interests have accumulated on the estimating infiltration rates (46), since people spend most of their time indoor and it is not feasible to measure indoor exposure for each individual. Both outdoor and indoor light scattering measurements

were conducted and combined with meteorological data and spatial property assessment data (46). Such ideas have already been applied to $PM_{2.5}$ mass, so it is promising to apply it to PM components. Furthermore, as more and more people are living in three dimensional landscape and exhibit a great deal of population mobility, neither traditional land use regression model nor dispersion model takes into account the vertical profile of the air pollution and the mobility of the population, especially in cities. A recent study which incorporates both three dimensional air pollution and population mobility has been developed in Hong Kong. Such research interest is quite promising in future.

To conclude, exposure assessment for PM components is complicated and need to be considered before exploring the association between PM components and adverse health effects. For example, due to the high spatial variability of PM components, the use of central site concentration alone may not reveal their real spatial variability. More concerns should be given to PM components infiltration rate from outdoor sources and personal monitoring methods.

Acknowledgements

None.

Footnote

Conflicts of Interest: The authors have no conflicts of interest to declare.

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doi: 10.21037/jphe.2018.03.03

Cite this article as: Yang Y, Pun VC, Sun S, Lin H, Mason TG, Qiu H. Particulate matter components and health: a literature review on exposure assessment. *J Public Health Emerg* 2018;2:14.