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New Methods for Personal Exposure Monitoring for Airborne Particles

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Abstract

Airborne particles have been associated with a range of adverse cardiopulmonary outcomes, which has driven its monitoring at stationary, central sites throughout the world. Individual exposures, however, can differ substantially from concentrations measured at central sites due to spatial variability across a region and sources unique to the individual, such as cooking or cleaning in homes, traffic emissions during commutes, and widely varying sources encountered at work. Personal monitoring with small, battery-powered instruments enables the measurement of an individual's exposure as they go about their daily activities. Personal monitoring can substantially reduce exposure misclassification and improve the power to detect relationships between particulate pollution and adverse health outcomes. By partitioning exposures to known locations and sources, it may be possible to account for variable toxicity of different sources. This review outlines recent advances in the field of personal exposure assessment for particulate pollution. Advances in battery technology have improved the feasibility of 24-hour monitoring, providing the ability to more completely attribute exposures to microenvironment (e.g., work, home, commute). New metrics to evaluate the relationship between particulate matter and health are also being considered, including particle number concentration, particle composition measures, and particle oxidative load. Such metrics provide opportunities to develop more precise associations between airborne particles and health and may provide opportunities for more effective regulations.

Keywords

exposure; particulate matter; personal monitoring; sensor technology

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Conflict of Interest

Kirsten A. Koehler declares that she has no conflict of interest.

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Compliance with Ethics Guidelines

Human and Animal Rights and Informed Consent

This article does not contain any studies with human or animal subjects performed by any of the authors.

Introduction

Particulate matter (PM) air pollution ranks as one of the leading causes of morbidity and mortality worldwide [1]. This high burden of disease reflects a range of adverse cardiopulmonary health effects that have been associated with air pollution exposures [2, 3] and the fact that exposure to air pollution is involuntary – we must breathe where we are, regardless of the air quality. Currently in the United States, the Environmental Protection Agency (EPA) requires states to monitor the mass concentration of ambient PM smaller than 2.5 μm ($PM_{2.5}$) and smaller than 10 μm (PM_{10}) at stationary, central locations, sometimes called *'area'* measurements. Measured concentrations are to be maintained below National Ambient Air Quality Standards (24-hr and annual averages for $PM_{2.5}$; and 24-hr for PM_{10}) to protect public health. A wealth of information has been gathered showing consistent associations among ambient air quality and many adverse health outcomes [4, 5]. Such associations are important because air quality regulations are currently limited to ambient air quality - regulations have not been implemented indoors, even for public spaces. Recent studies have shown that even at concentrations below current EPA regulations health effects persist [6, 7]. Such studies may also provide attenuated estimates of the relationship between PM and health because the epidemiological studies that rely on area measurements from central ambient air quality monitors to assign *'personal'* exposures are subject to exposure misclassification [8]. Exposure misclassification results from high levels of within- and between-individual variability in PM introduced by the fact that people are mobile, visiting multiple microenvironments daily (e.g. home, work, school, transit, eateries, etc.), spending a majority of their time indoors [9] and conducting activities that produce PM in their vicinity (the 'personal cloud'). Moreover, PM can vary across a region, meaning that area concentrations measured at a central location may not be representative of exposures in any of these environments where individuals spend their time.

Personal monitoring was pioneered in occupational studies to better characterize exposures of individual workers. In 1960, Sherwood and Greenhalgh [10] introduced the first small, battery-operated pump and air sampling device designed to directly measure personal exposure, a substantial improvement over taking a single area sample to assess exposure. Within a decade, personal sampling came to dominate industrial hygiene as the primary form of assessing exposures [11]. In early studies characterizing personal exposures to PM, cumulative samples were collected on filter media using simple air sampling inlets that were intended to capture "total dust" or "total suspended particulate". Recognizing the wide range of particle sizes relevant to human health (spanning three to four orders of magnitude in particle diameter), size-selective sampling was initiated in the 1970s. The occupational and environmental communities have taken different paths for size-selective particulate sampling. By the late 1990s, the industrial hygiene community had reached general consensus to assess PM exposures using samplers that reflect physiological penetration into different regions of the respiratory tract (inhalable, thoracic, and respirable fractions) [11]. In contrast, regulators of ambient air pollution (i.e., EPA) designated size-based metrics (PM_{10} and $PM_{2.5}$) based partially on health, but also on the sources of pollution that contributed to PM in each size range. Due to the different sampling strategies, exposure

assessment has been predominantly siloed into occupational and environmental categories, rarely capturing a more holistic view of exposures in all microenvironments.

More recently, improvements in personal sampling pumps, sensor technology, and battery technology have enabled researchers to investigate personal exposure to a variety of environmental pollutants beyond cumulative particulate or gas sampling. Direct-reading instruments (DRIs) incorporate sensors that provide a “real-time” indication of contaminant concentrations, allowing simultaneous high-temporal and spatial resolution measures of various contaminants when carried by an individual along their daily route. This review will outline the latest developments in cumulative and direct-reading instruments for personal exposure assessment of particulate air pollutants. A brief description of the operating principle, advantages, and disadvantages are compiled in Table 1. We will not go into great detail on the use of instruments that have been commonly used for personal exposure assessment for more than 5 years. The review will conclude with measurement of important covariates and some remaining challenges for studies deploying personal exposure assessment.

1. Approaches for Estimating PM Mass

1.1. Size-selective Methods for Cumulative Mass

Personal size selective samplers have long been used to collect particles for subsequent gravimetric or chemical analysis. We review these methods briefly here because they remain the most commonly used way to assess personal exposure to PM. Most size selective samplers remove particles larger than a certain size with a cyclone or impactor and then collect smaller particles onto a filter. A suite of samplers, called Personal Environmental Monitors (PEMs, MSP Corporation), rely on an impactor jet to collect larger particles onto an oil-soaked, sintered-metal plate with collection efficiency characterized by the diameter of the particle associated with 50% collection, the cutoff diameter, d_{50}. PEMS are available with various cutoff diameters (2.5 and 10 μm) flowrates (2, 4, and 10 L min^{-1}). These cutoff diameters are consistent with EPA National Ambient Air Quality Standards for $PM_{2.5}$ and PM_{10}. PEMS have been used extensively in environmental research, such as the study of adverse health effects from exposure to secondhand tobacco smoke [12] and the study of particulate triggers on asthma [13].

Other size-selective samplers collect particles according to inhalable, thoracic, or respirable conventions [14]. These conventions are based on how particles interact with the human respiratory tract with shallow collection efficiencies compared to $PM_{2.5}$ and PM_{10} [15]. Inhalable samplers are used for substances that are hazardous if deposited anywhere in the respiratory tract, collecting only those particles that can enter the respiratory system via the nose and mouth (d_{50} = 100 μm). These collection characteristics are achieved with a mouth-like opening (IOM, SKC Inc.) or a perforated curved-surface inlet (Button Aerosol Sampler, SKC Inc.). Thoracic samplers are used for substances that are hazardous when deposited in the lung airways and gas-exchange region (d_{50} = 10 μm). A parallel particle impactor (SKC Inc.) has been used to achieve these collection characteristics. Respirable samplers typically employ a cyclone inlet (e.g., Respirable Dust Aluminum Cyclone, SKC Inc.) to remove

large particles (d_{50} = 4 μm) with a filter to collect the smaller particles that can pass into the gas-exchange region. These samplers are used primarily in occupational settings.

Recent advances in size-selective samplers have sought to improve limits of detection for inhalable sampling and minimize sample losses in existing sampling cassettes. The 37-mm cassette (SKC Inc.) is inexpensive and readily available with pre-loaded, pre-weighed filters for easy field use, but does not conform to any of the size selective sampling criteria. The personal high-flow inhalable sampler head (PHISH), adapts a new inlet for the 37-mm cassette to approximate the inhalable criterion when operated at 10 L min^{-1} of flow [16, 17]. Although the PHISH is not commercially available, it is expected to cost approximately $10, a substantial cost savings over the IOM or Button samplers ($85–250). The increased flow rate of the PHISH compared to the IOM (2 L min^{-1}) or Button sampler (4 L min^{-1}) makes this method desirable when sampling durations are short or to achieve method limits of detection for chemical analyses of low concentration species. Another innovation in inhalable sampling is the use of Accu-cap filters (SKC Inc.), which consist of an acid-soluble cellulose acetate capsule attached to filter media (e.g. mixed-cellulose ester, PVC) [18, 19]. The Accu-cap allows quantification of all particles that enter the traditional 37-mm closed-face cassettes, including those caught on the filter and those that would have deposited on the walls of the cassette. Compared to filters alone, significantly more mass has been recovered using the Accu-caps in occupational environments [19].

Personal cascade impactors are also available to obtain the size distribution of a particulate exposure [20, 21]. Cascade impactors consist of sequential impactors in series with decreasing cutoff sizes. Particles above the cutoff size are collected onto impaction substrates, and those particles smaller than the smallest cutoff size are collected onto a filter. The size distribution of the aerosol can then be constructed from analysis of individual substrates. Relatively recent developments in cascade impactors include the use of polyurethane foam as a collection substrate [22] and the development of a micro-scale impactor using lithography [23].

1.2. Direct-Reading Instruments Using Light Scattering

Light scattering has been used as an indicator of particle concentration for over a century [24]. Photometers are a class of light-scattering device in which an assembly of particles are illuminated within a sensing zone at one time. For particles with a diameter from ~300 nm to ~10 μm, the light scattered is proportional to the mass concentration of the aerosol, although the relationship changes with particle type and size distribution [25]. Particles smaller than 300 nm do not scatter enough light to be detected with a photometer and particles larger than 10 μm are difficult to draw into the sensing zone. Personal, belt-mounted photometers allow rapid (up to 1 second resolution) measurement of particle mass concentrations, such as the Personal DataRam (pDR-1200 and pDR-1500, ~$5,500; Thermo Scientific), SidePak (AM510, TSI Inc.), and the microPEM [26]. The pDR-1200 has been evaluated in laboratory tests [27, 28]. Photometers can be operated with a size-selective inlet to obtain estimates of particulate matter in various size fractions (e.g., respirable, $PM_{2.5}$). Photometers have been used to assess personal particle exposures in widely varying environments from subway stations [29] to hookah bars [30].

Low-cost sensors (~$15, Shinyei PPD42NS; and ~$12, Sharp GP2Y1010AU0F) based on photometry have recently become available. The low cost of these sensors is partially enabled because a light-emitting diode is used as the light source. However, these sensors require integration with a data logger or other communication device and an enclosure for environmental use. The Shinyei sensor has been used in a distributed network to measure spatiotemporal variations of PM2.5 in China [31]. $PM_{2.5}$ measured with the Shinyei sensor at an EPA monitoring site have been shown to compare favorably to more expensive commercial photometers [32]. Although these sensors have been used as stationary environmental monitors to date, they could be enclosed into a battery-powered unit with data-logging capabilities for personal exposure assessment.

A low-cost, light-scattering device based on particle counting (~$400, DC1700, Dylos Corp) has recently been incorporated into environmental studies. In the DC1700, a small box fan pulls particles into a sensing area illuminated by a red laser. The light scattered by an individual particle in the sensing zone is used to place a count into one of two size bins (> 0.5 μm; or > 2.5 μm). The output of the Dylos is particle number concentration, which has been shown to scale with particle mass concentration for a given particle type and size distribution [33]. This instrument has been used to measure second hand smoke [34] and as part of an intervention to reduce exposure to second hand smoke [35]. Although rather large, the Dylos has been incorporated into a backpack for personal monitoring [36].

1.3. Dose-Based Samplers

Recently, samplers have been developed to estimate the fraction of particles that deposit in the human respiratory tract using polyurethane foam as a substrate [37, 38]. The foam plugs are small and operate at rates flow amenable to personal sampling. By estimating the deposited fraction, such samplers seek to provide a more physiologically-relevant estimate of dose. Foam plugs have a lower pressure drop than traditional filter media, for a given flow rate, that remains constant with loading [37]. As a result, inexpensive pumps may be able to operate foam-based devices without the need for automated flow control that substantially increases the cost of personal sampling pumps. However, this advantage is partially offset by the need for relatively expensive chemical analysis. Foam-based samplers are subject to humidity effects resulting in a high gravimetric limit of quantification that makes it impractical for personal sampling over short periods of time (<24 hours) [37]. Instead researchers have conducted chemical analyses of foam substrates for specific PM components or used them as size-selective inlets to other devices [37, 39–42].

1.4. PM Speciation

There is increasing evidence that some sources of particulate pollution are enriched in metals, polycyclic aromatic hydrocarbons and other toxic species yielding a mixture that is more detrimental to human health than other sources of pollution. For example, traffic-related air pollution is a particularly toxic component of PM and that it inflicts a major burden on public health [43]. For this reason, personal exposure assessment seeking to evaluate the contribution of specific sources to personal PM exposure have sought more specific metrics than $PM_{2.5}$ mass, including chemical speciation of tracers compounds that may indicate the influence of specific sources. PM speciation has traditionally involved

analyses of samples captured on filter media (e.g. inductively-coupled plasma, ICP, followed by optical emission spectrometry, OES, or mass spectrometry, MS). PM speciation is substantially more expensive (often >$50 per sample) than gravimetric analysis (~$15 per sample), often limiting the number of samples taken in a study. Moreover, method limits of detection often require fairly long sampling durations, which limits measurement time resolution. Advances in microfluidic technology allow for low-cost, rapid detection of some PM species. Paper-based devices have employed colorimetric methods to detect trace species (as low as nanogram masses) in collected air samples and biologic fluids [44]. Paper-based sensors have been developed to measure metals from air samples collected on traditional filtration media [45–48]. These paper-based devices are very low-cost (<$1 to produce, compared to ~$100 for a metals analysis by ICP-MS), can use simple devices like cellular phone cameras as color detectors, and have shown good linearity with traditional methods [44]. Pairing these devices with electrochemical detection can further improve method selectivity and sensitivity [44, 48]. The species available for quantification by this method are still limited, but the “lab on a chip” field is progressing rapidly and may present new opportunities for personal exposure assessment.

Black carbon is produced from the incomplete combustion of fossil fuels. In densely populated areas, the contribution from traffic is often considered more important than from other fossil fuel combustion activities, including industry [49]. In studies collecting PM filter samples, the absorbance of the filter can be measured with a transmissometer to evaluate the mass of black carbon (e.g. SootScan, Magee Scientific, Berkeley, CA, USA). This method can provide estimates of the time weighted average exposure to black carbon [50, 51]. However, if the goal is to evaluate exposures during commute times specifically, cumulative measures may not be suitable. A personal aethalometer (MicroAeth, AethLabs, San Francisco, CA, USA) has gained popularity for measuring black carbon at high temporal resolution (up to 1-second resolution) and for use en epidemiologic studies [52–54].

2. Beyond PM Mass

Although it is certain that PM is associated with a variety of adverse health outcomes, it is not known which metric (particle size, morphology or chemical composition) is most strongly associated with heath deterioration [55–57]. The assessment of personal exposures, which are highly dependent on individual activities, represents an opportunity to evaluate the short-term effects of novel pollution metrics that are not routinely monitored for regulatory purposes. As ambient $PM_{2.5}$ mass levels improve, especially in many developed regions of the world, other metrics, such as those discussed below, may provide stronger, more precise associations with health outcomes.

2.1. Ultrafine Particulates

Ultrafine particles (UFP, those with diameter less than ~0.1 μm) contribute nearly negligibly to $PM_{2.5}$ mass, but dominate the particle number concentration (PNC). UFP are known to carry large amounts of adsorbed toxic contaminants such as oxidants, metals and organic species that may produce oxidative stress in the body [58]. Traditionally, monitoring of UFP has relied on condensation particle counters (CPCs) to measure PNC and handheld units

have been used for personal exposure assessment (e.g. P-track, TSI, Inc., Shoreview, MN; [59]). However their use in personal monitoring is limited by the cost, size, weight, and maintenance requirements of this instrument.

In the last five years, substantial progress has been made to assess personal exposure to ultrafine particles (particles smaller than 100 nm). Personal DRIs for ultrafine particles are based on diffusion charging or light scattering after growth by condensation. The DiSCmini (Matter Engineering) is a personal diffusion charging device introduced by Fierz et al. [60]. In the DiSCmini, a positive corona is used to produce a high concentration of positive ions that attach to the particles entering the inlet. The charged particles then pass through an induction stage (or ion filter), a diffusion stage, and a high-efficiency particulate air (HEPA) filter. The diffusion stage and the HEPA filter are each connected to an electrometer, which measures the charge of depositing particles. The smallest particles deposit on the screen in the diffusion stage, whereas larger particles penetrate to the HEPA filter. Particle number concentration, mean diameter, and lung-deposited surface area concentration are estimated using the signals from the electrometers. The DiSCmini compares reasonably well with reference instruments under laboratory [61, 62] and field settings [63]. In urban settings, the DiSCmini was used to show that number concentration is generally inversely related to particle size and strongly influenced by microenvironment, number concentrations are highest near roads, and that HEPA filters in cars can substantially reduce exposures [64]. The DiSCmini has also been used to investigate the relationship among particle exposures and cardiovascular health risk during highway maintenance [65] and to investigate the spatial heterogeneity of ultrafine particles [66, 67].

The nanoTracer PNT1000 (Phillips Areasense) is another DRI based on diffusion charging for measuring personal exposure to ultrafine particles. As described by Marra et al. [68], particles entering the nanoTracer are first charged in by diffusion charging and then enter an electrostatic precipitation section. The charge on particles that pass through the precipitator and deposit onto a HEPA filter is measured with an electrometer. The total particle number concentration and mean particle size are derived from the signals of the electrometer with the electrostatic precipitator turned on and off. The nanoTracer has been compared to other instruments in the laboratory [69, 70]. It has been used to evaluate determinants of ultrafine particle concentrations in homes [71] and to investigate possible associations among ultrafine particle exposures and adverse cardiopulmonary health [72–74].

A personal ultrafine particle monitor (PUFP C100, Enmont LLC) became commercially available in late 2014 as described by Ryan et al. [75]. The PUFP C100 is a CPC that addresses many of the challenges when using CPCs for personal exposure assessment. The C100 draws aerosol through a tubular saturator with walls wetted with water. The temperature of the saturator is increased with distance causing supersaturation of water vapor at the centerline of the tube and condensation of water vapor onto the surface of the particles larger than a critical diameter (~20 nm). These particles grow until they are several micrometers in diameter and scatter a sufficient amount of light to be counted in a detector region. This instrument provides total particle number concentration from ~20 nm to ~2 μm. However, the CPC must have the water reservoir refilled periodically, which may require assistance from participants for sampling durations over 6 hours. Measurements made with

prototypes of this instrument have been shown to be highly correlated with those from benchtop, reference CPCs [76]. This instrument and prototypes have been used to investigate the impact of idling of school busses on ultrafine particle exposures [77, 75] and to evaluate ultrafine particle exposures among schoolchildren [75].

Other devices have been designed to collect ultrafine particles for subsequent analysis by electron microscopy or bulk chemical methods. Chemical and morphological information from electron microscopy can be used to distinguish certain types of nanoparticles apart from other nanoparticles and larger particles in a collected sample [78], although analysis can be expensive (~$300 per sample). Samples collected onto filters can be used for this purpose but require a flat featureless background (polycarbonate filters) and correction for less than 100% collection efficiency [79], which also depends on particle morphology [80]. They also require fairly complicated procedures to eliminate the background filter media for analysis by transmission electron microscopy (TEM), which provides better resolution than scanning electron microscopy (SEM) for particles smaller than 100 nm [78]. Alternatively, personal thermophoretic samplers to collect breathing zone samples over a fairly long (8 hr to 24 hr) time period directly onto TEM grids [81, 82]. These grids can then be analyzed by SEM or TEM without further preparation.

Several samplers have been developed to collect ultrafine particles for characterization by bulk chemistry methods. The Personal Nanoparticle Sampler (PENS) uses three stages (a respirable cyclone, a micro-orifice impactor with a d_{50} = 100 nm, and a filter) to enable measurement of respirable and nanoparticle exposures [83]. The impactor provides a sharp cutoff to collect nanoparticles separately from larger particles, although at a rather high pressure drop 14 kPa. Another sampler, the Nanoparticle Respiratory Deposition (NRD) sampler (Zefon Intl) [84], uses a respirable cyclone (d_{50} = 4 μm), a three-jet impactor (d_{50} = 300 nm), and finally eight nylon mesh screens to collect particles by diffusion. The collection efficiency of the mesh screens combined with that of the impactor mimics the total deposition of particles smaller than 300 nm in the human respiratory system. The reliance of particle collection by diffusion enables particle collection at substantially lower pressure drop than the PENS (3.5 kPa), which is important for personal sampling pumps. The filter from the PENS or the mesh screens from the NRD sampler can be analyzed by various chemical methods (e.g., inductively coupled plasma followed by optical emission spectroscopy, ICP-OES). The PENS sampler has been used for sampling of metalworking operations [85], and the NRD sampler for assessing welding fume exposures [86].

2.2. Oxidative Capacity

Although the exact mechanisms by which PM leads to adverse health outcomes are not entirely clear, exposure to PM has been shown to generate reactive oxygen species (ROS) and produce oxidative stress in cells [87, 88, 3, 89]. Persistent cellular oxidative stress may lead to cellular damage, cell death, and disease [3, 90]. Because a wide variety of species can produce these ROS, measuring these components of PM individually is not practical or cost-effective. Instead, chemical assays such as the dithiothreitol assay have been developed to assess the cumulative effect of these components to produce ROS, known as the aerosol oxidative capacity [91–95]. The dithiothreitol assay typically requires relatively large

masses of PM (5–40 μg per mL [93, 92]), necessitating sampling flow rates and durations longer than typical for personal monitoring. Recent advances in microfluidic technology have reduced the assay volumes such that low PM masses can be evaluated with a paper-based device [96, 97], similarly as described for the detection of metals (Section 2.4) and electrochemical sensors have been developed that can be used to measure oxidative capacity in airborne PM [98] or from extracted filter samples [99]. Both the electrochemical and paper-based devices have potential uses for personal exposure assessment.

3. Integration of Exposure Covariates

Epidemiologic analysis requires the collection of health measures and other important covariates often through questionnaires and exam visits. However, questionnaire data can be unreliable and subject to recall bias, and in most cases, it is unclear when exam visits should be scheduled (immediately after sampling, 8 hours later, 24 hours later). Several new approaches are described here to improve collection of these data.

3.1. Microenvironment and Location

As individuals move through an urban or suburban environment, pollution levels within their breathing zone may change rapidly with location (e.g., major roads versus office space) and time (e.g., rush-hour traffic versus weekend drive). Time-activity diaries are often used to account for participant location, but these diaries are time consuming for participants, and are often incomplete. Alternately, personal exposure assessment using direct-reading sensors can be paired with a global positioning system (GPS) receiver to track participant location [100]. Downloading the time series of participant location into a geographic information system (GIS) with known home, work, and asking participants about other locations visited during their sampling period can provide a more precise estimate of time-activity.

3.2. Activity Level

Although personal monitoring is the state-of-the-art method for exposure assessment, an estimate of inhaled dose cannot be made without knowledge of ventilation rate. According to the environmental health paradigm, inhaled dose should be more related to the health outcome than the exposure. Although it is possible to measure ventilation rate directly, such instruments require participants to wear chest straps that are uncomfortable for most. Ventilation rate is related to heart rate [26], and commercially available heart rate monitors may provide an opportunity to improve estimates of participant dose, particularly for activities like riding a bicycle, where both exposure and ventilation rate may be high. Sophisticated chest-mounted heart rate monitors (e.g. ActiHeart, CamNtech) provide high quality data and can additionally monitor inter-beat interval also providing measurements of heart rate variability, which may be an important health outcome to consider in studies on impacts of PM on cardiovascular health [101]. However, relatively inexpensive heart rate monitors that are wrist mounted may also prove useful to determine inhaled dose.

3.3. Health Data

Several recent studies have deployed home-use spirometers for panel studies of asthma and COPD patients [102–104]. These studies showed that data-logging units prevented

transcription errors by participants, improved compliance, and provided data that compared well to clinic spirometers. Ambulatory heart rate monitors have also been used in epidemiologic studies of air pollution, but are bulky and uncomfortable for many participants. Small, wearable sensors are marketed to elite athletes, but have clear usefulness for personal monitoring. Commercial sensors for cardiac rhythm are now available (e.g. ZioPatch, iRhythm Technologies and other sensors that can monitor blood chemistry are under development [105]). Additionally, sensors to track outcomes among susceptible populations may improve our understanding of how particulate air pollution contributes to disease. For example, units adapted to fit on an inhaler can track usage and the location of use of rescue medication for individuals with asthma or chronic obstructive pulmonary disease (Doser, Meditrack Products; Propeller, Propeller Health).

4. Outstanding Challenges and Opportunities in Air Pollution Exposure Assessment

A major limitation of personal exposure assessment is determining whether the participant has worn sampling equipment consistently (participant compliance). If sampling equipment is left in a home unattended, it does no better at reducing exposure misclassification than an area sample. Accelerometers on sampling equipment can be used to determine how long it remained stationary. To characterize the health effects associated with short-term (<2 hour) exposures, it will likely be necessary to have a better awareness of participant compliance. Other methodologies, such as proximity sensors that estimate the distance between the participant and the sampling equipment may be better suited to reach these goals.

We have outlined a framework in Figure 1 by which sensor data on personal exposures, microenvironmental data, and health data can be collected simultaneously and integrated with a central server to enable high level processing. With rapidly advancing sensor technology, it is crucial that methodology is developed to use the data appropriately. Ramachandran and colleagues [106, 107] showed that 15-minute average ambient air pollutant concentrations were routinely 3–4 times higher than the 24-hour average outdoor values, could vary by as much as an order of magnitude, and that within-day variability was comparable to between-day variability. Variability in personal exposures will likely be even larger than ambient levels because participants are actively involved with PM generating processes (cooking, cleaning, driving etc.). However, this data will also be correlated over time, complicating statistical analyses. DRIs can often provide high temporal resolution data, but most studies have not utilized the high temporal resolution data, instead simply averaging over longer time frames based on some classification (hourly, by microenvironment, etc.). However, these data have potential to help elucidate the appropriate time frame from exposure to health outcome. In the lower right panel of Figure 1 we illustrate how floating exposure and health outcome windows can be modeled to define the most relevant lag times for health effects. Wellenius et al. [6] found that ischemic stroke risk was most strongly associated with markers of traffic-related pollution (hourly $PM_{2.5}$ mass, black carbon, nitrogen dioxide concentrations) with the highest odds ratios occurring 12–14 hours before stroke onset, suggesting that short-term exposures may increase risk. Significant associations were not observed for sulfate, ozone, or carbon monoxide.

Additionally, Delfino et al. [103] found stronger associations between FEV1 and $PM_{2.5}$ mass when 1-hr and 8-hr maximum values were used, compared to 24-hour averages among asthmatic children. Developing data handling and statistical methodology will be crucial to use this high-resolution data appropriately while properly accounting for measurement error, correlation and confounding [108, 109].

The combination of location and exposure data allows researchers to apportion exposures to the various microenvironments in which people spend time and may help identify sources most strongly associated with health (Figure 1, lower left panel). For example, when a person is at home the PM from vacuuming or cooking may not be associated with health outcomes to the same extent as traffic-related particulate air pollution that is enriched in black carbon, metals, and polycyclic aromatic hydrocarbons when a person is in transit [43]. Such results may have important policy implications. The persistent observation of health effects at PM concentrations below regulatory standards suggests that there may not be a threshold level for which the effects of PM on health are not observed [110]. The lack of a threshold complicates policy decisions because reductions in PM mass concentrations will ultimately be limited by background concentrations from natural sources. Currently, regulations are based on central monitoring of mass concentration. Mass-only measurements are inherently difficult to use to identify the most toxic particles from the highly variable mixture encountered in daily life. Epidemiologic studies that employ personal exposure assessment may provide needed information on the associations of specific components of PM and health. Such monitoring, including both novel metrics of air pollution and acute health information, could provide a basis for evaluating variable toxicity of different sources of PM or components of PM. This information may allow individuals, particularly those most susceptible to the adverse effects of air pollution, to make choices to reduce their exposures to the most toxic components of PM. Ultimately, the information may contribute to new regulations or guidance honed to those specific sources of PM or components of PM (e.g. chemical composition, size, shape) most strongly associated with adverse health.

Other challenges in monitoring airborne particle exposures in different microenvironments remain. For example, due to differences in sampling approaches for PM in occupational and ambient environments (e.g. respirable mass for occupational exposure assessment vs. $PM_{2.5}$ mass for ambient exposure assessment) and differing mandates from distinct funding agencies (EPA and NIEHS vs. NIOSH), few studies have considered exposure assessments in both ambient and occupational environments. This distinction leaves researchers unable to consider a holistic view of PM exposure for individuals. Other challenges include agreement from participants' employers to allow monitoring equipment in the workplace. However, low cost and lightweight monitoring instruments can improve the feasibility of studies to cross these domains.

Finally, we wish to identify a few opportunities for integration of new sensor technology for exposure and health data between researchers, participants, and other stakeholders (Figure 1, lower center panel). High temporal resolution data could be wirelessly transmitted to participant computers or cell phones, allowing them on-demand information of their exposures and enabling them to view how personal activities influence their personal exposures. In occupational environments, such sensors could trigger alarms for health and

safety professionals to let them know when exposure thresholds are exceeded. Health and exposure data could be sent to parents of susceptible children or to health professionals of susceptible populations of children and adults (e.g. asthmatics, those with cardiovascular disease) to allow immediate intervention when needed to minimize or prevent exacerbation of disease.

5. Conclusions

Personal monitoring for particulate air pollution was pioneered in the 1960s, but significant advances in pump, sensor, and battery technology have improved the reliability of these sampling methods and improved feasibility for large-scale personal exposure assessment. This review has outlined personal exposure assessment approaches focusing on novel sensors developed over the last five years. Although size-selective sampling remains the most common method for measuring personal exposures to particulates, novel sensors that go beyond measuring PM mass provide alternate strategies for exposure assessment and may yield stronger, more precise associations with adverse health outcomes accounting for the variability introduced by the toxicity of sources. For example, particle number concentration is dominated by the smallest particles (<200 nm), those which contribute nearly negligibly to particle mass concentration. Until recently, particle number concentration was difficult to measure without the aid of heavy and expensive equipment, but personal monitors employing at least five measurement methods have been developed over the last 5–10 years. By pairing traditional or novel exposure measures with measures of health, activity, and important microenvironmental factors, we anticipate that information bias can be reduced compared to questionnaire-based approaches and central monitoring. More importantly, a holistic view of the influence of particulate air pollution on health may emerge.

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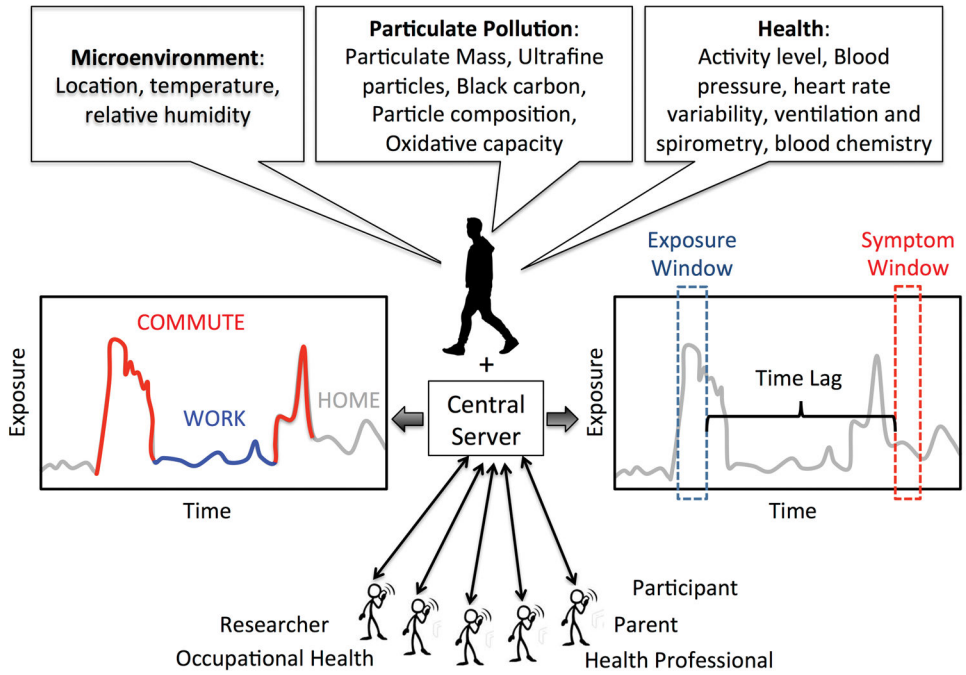

Figure 1.
Framework for personal exposure assessment and usage of highly resolved temporal data.

Table 1

Recent developments in personal exposure assessment for particulate matter.

Instrument	Operating Principle	Advantages	Disadvantages	Key Citations
Mass-based PM				
PHISH	Size-selective sampling inlet provides inhalable sample at 10 L min^{-1}.	Higher flow rate to achieve lower detection limits; low cost compared to traditional inhalable samplers.	High flow rate limits options of personal sampling pumps.	Characterization: [16]
Personal Cascade Impactors	Provides low resolution size distribution using inline impactors.	Particles collected on stages for gravimetric or chemical analysis by particle size.	High pressure drop requires expensive pump; analyses by particle size adds cost.	Characterization: [20–23].
PM Speciation (paper-based)	Reactions with metallic components of PM produce color change on device.	Samples collected for later analysis; Very low cost; Detect many species; Portable; Easy to perform test.	Low temporal resolution; Few PM species included in current devices.	Characterization: [46, 48, 111, 45]
Foam-based Samplers	Foam serves as porous filter capturing fraction of particles relevant to physiological dose.	Estimates particulate dose; Low pressure drop.	Low temporal resolution; Not amenable to gravimetric analysis.	Characterization: [37, 38, 112]
Light Scattering Instruments				
MicroPEM	Light-scattering detects particles larger than ~300 nm.	Lightweight internal pump; High temporal resolution.	Relatively expensive.	Characterization: [26]
Shinyei PPD42NS/ Sharp GP2Y1010AU0F	Light-scattering detects particles larger than ~500 nm.	Very low cost; No sampling pump; High temporal resolution.	Not a size-selective sampler; user must outfit sensor with data logging capabilities.	Characterization: [31, 32]
Dylos	Light-scattering detects particles larger than ~500 nm.	Low cost; Internal fan used to provide air flow; High temporal resolution	High particle diameter limit of detection for a particle counter	Characterization: [33] Application in recent studies: [34–36]
UFP				
Diffusion Classifier (DiscMini, Matter Aerosol, Switzerland; NanoTracer, Philips Aerasense, Netherlands)	Particles charged and drawn to a electrometer that converts signals to particle surface area, number concentration, and mean diameter.	Internal pump; High temporal resolution.	Relatively heavy and expensive.	Characterization: [60–63, 68–70] Application in recent studies: [65–67, 64, 72–74]
NRD	Three stages: a respirable cyclone, three jet impactor, and mesh screens provide an estimate particle deposition in the respiratory tract for diameter <300nm.	Mesh screens amenable to various chemical analyses methods.	Low temporal resolution.	Characterization: [84] Application in recent studies: [86]
PENS	Three stages: a respirable cyclone, a micro-orifice impactor and a filter give measures of respirable and nanoparticle exposures.	Measure nanoparticles (<100 nm) separately from larger particles.	High pressure drop requires expensive sampling pump; Low temporal resolution.	Characterization: [83] Application in recent studies: [85]

Instrument	Operating Principle	Advantages	Disadvantages	Key Citations
MiniCPCs	Particles are grown by condensation of water vapor to a size that can scatter enough light to be detected and counted.	Detects particles as small as 20 nm; High temporal resolution.	Water reservoir must be refilled; current version has short battery life (<6 hours).	Characterization: [75, 76] Application in recent studies: [77, 75]
Thermal Precipitator	A large temperature gradient is developed between two parallel plates. Increased energy of molecules on warm side drives particles to a collection substrate.	Efficient for particles less than ~300 nm; Electron microscopy paired with EDS can provide information on size, shape, and composition of particles.	Low temporal resolution; Microscopy methods are required for particle detection, which can be expensive.	Characterization: [81, 82]
Black Carbon				
MicroAeth (AE51, AethLabs, San Francisco, CA)	Aethalometer detects changes in filter transmissivity.	Built in pump; High temporal resolution; Low LOD.	Low built-in battery life; Variability in response with aerosol optical properties.	Characterization: [113–116] Application in recent studies: [52–54].
Filter-based reflectance/absorbance (e.g. SootScan, Magee Sci, Berkeley, CA)	Detects changes in filter transmissivity/ Absorbance.	Samples can be collected for later analysis; Samples stable for long period of time; Multiple wavelengths (880 nm sensitive to BC, 370 nm sensitive to aromatic OC).	Low temporal resolution.	Characterization: [117] Application in recent studies: [51, 50]
Oxidative Capacity				
Paper-based sensors	Reactive species of PM on filter cause color change on device via DTT assay.	Samples can be collected for later analysis; Very low cost; Can detect oxidation from many species; Portable; Easy to perform test.	Low temporal resolution; Uncertainty regarding physiological significance.	Characterization: [97, 96]