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Authors
Fischer, Susan L
Koshland, Catherine P.

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Field performance of a nephelometer in rural kitchens: effects of high humidity excursions and correlations to gravimetric analyses

Susan L. Fischer*, Catherine P. Koshland
Environmental Health Sciences, School of Public Health
University of California, Berkeley
Berkeley, CA 94720

*corresponding author
sfischer@berkeley.edu, 140 Warren Hall #7360, tel. 510-643-4613, FAX: 510-642-5815

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Abstract

Rural kitchens of solid-fuel burning households constitute the microenvironment responsible for the majority of human exposures to health-damaging air pollutants, particularly respirable particles and carbon monoxide. Portable nephelometers facilitate cheaper, more precise, time-resolved characterization of particles in rural homes than are attainable by gravitational methods alone. However, field performance of nephelometers must contend with aerosols that are highly variable in terms of chemical content, size, and relative humidity. Previous field validations of nephelometer performance in residential settings explore relatively low particle concentrations, with the vast majority of 24-hour average gravitational $PM_{2.5}$ *concentrations falling below 40* $\mu g/m^3$*. We investigate relationships between 24-hour gravitational particle measurements and nephelometric data logged by the personalDataRAM in highly polluted rural Chinese kitchens, where gravitationally determined 24-hour average respirable particle concentrations were as high as 700* $\mu g/m^3$*. We find that where relative humidity remained below 95%, nephelometric response was strongly linear despite complex mixtures of aerosols and variable ambient conditions. Where 95% relative humidity was exceeded for even a brief duration, nephelometrically determined 24-hour mean particle concentrations were nonsystematically distorted* *relative to gravitational data, and neither concurrent relative humidity measurements nor use of robust statistical measures of central tendency offered means of correction. This nonsystematic distortion is particularly problematic for rural exposure assessment studies, which emphasize upper quantiles of time-resolved particle measurements within 24-hour samples. Precise, accurate interpretation of nephelometrically resolved short-term particle concentrations requires calibration based on short-term gravitational sampling.*

Introduction

Portable nephelometers have been welcomed as an advancement for assessment of human exposures to particles (Balakrishnan *et al.*, 2004; Ezzati *et al.*, 2000a; Ezzati *et al.*, 2000b; Lanki *et al.*, 2002; Liu *et al.*, 2003; Muraleedharan & Radojevic, 2000; Quintana *et al.*, 2000; Quintana *et al.*, 2001; Radojevic & Hassan, 1999; Rea *et al.*, 2001; Wallace *et al*., 2006; Wallace *et al.*, 2003; Williams *et al.*, 2000; Wu *et al.*, 2005a). However, most previously published environmental investigations of nephelometer performance relative to gravitational determinations depict controlled laboratory settings or relatively low (1 – 60 μg/m^3) particle concentrations (Allen *et al.*, 2004; Chakrabarti *et al.*, 2004; Howard-Reed *et al.*, 2000; Jenkins *et al.*, 2004; Lanki *et al.*, 2002; Liu *et al.*, 2002; Quintana *et al.*, 2000; Richards *et al.*, 1999; Sioutas *et al.*, 2000; Wallace *et al*., 2006; Wallace *et al.*, 2003; Wu *et al.*, 2005b). Our research explores the performance of a commercially available portable nephelometer, the MIE personal DataRAM (pDR-1000, Thermo Electron, Franklin, MA, USA), in a highly polluted field context. We monitored indoor particle concentrations in rural Chinese kitchens for which 24-hour average gravitational respirable particle (RSP) measurements ranged 28 μg/m^3 to 696 μg/m^3, with a mean(se) of 202(40) μg/m^3. This research extends validation of nephelometer performance to a multi-source, highly variable pollution regime that is extremely important from the standpoint of global human exposures.

Indoor air quality in rural kitchens: a public health priority

To understand the importance of characterizing air pollution in rural kitchens in which small-scale combustion of solid fuels routinely transpires, consider that inhalation intake per unit emission is two to three orders of magnitude higher for indoor combustion sources than for outdoor sources (Evans *et al.*, 2002; Lai *et al.*, 2000; Smith, 1988; Wallace, 1996). Compounding the problem of greater intake for indoor sources is that solid fuel/stove combinations are two to three orders of magnitude more polluting than their gas counterparts: for example, a study of emissions factors from 28 Chinese fuel-stove combinations indicates that solid fuels typically emit 10-100 g products of incomplete combustion (PICs) per MJ of delivered energy, whereas Chinese stoves burning liquid petroleum gas (LPG), coal gas, or natural gas emit less than 0.03 g-PIC/MJ (Zhang *et al.*, 2000). Another aggravating factor is the prevalence of associated exposures: an estimated 1.06 billion people relied partially or exclusively on solid fuels for cooking or heating in China alone in 2001 (NBS, 2002).

Need for time-resolved data: rural kitchens and general exposure assessment

Time-resolved particle concentration data from rural households is desirable as a means of indicating concentrations and exposures associated with high pollution periods and illuminating the distribution of exposures across gender and age. Additionally, more than two dozen human exposure studies (Michaels & Kleinman, 2000) and several animal studies offer evidence for PM-mediated health effects at time scales shorter than 24 hours. For example, acute pulmonary inflammation (latency period 6 hr) has been observed in response to 1-hour exposures to diesel exhaust (Salvi *et al.*, 1999), and episodes of asthma have been found to correlate more strongly to 1-hour and 8-hour PM_{10} concentrations than to 24-hour means (Delfino *et al.*, 1998; Delfino *et al.*, 2002). Cardiovascular effects, such as reduced heart rate variability, of short-term human exposures to fine particles have also been investigated (Gold *et al.*, 2000; Le Tertre *et al.*, 2002; Sullivan *et al.*, 2005). Resolving epidemiologic and dose-response relationships calls for finer temporal resolution of exposures.

Challenges to time-resolving field devices for particle monitoring

Indoor aerosols in environments frequented by human beings are often chemically and physically unstable and extremely heterogeneous, and describe different (between microenvironments) and temporally variable (within a single microenvironment) relationships between mass concentration determined by scattering and mass concentration as determined by standard gravimetric methods. These differences are especially acute in rural solid fuel-burning kitchens where relative humidity is variable, some level of tobacco smoking is typical, and more than one fuel/stove combination is often used during a given monitoring period.

Scope of this paper

While the use of a portable nephelometer for indoor air quality assessments in rural kitchens holds promise for high-precision, time-resolved measurements, there remain critical issues to resolve regarding signal interpretation. Published research results (reviewed below) based on nephelometric measurements in rural field settings are inconsistent with regard to data quality (i.e., nominal (factory-calibrated) readings vs. data scaled to fit gravitational field samples, particle size fraction of gravimetric calibration) and treatment of temporal resolution (Balakrishnan *et al.*, 2004; Brauer, 1995; Brauer *et al.*, 1996; Ezzati *et al.*, 2000a; Ezzati *et al.*, 2000b). Several studies have observed humidity-dependence of light scattering signals in ambient environments (e.g., Day & Malm, 2001; Day *et al.*, 2000; Richards *et al.*, 1999; Thomas & Gebhart, 1994). However, previously published field research using nephelometers in solid fuel-using rural households does not explicitly contend with high relative humidity excursions.

After reviewing previous work germane to this investigation, we present results from concurrent optical and gravitational sampling in a rural village in northeastern China. Our particle data are augmented by time-resolved logs of temperature and relative humidity, as well as by surveys of household structural features and pollution-related behaviors. The kitchens in which sampling transpired represent a wide range of fuel/stove types, cooking and heating patterns, and tobacco-smoking behaviors. We ask of our data: what is the relationship between 24-hour gravitational and optical samples? How do short-term high-humidity excursions affect correlations between 24-hour gravitational and optical samples? What are the implications for rural indoor air quality and exposure assessment studies?

Previous work characterizing pDR response relative to gravimetric measurements

A number of laboratory and field investigations have characterized correlations between gravimetric particle samples and nominal (factory calibration based on SAE Fine test dust[1]) data logged by the MIE personalDataRAM, henceforth pDR. Compiled in Table 1, these studies are briefly reviewed below.

Laboratory- and field-based characterization of humidity effects on pDR response

Two independent laboratory investigations observe that at relative humidity (RH) greater than 70% the pDR substantially and nonlinearly overestimates mass concentration (Chakrabarti *et al.*, 2004; Sioutas *et al.*, 2000). For a well-characterized particle distribution, Chakrabarti *et al.* and Sioutas *et al.* provide empirical evidence of pDR adherence to theoretical trend regarding sensitivity of scattering signal to relative humidity. However, strong dependence of humidity-induced distortion of the optical signal on particle size and composition precludes applying a theoretically-derived humidity correction where the particles of interest are not physically and chemically well-characterized (McMurry & Stolzenburg, 1989), such as rural kitchens in which several fuels as well as tobacco are burned, humidity conditions can fall below 30% and often peak above 90%, and diurnal particle concentrations can range from less than 50 $\mu g/m^3$ to several *thousand* $\mu g/m^3$.

In low-pollution field environments in which particles were not chemically and physically well-characterized, two investigations with the pDR report successful statistically-based correction for relative humidity effects on the relationship between nephelometric signal and gravimetric analysis (Richards *et al.*, 1999; Wu *et al.*, 2005b). Above 95% relative humidity, the upper limit of manufacturer's specifications for pDR operation, the scattering signal's dependence on relative humidity is too sensitive to enable empirical correction, given the precision and accuracy limitations of portable relative humidity data-logging devices (Richards *et al.*, 1999).

Corrrelations between pDR and gravimetric measurements in controlled settings

Jenkins *et al.* (2004) observe strongly linear relationships (r^2 =0.98-9.999) between pDR response and 24-hour gravimetric determination of respirable particles (RSP) in three laboratory simulations of environmental tobacco smoke (ETS), cooking oil fumes, and cedar wood smoke.

Correlations between pDR and gravimetric measurements in field settings

Several researchers have explored correlations between the pDR's nephelometric signal and gravitationally determined mass concentration in ambient environmental, indoor, and personal exposure settings (Allen *et al.*, 2004; Chakrabarti *et al.*, 2004; Howard-Reed *et al.*, 2000; Lanki *et al.*, 2002; Liu *et al.*, 2002; Quintana *et al.*, 2000; Sioutas *et al.*, 2000; Wallace *et al.*, 2006; Wu *et al.*, 2005b). These studies demonstrate: (a) significant linear relationships of varying strength between scattering coefficients (nominal nephelometric signal) and mass concentrations in field environments, with stronger relationships obtaining in those settings for which relative humidity and volatile fraction are reasonably stable; (b) that the pDR must be field-calibrated for each distinct microenvironment in which it is used, due to situation-specific ratios of optical signal to gravimetric concentration; (c) where there exist several different sources and humidity regimes, calibration based on integrated (24hr) measurements may not apply to short-term pollution episodes, particularly those associated with cooking. In particular, Liu *et al.*'s (2002) study of residential indoor air quality found significant ($p<0.0001$) disparity between short-term particle concentrations (scaled to 24-hour gravitational measurements) reported by pDR and Radiance M903 (Radiance Research, Seattle, WA; scattering at 530 nm) nephelometers.

Previous use of nephelometers for temporal disaggregation in rural field settings

Three previously published studies of indoor air quality in rural households use portable, passive-mode nephelometers to afford some level of temporal disaggregation not available from filter samples alone (Balakrishnan *et al.*, 2004; Brauer *et al.*, 1996; Ezzati *et al.*, 2000a; Ezzati *et al.*, 2000b). Brauer *et al*. infer from short-term measurements with the Radiance M903 nephelometer that peak particle concentrations in unvented biomass-burning kitchens are significantly higher ($p<0.05$) than LPG-burning and improved-stove counterparts, without quantifying the disparity. Using time-resolved particle data in kitchens, the other two studies demonstrate methodology for quantitatively characterizing exposures of cooks in India (Balakrishnan *et al.*, 2004) and Kenya (Ezzati *et al.*, 2000b).

Methods

Site

Field measurements represent wintertime kitchens of a rural village in China's Jilin province, which borders Inner Mongolia Autonomous Region to the west and North Korea and Russia to the southeast. Homes in Hechengli village (pop. 720) are small (typically 56 m^2), single-story dwellings with uninsulated exterior walls of three-layer brick and concrete (2.5 cm thick) or earthen floors. Winters are cold, with soil freezing to a depth of 1.7 m and ambient temperatures routinely dipping below -20 °C.

Space heating, as well as some cooking, in Hechengli is primarily accomplished by the use of *kang*s. The traditional Han-style *kang* is an internal flue structure made from fired bricks. The internal flue circulates hot exhaust beneath a raised floor. After circulating under the floor of the living space, hot gas exits the *kang* via a brick chimney 5-6 m high. *Kang* stove structures in Hechengli are fairly uniform in design insofar as they all exhaust through chimneys, have doors to close the solid fuel combustion chamber, and are sealed with respect to the combustion chamber and cooking vessels (Figure 1).

In addition to solid-fuel burning *kang*s, a variety of other combustion devices are found in Hechengli village households: free-standing solid fuel stoves for boiling water and/or augmenting space heating, LPG stoves, and producer gas[2] stoves. Cigarette-smoking also plays a significant role in generating indoor air pollution.

Sample

During February-March 2003 and February-March 2004, a total of 37 distinct households were monitored; some households were monitored more than once (not more than twice in a given field period), for a total of 70 household-days of sampling. The sample represents the full range of fuel/stove types, cooking styles, smoking intensities, and heating practices observed in Hechengli village's 224 households. The sampling period was 24 hours. Optical time-resolved particle data were logged for 65 samples. Gravitational sampling of respirable particles was undertaken for 23 household-days. Two samples were excluded from analysis due to power failure of the nelphelometer; thus, 21 pairs of optical and gravitational particle measurements were available from village kitchens.

To help delineate the extent to which different sources yield different nephelometric response, several additional opportunistic samples were collected from indoor environments in which a single source dominated: heavy tobacco smoking (approximately 40 cigarettes smoked in a 40 m^3 room over an 8-hour period), n=4; laboratory-simulated coal-burning fire pit (Tian, 2005), n=2; solid-fuel (coal) burning stove used for heating water, n=2; and non-smoking room in a Chinese hotel, n=4. Monitoring periods were 24 h r for these samples except where a specific activity of shorter than 24-hour duration was of interest, namely laboratory simulation of a coal-burning fire pit (1.5 h r) and rooms in which heavy tobacco smoking occurred (8 hr). Two of these samples (both were heavy smoking events) were excluded from analysis due to human tampering with equipment.

Optical particle measurements

Time-resolved particle measurements were conducted with the MIE personalData RAM operating in passive mode. The personalDataRAM (pDR) is a nephelometer (light-scattering device) that measures particle concentration on the basis of scattering from a pulsed, high-output near-infrared LED at 880 nm between 45º and 90º. The geometry and optics of the passive-mode pDR are such that particles sized 0.1-10 μm are emphasized, with maximum response at 0.5-2 μm (MIE, 1990; Wallace *et al*, 2006). The pDR provides temporal resolution as fine as 1 second and has a dynamic operating range of 0.001- 400,000 $\mu g/m^3$ based on a scattering coefficient range of 1.5×10^{-6}- 0.6 m^{-1} and calibration on SAE Fine test dust.

Two pDR's were cleaned using pressurized air before each field mission. They were simultaneously zeroed in a manufacturer-supplied bag that was emptied of air and then positively pressurized using a hand pump with a Grade BQ filter tube. Every 6 or 7 days, pDR's were retrieved from the field in order to download data and perform quality assurance and control, which comprised checking that zeroes had not positively drifted during the field session by placing the pDR's in the zero bags and noting readings to be no more than 2 $\mu g/m^3$ after three air changes of Grade BQ HEPA-filtered air, checking that pDR's gave comparable responses (within 3 $\mu g/m^3$ over a 15-minute period) when placed side-by-side in the same indoor microenvironment, and re-zeroing the instruments. This weekly quality control is compliant with the manufacturer's suggestion that for an average exposure of 1 mg/m^3 PM_{10}, equipment should be re-zeroed once every 2 weeks.

Gravitational particle measurements

A NIOSH-compatible 10 mm, multiple-inlet, conductive plastic cyclone (GS-3, SKC, Inc., cat no. 225-100) was used in conjunction with a battery-operated programmable occupational sampling pump (aircheck sampler, SKC, Inc., model 224-PCXR8) to collect the respirable particle fraction[3] (RSP) for gravitational analysis.

To reduce sensitivity of gravitational analysis to filter conditioning and to save labor, matched-weight 37 mm, 5 µm pore size PVC filters (SKC, Inc. cat. no. 225-8202) were chosen. Each matched-weight cassette comprises two filters matched (within 25 µg) in weight and loaded into a cassette in a controlled laboratory environment (68±0.5 ºF, 50±0.5 % RH); sample weight is taken as the difference between the mass of the top (exposed) and bottom (unexposed) filter.

Pump flow rates were set to 2.75 l/min. prior to field deployment using a NIST-traceable SKC UltraFlo Calibrator (cat. no. 709), a bubble-meter the accuracy and repeatability of which is rated to ±0.5%. Pumps were programmed to run for 8 hr distributed (1 min. on, 2 min. off) over the course of a 24-hour sampling period. Post-sample flow rates were measured with the exposed filter in place, after which filter samples were removed from the cyclone assembly and sealed to await gravitational analysis in Berkeley, California.

Filters were weighed on a Cahn Microbalance (Model 29, Cahn Instruments, Madison, Wisconsin), 0.001 mg resolution, in a controlled-environment weighing room. Prior to weighing, filters were equilibrated for 24 hr at 40±5% RH and 75±5 ºF. To control static effects, each filter was placed on an ionizing anti-static unit for 2 minutes prior to weighing. Quality control included weighing the calibration weight and/or a control filter after every fifth sample was weighed, and recalibrating the balance for deviations between 1 and 5 µg or recalibrating and re-weighing in the case of deviations greater than 5 µg. Under this protocol, the limit of detection was 18 µg/m^3. Gravimetric readings were adjusted for the non-zero mean (-7.42(2.83) µg) of twelve blanks.

Temperature and relative humidity

Time-resolved measurements of temperature and relative humidity were logged by HOBO-T/RH devices (Onset Computer Corporation, Pocasset, MA). Field logistics and the limited memory capacity of the HOBO-T/RH necessitated logging at 3-min. intervals, which did not sacrifice information given the temporal resolution (10 min.) of the RH sensor. HOBO-T/RH units offer NIST-traceable temperature accuracy of ±0.7ºC at 21ºC, with 0.4ºC resolution over a range of -20 –70ºC. The relative humidity sensor is rated to ±5% over a range of 25 –95% RH at 5–50ºC.

Results and discussion

Over the course of fieldwork, the mean(se) of 24-hour gravitational RSP samples (n=21) in Hechengli village kitchens was 202(40) $\mu g/m^3$, with a range of 28.3 $\mu g/m^3$ to 696 $\mu g/m^3$. On a three-minute basis, relative humidity ranged 16.2-100%, and within-sample peak relative humidities spanned 59-100%, averaging 85.1(2.4)%.

Variable optical response ratios between microenvironments

Ratios of nominal optical to gravimetrically determined mass concentrations, disaggregated by microenvironment, are shown in Table 2. Characterizations of response ratio for two atypical microenvironments, namely heavy cigarette smoking as the sole indoor pollution source (n=2) and kitchens in which relative humidity concentrations above 95% were logged (n=5) are limited to small numbers. Nevertheless, statistically significant differences in mean relative response are demonstrated by the three microenvironments most relevant to this study, namely:

1. kitchens in which RH does not exceed 95% (n=16, GM(GSD) of response ratio=2.13(1.47)),
2. kitchens with high-humidity excursions (n=5, GM(GSD) of response ratio=7.40(4.66))
3. heavy smoking indoors (n=2, GM(GSD) of response ratio=27.13(1.55)).

Direct observation of room conditions and human behavior, as well as observed lack of well-mixedness (quantitatively indicated by disparities in highly unstable (in time and magnitude) peak concentrations logged by collocated pDR's) indicate that the two heavy smoking samples represent not uniform ETS but include dense smoke plumes from cigarettes of curious persons who scrutinized equipment at close range. Wisps of mainstream smoke in which condensation has occurred, in the absence of high RH (mean(se)=42(0.2)%, range 29-59%) account for several 1-minute particle concentration peaks for which the nominal pDR reading exceeded 100 mg/m^3. These field observations are in accord with Jenkins *et al.*'s suggestion that poorly-mixed tobacco smoke may constitute a nephelometric regime distinct from well-mixed ETS. Samples plagued by condensation events remain significantly different from other sample sub-groups (see note, Table 2) when high-humidity kitchens and heavy smoking are grouped together.

As cooking episodes may be associated with high relative humidity excursions or dense plumes of condensed-phase species, our observations as well as previous works (see Table 1) call into question two assumptions implicit in previous use of the pDR to reconstruct human exposures to PM in rural Indian kitchens (Balakrishnan *et al.*, 2004). First is the assumption that the relationship between gravimetric $PM_{3.5}$ and optically delineated PM is constant in time within a given household, or, more simply, that nephelometric response per unit particle mass is constant during cooking and non-cooking episodes. A second questionable assumption is that the ratio of cooking to non-cooking particle concentrations is constant across fuel/stove types.

Ratios of nominal pDR response to gravitational samples range from 0.6 to 2.3 in previously published sources (Table 1) and 0.78 to 7.4 in 24-hour kitchen measurements of our research (Table 2). Thus, previously reported 14-hour mean particle concentrations in Kenyan households (Ezzati *et al.*, 2000a; Ezzati *et al.*, 2000b), based on nominal pDR data, may substantially deviate from gravitationally-determined particle concentrations. Nonuniform deviation from gravitational measures is particularly likely for those measurements made in the near-zone of (less than 1m from) combustion sources, where well-mixedness does not always prevail. While the near-zone may be more relevant from the standpoint of assessing cooks' exposures to air pollution, it is also where condensed species are more likely to produce spurious—in the sense of biasing nominal response upward and not being systematically interpretable as mass concentration—pDR data.

Insignificant correlation between gravitational and optical means, full dataset

Analysis of 21samples from kitchen microenvironments does not support a strong or significant linear relationship between the optical and gravitationally determined mass concentrations (r^2=0.11, Prob>F =0.13).

Strong correlations between nephelometric and gravitational measurements where RH <95%

Omitting those samples for which relative humidity was logged in excess of 95% (n=5) at some point during the sampling period, the remaining (n=16) samples describe a highly significant and strong linear relationship (r^2=0.92, Prob>F < 0.0001) depicted in Figure 2, with nominal pDR data overestimating gravimetric RSP by a factor of 2.1(0.17). Omitting the outlier at 644 $\mu g/m^3$ neither significantly alters the relationship between gravimetric and nephelometric measurements nor diminishes its significance, but does reduce explanatory power (r^2=0.82).

The observed relationship (r^2=0.92) between gravimetric and nephelometric measures of PM in kitchens for which RH nowhere exceeded 95% is among the strongest observed (compare to Table 1), notwithstanding the diversity of fuels, cooking and heating practices, smoking behaviors, and ambient conditions. As long as relative humidity does not exceed 95%, the strong signal-to-noise ratio in high-pollution environments of these rural village kitchens largely compensates for issues which compromise the precision and accuracy of nephelometric methods.

No correlation between nephelometric and gravitational measurements if RH >95%

When the five samples for which relative humidity exceeded 95% are examined, no significant linear relationship to gravitational RSP is found (r^2=0.23, Prob>F = 0.42). The temporal duration for which relative humidity in excess of 95% was logged (ranging 27 to 485 minutes) was a strong and borderline significant predictor of nominal mass concentration reported by the pDR (r^2=0.71, Prob>F =0.074).

Within-sample censoring of particle concentrations (1-min. resolution) logged during high-humidity episodes from 24-hour average mass concentrations computed from pDR data did not significantly improve their correlation to gravimetrically determined RSP. In part the failure of this simple censorship is due to the response time of the relative humidity logger, which lags that of the nephelometer by a variable duration (typically 10-15 minutes).

Sensitivity of measures of central tendency to censorship of condensation-distorted samples

Censoring the optical PM dataset to remove samples plagued by condensation events significantly affects short-term peak measures of PM as well as 24-hour averages for summary statistics constructed both as arithmetic and geometric means (Table 3). For these data, reporting geometric means does not confer immunity to distortion from condensation events. Similarly, ranges of observed measurements of PM are also dramatically different between censored and uncensored datasets (Table 4). On 24-hour and 1-hour approximate[4] peak bases, upper bounds of observed PM ranges were overestimated by factors of 2 and 70, respectively, when the dataset was not purged of 24-hour samples suffering short-term condensation-induced optical distortion.

The within-sample median (constructed for each 24-hour sample from concentrations logged at 1-minute intervals) is robust to relative humidity-induced distortion. Thus, the most robust measure of central tendency—the within-sample time-resolved median concentration—reported by previous use of a pDR in rural Kenyan homes (Ezzati *et al.*, 2000a; Ezzati *et al.*, 2000b) may not be afflicted by distortion induced by condensation in the optical chamber. Unfortunately, the within-sample median defies ready interpretation in terms of traditional gravimetric measurements due to the highly skewed nature of such environmental concentrations, and nephelometrically determined mean particle concentrations may be severely and nonsystematically distorted by short-term scattering peaks induced by high-humidity excursions or condensation in the optical chamber (Table 3).

Ezzati *et al*. (2000b) also raise the question of whether magnitudes of short-term peak exposures, rather than average daily exposures, drive adverse respiratory and ocular health endpoints. However, as indicated by the research reviewed above and by our findings, short-term nephelometric peaks cannot be conclusively interpreted on the basis of nominal readings or as scaled according to correlation with longer-term gravimetric samples. Our data demonstrate that metrics based on upper quantiles of temporally resolved particle concentrations are particularly sensitive to nephelometric distortion induced by condensed phase species (Table 4).

Conclusion

Under conditions for which humidity is highly variable but nowhere in excess of 95%, 24-hour average particle concentrations registered by a portable nephelometer are highly correlated (r^2=0.92, n=16) with gravitational respirable particle (RSP) measurements in Hechengli village, despite multiple fuel use, highly variable and sometimes intense tobacco smoking patterns, different cooking and heating stoves, and variable combustion-related behaviors. On a 24-hour basis, we observe nominal readings of the pDR to overestimate mass concentration by a factor of 2.1(0.2). Our observations during high-humidity (≥95% RH) episodes, as well as in the presence of dense, poorly mixed, tobacco smoke, emphasize that extreme short-term excursions registered as high nominal particle concentrations may be artifacts of condensation in the sensing chamber. Accordingly, upper quantiles (both between 24-hour samples and within temporally-resolved samples) of PM observations, which are of particular interest to exposure-focused studies, are especially vulnerable to nephelometric distortion. Previously published nephelometric results from rural households do not take into account optical distortion mediated by condensed species. Datasets for which these distorted samples have not been censored may yield substantially inaccurate summary statistics in rural field contexts, both for the most common measures of central tendency (geometric and arithmentic mean) and for metrics based on upper quantiles. Research reviewed here and observations in single-source microenvironments in Hechengli substantiate that precise and accurate quantitative use of temporally disaggregated nephelometer data requires temporally disaggregated gravimetric calibration and that field calibration must be performed for each distinct microenvironment in which the nephelometer is employed.

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Tables

study	r^2, *n*	*range* ($\mu g/m^3$)	*relative*$^{\eta}$ *response*	*application*	*neph. intake*	*mass frac.*
Allen *et al.*, 2004	0.74, 194	~1-24	1.3	personal exposure monitoring	passive	$PM_{2.5}$
	0.66, 193	~1-42	1.16			
Chakra-barti *et al.*, 2004	0.16, 20	~5-25	1.53§	outdoor, Los Angeles	2.5 µm cut	$PM_{2.5}$
	0.56, 18	~5-25	1.06§	outdoor w/ empirical humidity correction		
Howard-Reed *et al.*, 2000	0.66, 34	~ 2-35	1.1	elderly population exposure; Baltimore, MD and Fresno, CA	passive	$PM_{2.5}$
Jenkins *et al.*, 2004	0.98, ~20	28-769	2.01	chamber ETS$^{\Phi}$	passive	RSP, 4.0 µm cut
	0.999, 15	82.5-2660	1.87	laboratory-generated cooking oil smoke		
	0.999, 15	48.5-684	0.92	cedar wood smoke (laboratory)		
	0.00013, 12	<50	0.016	propane stove (lab.)		
Lanki *et al.*, 2002	0.86, 308	~3- 60	1.85	exposure elderly population, Finland	2.5 µm cut	$PM_{2.5}$
Liu *et al.*, 2002	0.44, 16	~2- 25	1.01	personal, no cooking	passive	$PM_{2.5}$
	0.60, 16	~2- 20	1.14	personal w/ cooking		
	0.84, 16	~1- 40	1.64	indoor, no cooking		
	0.77, 16	~2- 40	1.69	indoor, cooking		
Quintana *et al.*, 2000	0.42, 83	3-26	1.41	indoor	passive	$PM_{2.5}$
	0.20, 81	11-53	0.60			PM_{10}
	0.62, 25	2- 14	2.63	outdoor, Alpine, CA (San Diego region)	2.5 µm cut, inlet heated	$PM_{2.5}$
	0.16, 25	9- 49	0.94			PM_{10}
Sioutas *et al.*, 2000	— , 39	< 100	0.93	ambient Los Angeles	2.5 µm cut, diffusion dryer	$PM_{2.5}$
	0.80, 39	~20-320	1.23	nitrate corrected, ~3-fold concentrated		
	0.88, 39	~20-320	1.21	nitrate uncorrected		
Wallace *et al.*, 2003	— , 28	not reported	1.5	ambient, official EPA monitoring sites,	passive	$PM_{2.5}$
Wallace *et al.*, 2006	0.87, 784	~3-110	1.71	indoor residential	passive	$PM_{2.5}$
	0.70, 728	~3-200	1.51	personal exposures		
Wu *et al.*, 2005b$^{\xi}$	0.5, 260	~2-38	1.6$^{\Omega}$	indoor	passive	$PM_{2.5}$
	0.35, 48	~4-32	2.3$^{\Omega}$	outdoor, Alpine, CA	active, unheated	
	0.5, 60	~3-14	1.1$^{\Omega}$		heated inlet	
	0.31, 246	~2-23	2.3$^{\Omega}$		heated + humidistat	

η: Ratio of nominal nephelometric to grav. mass conc'n; based on linear regression coefficient, save where noted.
§: Constructed from geometric means of measurements, rather than as linear regression coefficients.
Φ: Machine-smoking protocol constructed to emphasize mainstream tobacco smoke.
ξ: Wu *et al.* (2005b) develop quality control procedures for significantly improved correlations.
Ω: Wu *et al.* (2005b) report median ratio of pDR and gravimetric measurements.

Table 1, PUBLISHED CORRELATIONS BETWEEN pDR AND GRAVITATIONAL PM: Relationships between nominal pDR measurements and gravitationally determined particle concentrations for laboratory, field (indoor and ambient), and personal exposure microenvironments, 24-hour basis.

Level	*n*	*response ratio*$^{\varphi}$, *geo. mean (GSD)*
kitchen with RH<95% (1-min. resolution)	16	2.13 (1.47)
kitchen with high RH (>95%) excursion	5	7.40 (4.66)
village office with heavy smoking (8-hour)	2	27.13 (1.55)
solid-fuel coal stove for boiling water	2	0.78 (1.03)
laboratory mock coal fire pit, water boiling, (1.5 hr.)	2	0.91 (1.23)
non-smoking hotel room, Yanbian, China	4	2.30 (1.14)
whole-model test statistics: (Prob>F) =0.0002, r^2=0.59, n=32		

φ: Response ratio constructed as geometric mean (geometric standard deviation) of individual samples' ratios of optical to gravitational mass concentration. Lack of correlation in high-humidity and dense smoke micro-environments precludes presenting relative response in terms of linear regression coefficient, as in Table 1 and in accord with dominant reporting convention.

note: coarser aggregation, with heavy-smoking and high-humidity kitchen samples grouped together as "condensation-corrupt data" and mock fire pit and solid-fuel coal stove grouped as "near-zone coal fires," yields similar results with diminished explanatory power (r^2=0.46, Prob>F =0.0006), condensation-corrupt data being significantly different from other groups and there being insufficient power to distinguish between relative response of non-condensation sub-groups.

Table 2, OBSERVED RATIOS OF pDR TO GRAVITATIONAL PM MEASUREMENTS: Means and standard errors of mass concentration ratios of nephelometrically determined PM (raw pDR data) to gravitational RSP in 6 distinct microenvironments. Unless otherwise noted, all measurements are associated with indoor environments in Hechengli village, 24-hour basis.

		24-hour average PM (mg/m³)	*24-hr. median PM (mg/m³)*	*maximum 1-hr. PM (mg/m³)*	*maximum PM (mg/m³)*
ARITHMETIC MEAN (SE)	censored (n= 43)	0.312 (0.039)	0.164 (0.032)	1.88 (0.25)	8.30 (1.50)
	uncensored (n=58)	0.521 (0.070)	0.148 (0.024)	5.42 (1.13)	34.5 (7.3)
	2-way ANOVA: Prob > F, r^2	***0.0019, 0.13***	*0.48, 0.007*	***0.0024, 0.13***	***<0.0001, 0.23***
GEOMETRIC MEAN (GSD)	censored (n=43)	0.233 (2.2)	0.099 (2.9)	1.26 (2.6)	4.35 (3.4)
	uncensored (n=58)	0.333 (2.7)	0.096 (2.6)	2.22 (3.9)	9.94 (5.9)
	2-way ANOVA: Prob > F, r^2	***0.0011, 0.14***	0.88, 0.0003	***0.0003, 0.18***	***<0.0001, 0.24***

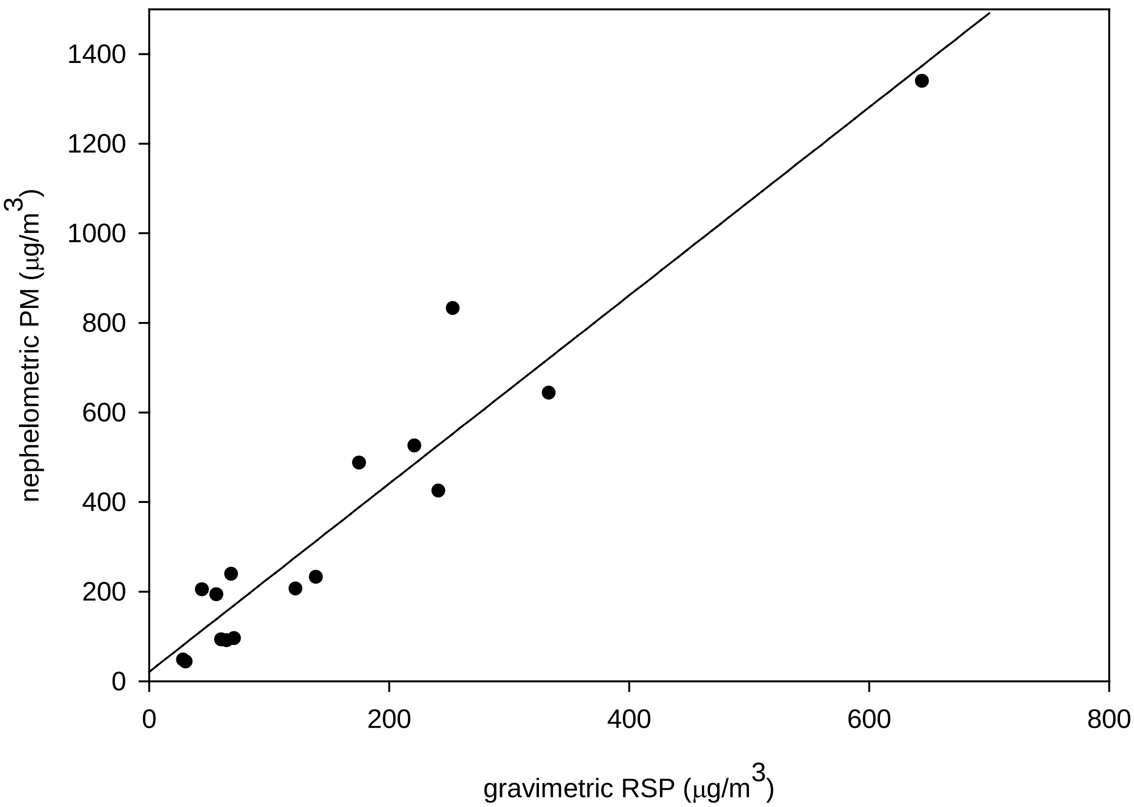
Table 3, PM SUMMARY STATISTICS, CENSORED & UNCENSORED: Summary statistics and ANOVA based on arithmetic (*top*) and geometric (*bottom*) means of nephelometrically determined PM (calibrated to gravimetric RSP) in 24-hour samples in Hechengli village kitchens. Groups represent uncensored dataset and that censored to exclude samples afflicted by condensation in the nephelometer's optical chamber.

	24-hour average PM (mg/m³)	*24-hr. median PM (mg/m³)*	*maximum 1-hr. PM (mg/m³)*	*maximum PM (mg/m³)*
censored (n= 43)	0.0323-1.44	0.0042-1.28	0.159-6.2	0.328-40.4
uncensored (n=58)	0.0323-**2.63**	0.0042-1.28	0.159-**454.5**	0.328-**194**

Table 4, RANGE OF OBSERVED PARTICLE CONCENTRATIONS, CENSORED & UNCENSORED: Range of observations in 24-hr. kitchen samples of PM (optical determination calibrated to gravimetric sub-sample) in Hechengli village, with deviations of the uncensored dataset highlighted (*bold font*).

Figures

Figure 1, HECHENGLI VILLAGE KITCHEN: A Han-style kitchen showing the *kang* and a second solid-fuel (coal) stove for boiling tea water and fueling the radiant heating system.



neph-PM =21 µg/m^3 + 2.1 grav-RSP
intercept not significantly different from zero (Prob>t = 0.57), standard error of slope = 0.17.
Figure 2, OBSERVED RELATION OF OPTICAL TO GRAVITATIONAL PM: Linear fit of nephelometrically determined PM to gravimetric RSP in 24-hr. kitchen samples for which relative humidity did not exceed 95% (r^2=0.92, Prob>F < 0.0001, n=16). Omitting the outlier at 644 µg/m^3 yields a similarly significant relationship with diminished power (r^2=0.82, Prob>F < 0.0001, n=15). Censoring the outlier neither significantly changes the slope (2.3, se=0.3) nor perturbs the intercept from zero (5.4 µg/m^3, se = 47 µg/m^3).

Endnotes

[1] SAE Fine test dust is log-normally distributed with MMD between 2 and 3 µm, GSD 2.5, bulk density 2.60-2.65 g/cm^3, and refractive index 1.54.
[2] Producer gas comprises ca. 21% CO, 12% H_2, 2% CH_4, 14% CO_2, and 51% N_2. In Hechengli village, a nascent energy project generates producer gas from wood chips and agricultural residues. Some households had limited availability of producer gas during the second of two field missions.
[3] This fraction is preferred by many occupational hygienists, in part because associated cyclones' gentle 50% cutpoint at 4.0 µm admits a particle fraction more similar to what is deeply inhaled by human beings than sharply-cut fractions at 2.5 or 10 µm.
[4] Although our nephelometric dataset was purged of samples that were nonsystematically distorted via condensation in the optical chamber, approximate 1-hour peak PM concentrations of our study are semi-quantitative in that nephelometric response during high-pollution episodes may depart from that derived from 24-hour calibration. Published laboratory studies which explore nephelometric response to cooking-generated aerosols suggest that optically-determined 1-hour peak PM concentrations might overestimate mass by about 10% (Jenkins *et al*. (2004), cooking oil) or underestimate 1-hour peaks by a factor of 2.5 (Brauer *et al*. (2000), frying potatoes).