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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. Our investigation of relationships between 24-hour optical and gravitational particle measurements in rural Chinese kitchens depicts that where relative humidity remained below 95%, nephelometric response was strongly linear despite complex mixtures of aerosols. Where 95% relative humidity was exceeded for even a brief duration, nephelometric data were nonsystematically distorted, 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 both within and between samples. Precise, accurate interpretation of optically resolved short-term particle concentrations requires short-term gravitational sampling concurrent with optical methods.

Introduction

Indoor air quality in rural kitchens: a public health priority

Although indoor air pollution has garnered increasing attention since the *First International Indoor Climate Symposium* (Denmark, 1978), research has concentrated on established market economies, e.g., Western Europe, Japan, U.S.A. The emphasis has been on sick building syndrome, tobacco smoking, and potential correlates to asthma. Far fewer resources have been dedicated to indoor air quality (IAQ) in rural kitchens, although they constitute the most significantly polluted air pollution microenvironment in terms of number of people exposed, magnitudes of exposure, and healthy life lost [1].

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 2 to 3 orders of magnitude higher for indoor combustion sources than for outdoor sources [2-5]. Compounding the problem of greater intake for indoor sources is that solid fuel/stove combinations in China are 2 to 3 orders of magnitude more polluting than their gas counterparts: 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 [6]. 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 [7].

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

Temporally disaggregating daily 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 [8] and several animal studies offer evidence for PM-mediated health effects at time scales shorter than 24 hours. For example, inflammatory reactions (latency period 6-hr) have been observed in response to 1-hr. exposures to diesel exhaust [9], and episodes of asthma have been found to correlate better to 1-hr. and 8-hr. PM_{10} concentrations than to 24-hr. means [10, 11]. Resolving epidemiologic and dose-response relationships calls for finer (than 24-hr.) 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 micro-environment) 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 in 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. Moreover, these rural field studies do not explicitly contend with high relative humidity excursions [12-15].

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

A number of laboratory and field investigations have characterized correlations between the optical signals of portable nephelometers and gravimetric particle samples. Compiled in Table 1, these studies are briefly reviewed below.

Laboratory and theoretical investigation of humidity effects on optical signal

Two independent laboratory investigations observe that at relative humidities (RH) above 70% a portable nephelometer known as the personalDataRAM (MIE, Inc., Bedford, MA), henceforth pDR, substantially and nonlinearly overestimates mass concentration [16, 17]. Above 95% RH, the overestimation of mass concentration exceeds a factor of 5 and is exponentially sensitive to relative humidity. For a well-characterized particle distribution, Chakrabarti et al. [16] and Sioutas et al. [17] provide empirical evidence of pDR adherence to the theoretical trend regarding sensitivity of scattering signal to relative humidity with their laboratory results conforming to the same approximately exponential curve. While these studies offer evidence for correcting pDR signals based on concurrently monitored relative humidity, strong dependence of humidity-induced distortion of the optical signal on particle size and composition precludes applying a humidity correction where the particles of interest are not physically and chemically well-characterized [18], 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 variation in particle concentrations spans less than 50 $\mu g/m^3$ to several *thousand* $\mu g/m^3$.

Optical/gravimetric correlations in controlled settings and applicability to field conditions

Two studies investigate nephelometric response to a variety of laboratory-generated PM simulating cooking, wood-burning, gas stove use, and tobacco smoke [19, 20]. These studies indicate that: (a) for single-source particulate matter, nephelometric correlation to gravitational methods is highly linear in controlled laboratory conditions; (b) different laboratory protocols suggest different relative nephelometric responses for a given particle type (e.g., ETS, frying food); (c) laboratory calibration for field applications is complicated by the mixing-dependence of optical response for tobacco smoke and by the variability of observed response for different sources and particle-generation protocols. In particular, these studies demonstrate that material source does not suffice to characterize nephelometric response, which is sensitive to human behaviors and microenvironmental mixing dynamics.

For simulated ETS and cooking-derived PM in a laboratory setting, Brauer et al. [19] investigate the pDR's correlation to mass concentrations determined by the $PM_{2.5}$ Harvard Impactor and by piezobalance (TSI model 8510, St. Paul, MN) equipped with a $PM_{3.5}$ impactor. In laboratory microenvironments associated with toasting bread, ETS, and frying potatoes, nominal (factory-calibrated to SAE Fine test dust[1], a.k.a. "Arizona Road Dust"), nephelometric response overestimates gravitational $PM_{2.5}$ by factors ranging 3.14 to 5.18. Brauer et al. also observe that single particle sources in restaurants, bars, and kitchens yield relationships similar to their laboratory analogues, but their field data portray greater within-source variability than would allow for distinction between sources based on relative response observed in the laboratory.

Jenkins et al. [20] observe strongly linear relationships (r^2 =0.98-9.999) between pDR response and 24-hr. gravimetric determination of respirable particles (RSP) in three laboratory simulations of ETS, cooking oil fumes, and cedar wood smoke. For particles generated by a propane stove, pDR and gravimetric results were uncorrelated (r^2=0.0013), in part due to the low-signal/noise ratio for both gravimetric and optical methods at the low particle concentrations (maximum 46 $\mu g/m^3$) attainable within the controlled temperature of the test chamber. While Jenkins et al. suggest that higher optical response obtains for dense human-generated ETS than for the aged and diluted sidestream tobacco smoke generally associated with chamber studies, their chamber ETS protocol, which emphasizes mainstream smoke, produces a two-fold lower relative response than the dilute chamber smoke of Brauer et al. [19].

Optical/gravitational correlations in field settings

Several researchers have explored correlations between nephelometric signal and gravitationally determined mass concentration in ambient environmental, indoor, and personal exposure settings [12, 16, 17, 19, 21-24]. These studies demonstrate: (a) linear relationships of varying strength between scattering coefficients (nominal nephelometric signal) and mass concentrations in well-defined field environments 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 (24-hr.) measurements may not apply to short-term pollution episodes, particularly those associated with cooking.

Brauer [12] investigated PM levels inside and outside of homes in Mexico (n=22) and in British Columbia, Canada (n=6) where biomass combustion occurred indoors. In both indoor environments, highly significant (r^2=0.79-0.95) linear relationships between gravitationally determined PM_{10} and $PM_{2.5}$ and a passive-mode nephelometer were borne out over a range of PM concentrations (10-1,600 $\mu g/m^3$), with $PM_{2.5}$ being more closely correlated to the nephelometer signal than PM_{10}. Optical to gravitational mass ratio observed in British Columbia was significantly different from that in Mexico, emphasizing that interpretation of nephelometric data is application-specific, even where dominant sources are in the same broad category (e.g., small-scale combustion of wood). Brauer attributes weaker correlations among outdoor measurements by collocated nephelometer and impactor (r^2=0.5) to variable water content and volatile fraction.

Two studies investigate ambient Los Angeles air basin PM and attempt to clarify driving factors in nephelometric response [16, 17]. With relative humidity reduced to less than 50% by use of a diffusion dryer tube and active-mode operation (2.5 µm cut), Sioutas et al. [17] find particle size to dominate chemical composition as a determinant of the linear relationship between scattering signal and mass concentration for ambient air in the Los Angeles basin.

Several studies of environmental concentrations and personal exposures deemed fluctuations of relative humidity inconsequential on the basis of humidity being less than 60% for the vast majority of the monitoring period and less than 70% for indoor environments in general [19, 21-23, 25]. However, Liu et al.'s [23] study of residential indoor air quality found the disparity between short-term particle concentrations reported by pDR's and Radiance nephelometers calibrated gravimetrically on a 24-hr. basis to be statistically significant ($p<0.0001$) during cooking periods, especially where baking and frying occur. Both of these light-scattering devices were highly correlated to gravimetric measurements on a 24-hr. basis, and these 24-hr. correlations were not affected by whether tobacco smoking or cooking activities transpired within the 24-hr. period. The authors warn that for analysis of temporally disaggregated nephelometric data, gravimetric calibrations based on 24-hour. samples should be "used with caution" [23].

Previous use of nephelometers for temporal disaggregation in rural field settings

Three previously published studies of indoor air quality in rural households use the pDR to afford some level of temporal disaggregation not available from filter samples alone [13, 15, 26]. Brauer et al. [13] infer from short-term nephelometric measurements 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. The other two studies demonstrate methodology for quantitatively characterizing exposures of cooks in India [15] and Kenya [26].

Ezzati et al. use the pDR to monitor time-resolved (1-min.) concentrations of PM in 14-hr. samples in Kenyan households using firewood in a traditional three-stone fire, firewood in three different locally available metal body/ceramic liner stoves, charcoal in an unlined *jiko* stove, charcoal in a "Kenya ceramic"(-lined) *jiko*, or charcoal in a metal-lined *roketto* stove [26, 27]. No scaling of nominal data to gravitational samples is reported. In addition to mean and median concentrations, Ezzati et al. compare within-sample 75th and 95th percentiles of time-resolved (1-min.) PM concentrations as indicators of active use periods and worst-case stove performance, situations that contribute heavily to personal exposures.

In a study involving gravimetric $PM_{3.5}$ measurements in 412 rural homes in Andra Pradesh, Balakrishnan et al. [15] use time-resolved (1-min.) pDR logs in a subsample (10%) of homes to indicate the relative ratio of particle concentrations associated with cooking and non-cooking households.

Methods

Site

Field measurements represent wintertime kitchens of a rural village in China's Jilin Province, which borders Heilongjiang Province to the north, Inner Mongolia Autonomous Region to the west, Liaoning Province to the southwest, 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* (炕) 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). Cooking is performed atop one or two heavy iron circular plates built into the brick *kang* structure. These surfaces are of diameters 0.5-0.8 m.

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 Villages 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. One sample was excluded from analysis due to power failure of the nelphelometer; thus, 22 pairs of optical and gravitational particle measurements were used in this analysis.

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-hr. period), n=4; laboratory-simulated coal-burning fire pit [28], 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-hr. for these samples except where a specific activity of shorter than 24-hr. duration was of interest, namely laboratory simulation of a coal-burning fire pit (1.5-hr.) 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 (MIE, Inc., Bedford, MA) 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. It 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 $1.5x10^{-6}$- 0.6 m^{-1} and calibration on SAE Fine test dust (see endnote 1).

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 drifted during the field session by placing the pDR's in the zero bags and noting readings, checking that pDR's gave comparable responses when placed side-by-side in the same 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 checked for zero drift once every 2 weeks. By initially zeroing the instruments in a laboratory in California and pumping at least 3 air changes (until both instruments registered no more than 0.001 mg/m^3) of Grade BQ-filtered air into zero-bags before beginning the zero process, we assured that zero's were the same as would have obtained under much cleaner ambient conditions than my roadside hotel rooms offered

Gravitational particle measurements

A NIOSH-compatible 10 mm, multiple-inlet, conductive plastic (impervious to electrostatic effects) 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. The multi-inlet cyclone is designed to eliminate problems associated with the Dorr-Oliver cyclone through a higher specified flow rate (2.75 l/min); tangential inlet design, which reduces particle loss to impaction; and multiple inlets to reduce sensitivity to orientation.

To reduce sensitivity of gravitational analysis to filter conditioning and to save labor, matched-weight 37 mm, 5 µm pore size PVC filters were chosen. Each matched-weight cassette comprises two filters matched (within 25 µg) in weight and loaded into a cassette in 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-hr. 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.

Filters were weighed on a Cahn-29 Electrobalance (0.001 mg resolution) in Professor Katherine Hammond's (Environmental Health Sciences, UC Berkeley) weighing room. Prior to weighing, filters were equilibrated for 24 hours 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. Because matched-weight filters were used, with the difference between filters representing the mass of the sample, no pressure correction was necessary. As specified by the Hammond Lab weighing protocol, 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 $\mu g/m^3$. Gravimetric readings were adjusted for the non-zero mean (mean(se) =-7.42(2.83) µg) of twelve blanks.

Temperature and relative humidity

Time-resolved measurements of temperature and relative humidity were logged by Onset Corporation's HOBO-T/RH devices. Like the HOBO-CO, the HOBO-T/RH is inexpensive ($84) and portable (60×48×19 mm, 29 g), boasting non-volatile memory and 1 year of operation on a user-changeable battery. Field logistics and the smaller 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 –20ºC -70ºC. The relative humidity sensor is rated to ±5% over a range of 25% RH-95% RH at 5ºC -50ºC.

Results and analysis

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=6) 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, mean(std. err.) of response ratio=2.3(0.2)),
2. kitchens with high-humidity excursions (n=6, mean(std. err.) of response ratio=12(5))
3. heavy smoking indoors (n=2, mean(std. err.) of response ratio=28(9)).

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) illustrate 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 PM peaks for which the raw optical signal 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 [20]. Samples plagued by condensation events remain significantly different when high-humidity kitchens and heavy smoking are grouped together (n=8).

Inasmuch as cooking episodes may be associated with high relative humidity excursions or dense plumes of condensed-phase species, our observations call into question two assumptions implicit in previous use of the pDR to reconstruct human exposures to PM in rural Indian kitchens [15]. 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, equivalently, that nephelometric response per unit particle mass is constant during cooking and non-cooking episodes. A second assumption that appears to be embedded in this study (authors do not indicate whether pDR data processing was disaggregated by fuel/stove type) is that the ratio of cooking to non-cooking particle concentrations is constant across fuel/stove types.

Given ratios of nominal pDR response to gravitational samples ranging from 0.92 to 5.5 in previously published sources (Table 1) and 0.80 to 12 in 24-hr. kitchen measurements of our research (Table 2), previously reported 14-hr. mean particle concentrations in Kenyan households [26, 27] may substantially deviate from gravitationally-determined particle concentrations. Nonuniform deviation from gravitational measures is particularly likely because these measurements were 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

Over the course of fieldwork, the mean(se) of 24-hr. gravitational samples (n=22) in Hechengli Village kitchens was 156(23) µg/m^3, with a range of 28.3 µg/m^3 to 696 µg/m^3. The 24-hr. mean(se) of relative humidity was 65.3(1.9)% (range 35.9-90.9%). On a three-minute basis, relative humidity ranged 16.2-100%, and per-sample peak relative humidities spanned 59-100% (mean(se)=85.6(2.4)%).

Analysis of 22 samples from kitchen microenvironments does not support a strong or significant linear relationship between the optical and gravitationally determined mass concentrations (r^2=0.12, Prob>F =0.113).

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

Omitting those samples from which relative humidity was logged in excess of 95% (n=6) 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(se=0.17).

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 stronger 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; optical properties of PM in Hechengli Village kitchens are reasonably uniform on a 24-hr. basis.

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

When the 6 samples for which relative humidity exceeded 95% are examined, no significant linear relationship to gravitational RSP is found (r^2=0.019, Prob>F = 0.79). The temporal duration for which relative humidity in excess of 95% was logged (ranging 3 to 485 minutes) was a strong and significant predictor of the optically determined mass concentration (r^2=0.71, Prob>F =0.034). Incorporating gravitationally determined RSP did not improve model fit, nor was gravimetric RSP a statistically significant effect (Prob>F =0.93) in the two-effect model. Within-sample censoring of data points (1-min. resolution) logged during high-humidity episodes from 24-hr. optical average mass concentrations did not significantly improve their correlation to gravimetrically determined RSP; rather, a strong linear relationship between the thus-censored dataset and original average (plagued by high humidity excursions) prevailed (r^2=0.89, Prob>F =0.0047). This suggests that optical effects of condensation in the optical chamber are not as localized in time as are ambient excursions in relative humidity.

Our finding that simple censoring based on time-resolved relative humidity fails to correct pDR data plagued by high humidity excursions partly explains poor correlations to gravimetric samples observed by Quintana et al. [21], even after attempting humidity correction: their data processing algorithm involved exclusion of data coincident with high-humidity excursions (>85%) and interpolation with a cubic spline. Quintana et al.'s censorship of data points coincident with >85% RH led to exclusion of 10-25% of pDR data.

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 peak measures of PM as well as 24-hour averages for summary statistics constructed as both arithmetic (Table 3) and geometric (Table 4) means: 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 5): on 24-hr. and 1-hr. 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 samples suffering condensation-induced optical distortion.

While the 24-hr. median (constructed for each 24-hr. sample from concentrations logged at 1-minute intervals) is robust to relative humidity-induced distortion, this steadfast measure of central tendency refuses straightforward interpretation in terms of gravitationally determined RSP [29]. Thus, while the most robust measure of central tendency—the median—reported by previous use of a pDR in rural Kenyan homes [26, 27] may not be afflicted by distortion induced by condensation in the optical chamber, reported 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.

Ezzati et al. [26] 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 by correlation to longer-term gravimetric samples. In particular, our observations indicate that metrics based on upper quantiles of temporally resolved particle concentrations are particularly sensitive to nephelometric distortion induced by condensed phase species.

Conclusion

Under conditions for which humidity is highly variable but nowhere in excess of 95%, 24-hr. average 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, and different cooking and heating stoves and behaviors. On a 24-hr. basis, we observe nominal readings of the personalDataRAM (MIE, Inc., Bedford, MA) to overestimate mass concentration by a factor of 2.1(std. err.=0.17). Data from Hechengli 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-hr. 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 use of the nephelometer in rural households does not take into account optical distortion mediated by condensed species. Mean optically-determined PM concentrations of 24-hr. samples for which the scattering signal suffers even short-term condensed phase distortion are not readily corrected through point-by-point censorship on the basis of concurrently logged relative humidity measurements. Moreover, 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[η] response*	*application*	*neph. intake*	*mass frac.*
Brauer et al. 2000[¶] [19]	0.93, 6	26-270	3.14	toasting bread	fan with no size selection	$PM_{2.5}$
	0.99, 5	57-381	4.59	chamber ETS[c]		
	0.87, 5	43-250	5.18	frying potatoes		
Jenkins et al. 2004 [20]	0.98, ~20	28-769	2.01	chamber ETS[Φ]	passive	RSP, 4.0 µm cut
	0.999, 15	82.5-2660	1.87	cooking oil		
	0.999, 15	48.5-684	0.92	cedar wood smoke		
	0.00013, 12	<50	0.016	propane stove		
Brauer 1995[£] [12]	0.85, 31	30- 1490	1.68	rural Mexico, indoors, all fuels	passive	$PM_{2.5}$,
	0.83, 32	40- 1660	1.27			PM_{10}
	0.81, 15	30- 1490	1.66	rural Mexico, indoors, biomass only		$PM_{2.5}$,
	0.79, 16	50- 1660	1.30			PM_{10}
	0.95, 5	40- 160	2.82	rural Mexico, indoors, LPG only		$PM_{2.5}$,
	0.84, 5	40- 680	0.72			PM_{10}
	0.50, 11	20- 50	3.70	rural Mex., outdoors		$PM_{2.5}$,
	0.84, 6	10- 50	6.45	indoors, wood fuel, British Columbia		$PM_{2.5}$,
Sioutas et al. 2000 [17]	— , 39	< 100	0.93	ambient L.A. air	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		
Chakrabarti et al. 2004 [16]	0.16, 20	~5-25	1.53[§]	ambient L.A. basin	2.5 µm cut	$PM_{2.5}$
	0.56, 18	~5-25	1.06[§]	ambient w/ empirical humidity correction		
Howard-Reed [24]	0.66	~ 2-35	1.1	elderly population exposure	passive	$PM_{2.5}$
Quintana et al. 2000 [21]	0.42, 83	3-26	0.71	indoor air	passive	$PM_{2.5}$
	0.20, 81	11-53	1.66			PM_{10}
	0.62, 25	2- 14	0.38	outdoor air	2.5 µm cut, inlet heated	$PM_{2.5}$
	0.16, 25	9- 49	1.06			PM_{10}
Liu et al. 2002 [23]	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		
Lanki et al 2002 [22]	0.86, 308	~3- 60	1.85	exposure elderly population, Finland	2.5 µm cut	$PM_{2.5}$

η: ratio of nominal optical to grav. mass conc'n; based on linear regression coefficient, save where noted.

¶: 6-hr. measurements using the Radiance Research nephelometer based on scattering at 530 nm.

£: 9-hr. measurements using the Radiance Research nephelometer based on scattering at 530 nm.

§: Constructed from geometric means of measurements, rather than as linear regression coefficients.

Φ: Machine-smoking protocols differ, mainstream accounting for 11% of [20], filtered from [19].

Table 1, PUBLISHED CORRELATIONS BETWEEN OPTICAL AND GRAVITATIONAL PM: Relationships between optical and gravitationally determined PM for laboratory, field (indoor and ambient), and personal exposure microenvironments. 24-hour basis, pDR (scattering at 880 nm).

Level	*n*	*response ratio*$^{\varphi}$*, (mean (se))*	*CV*
kitchen with RH<95% (1-min. resolution)	16	2.30 (0.24)	42%
kitchen with high RH (>95%) excursion	6	11.7 (5.0)	104%
village office with heavy smoking (8-hr.)	2	28.4 (8.6)	43%
solid-fuel coal stove for boiling water	2	0.789 (0 19)	34%
laboratory mock coal fire pit, water boiling, (1.5 hr.)	2	0.920 (0.14)	22%
non-smoking hotel room, Yanbian, China	4	2.31 (0.15)	13%
whole-model test statistics: (Prob>F) <0.0001, r^2=0.63, n=32			

φ: Response ratio constructed as arithmetic mean of individual samples' ratios of optical to gravitational mass concentration due to lack of correlation in high-humidity and dense smoke microenvironments. Note that **Table 1** presents relative response in terms of linear regression coefficient, 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 small sample size being unable to resolve relative response of non-condensation sub-groups.

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

	24-hour average PM (mg/m^3*)*	*24-hr. median PM (mg/m*^{3}*)*	*maximum 1-hr. PM (mg/m*^{3}*)*	*maximum PM (mg/m*^{3}*)*
censored (n= 43)	0.312 (0.039)	0.164 (0.032)	1.88 (0.25)	8.3 (1.5)
uncensored (n=58)	0.521 (0.07)	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***

Table 3, PM SUMMARY STATS (arithmetic), CENSORED & UNCENSORED: Summary statistics as arithmetic mean (standard error) and ANOVA results for optically determined PM (calibrated to gravimetric RSP) in 24-hr. samples in Hechengli Village kitchens. Groups represent the full (uncensored) dataset and that censored to exclude samples afflicted by condensation in the nephelometer's optical chamber.

	24-hour average PM (mg/m^3*)*	*24-hr. median PM (mg/m*^{3}*)*	*maximum 1-hr. PM (mg/m*^{3}*)*	*maximum PM (mg/m*^{3}*)*
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***

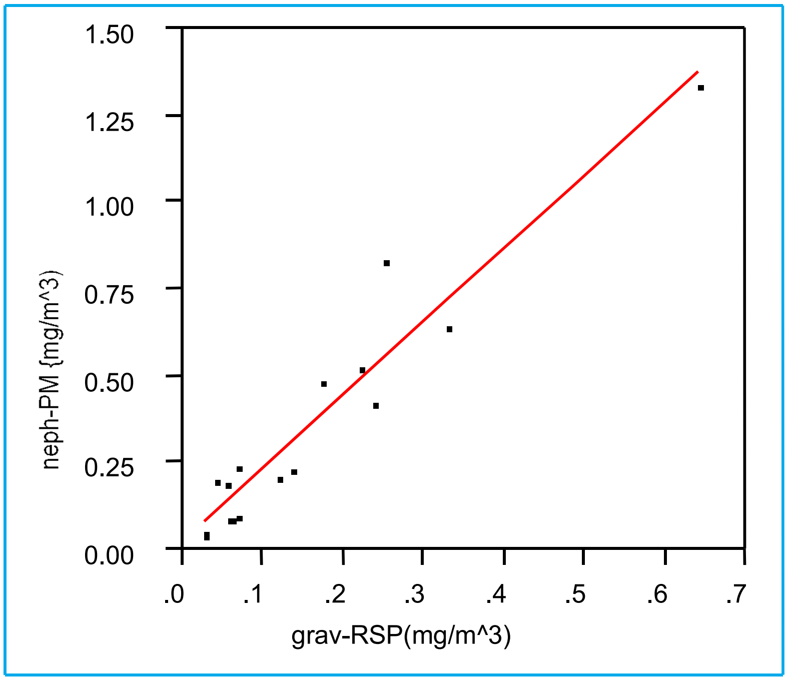
Table 4, PM SUMMARY STATS (geometric), CENSORED & UNCENSORED: Summary statistics as geometric mean (geometric standard deviation) and ANOVA for optically determined PM (calibrated to gravimetric RSP) in 24-hr. samples in Hechengli Village kitchens. Groups represent uncensored dataset and that censored to exclude samples afflicted by condensation in the nephelometer's optical chamber.

	24-hour average PM (mg/m^3*)*	*24-hr. median PM (mg/m*^{3}*)*	*maximum 1-hr. PM (mg/m*^{3}*)*	*maximum PM (mg/m*^{3}*)*
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 5, RANGE OF OBSERVED PM, 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* (炕) and a second solid-fuel (coal) stove for boiling tea water and fueling the radiant heating system.



neph-PM = 0.021 mg/m^3 + 2.1 grav-RSP
intercept not significantly different from zero (Prob>t = 0.57)

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).

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[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. It was also deemed appropriate for this study insofar as it is more closely matched than $PM_{2.5}$ to the particles registered, by virtue of aerodynamics combined with optical sensitivity, in the pDR's scattering chamber.

[4] Although our nephelometric dataset was purged of samples that were nonsystematically distorted via condensation in the optical chamber, approximate 1-hr. peak PM concentrations of our study are semi-quantitative in that nephelometric response during high-pollution episodes may depart from that derived from 24-hr. calibration. Published laboratory studies (Table 1) which explore nephelometric response to cooking-generated aerosols suggest that optically-determined 1-hr. peak PM concentrations might underestimate[4] mass by about 10% (Jenkins et al., cooking oil) or overestimate 1-hr. peaks by a factor of 2.5 (Brauer et al., frying potatoes).