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Analysis of time series of particle size distributions in nano exposure assessment

Rinke H. Klein Entink*, Cindy Bekker, Wouter F. Fransman, Derk H. Brouwer

Institute for Applied Scientific Research (TNO), Zeist, The Netherlands

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ABSTRACT

Real-time exposure measurements to nano-sized particles may result in large amounts of time series data on particle size and total number concentration. Analysis of the particle size distribution have thus far been limited to either graphical analysis of the distribution over time or an evaluation of the mode over time. For large time series data, graphical analysis of distributions is complicated and an assessment of the mode ignores the important aspect of the variance in particle size. A statistical method of analysis is proposed that overcomes those problems, based on a multilevel modeling approach and assuming a lognormal model for the particle size distribution. Two empirical examples illustrate the advantages of the proposed model, showing that useful summaries and inferences can be obtained, even for large data sets. The model thus provides a tool for practitioners to deal with large amounts of particle size distribution data obtained from real-time nano measurement devices.

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1. Introduction

Because of the increasing number of workers involved with nanotechnology and the potential health effects of working with these nanomaterials, assessment of the exposure of workers to (manufactured) nano particles or more specifically nanoobjects and their agglomerates and aggregates (NOAA), (ISO 2012) at the workplace receives considerable attention. To locate sources of emission and to characterize different work situations in order to gain knowledge on exposure and how to reduce l exposure levels, workplace aerosol measurements are performed. Because the size and associated surface area of the particles in the (workroom) air is one of the most important parameters for studying manufactured nano particles with respect to potential risk, most sampling methods for measuring nano-sized particles focus on both particle number concentration and particle size distribution (PSD) using real-time size, resolved devices, e.g. Scanning Mobility Particle Sizer (SMPS), Electrical Low Pressure Impactor (ELPI), Aerodynamic Particle Sizer (APS), etc.) rather than on the total particle number concentration in a certain size range alone (e.g. optical counters like the Condensation Particle Counter (CPC), and diffusion charging based devices like DiscMini, Nanotracer, etc.).

However, little attention has been paid on how to (statistically) analyze and report these measurement results. Both particle number concentration and PSD have been studied using graphical methods (Brouwer et al., 2004; Demou et al., 2008; Evans et al., 2010; Bekker et al., 2014). Although the authors showed that useful information could be retrieved, graphical analysis is limited to making qualitative inferences. Quantitative analyses have often been limited to averages or,

* Correspondence to: TNO, P.O. Box 360, 3700 AJ, Zeist, The Netherlands. Tel.: +31 888 66 2614.
E-mail address: Rinke.kleinentink@tno.nl (R.H. Klein Entink).

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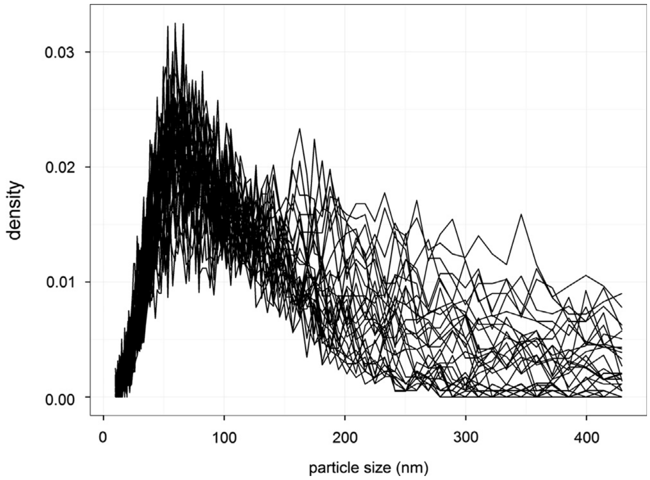

Fig. 1. Empirical particle size distributions, 30 time points recorded by an SMPS measurement device.

in the case of PSD, using the mode of the distribution. The latter thereby ignores that, to characterize a distribution, the variance is important as well. A complicating factor is that the use of real-time size, resolved devices differ from traditional time-integrated sampling methods for exposure in such a way, that the resulting time series measurements capture the exposure dynamics (e.g, changes in exposure levels over time during task-based measurements). Such time series are also more difficult to analyze because the measurements are not independent; Real-time measurements often result in autocorrelation patterns over time and require special statistical treatment to avoid biased estimates of standard errors (O'Brien et al., 1989). To overcome this problem, Pfefferkorn et al. (2010) and Klein Entink et al. (2011) have modeled particle number concentrations using Auto-Regressive Integrated Moving Average (ARIMA) time series models.

For the analysis of time series of PSD, no such method has thus far been proposed. Figure 1 shows 30 empirical PSDs from the recording of an experimental setting with an SMPS. This figure illustrates three important discussion points for the analysis of time series of PSD. Firstly, the empirical distribution is not smooth, but a rough, edgy line, possibly due to sampling variability of the instrument. This makes it harder to compare curves taken at two specific time spots graphically, or tell with certainty that their modes differ. Secondly, it shows that the mode might be a useful summary, but not a complete summary of what is happening. A summary using only the mode of a distribution misses important aspects of spread in particle sizes, for instance, not acknowledging that indeed two modes are the same but the tails of one distribution stretch further into the higher particle size ranges. Therefore, a measure of the variance should be included in an analysis as well. Thirdly, the sampling frequencies of real-time instruments may, depending on the type of device, result in a new curve every 1–10 s. A graphical inspection of all those curves would be simply unwieldy.

Therefore, in this paper we present a multilevel statistical model that overcomes those problems and allows the researcher to analyze time series of PSDs in the context of exposure assessment of NOAA. In the next section, the statistical modeling approach and the Bayesian analysis of the model is explained. In two empirical examples, the paper then describes the analysis of a continuous-drop experiment with calcium carbonate nano powder. The first example was recorded with an SMPS, the second example with an ELPI measurement device. A discussion of the proposed method concludes the paper.

2. Methods

A model is developed that consists of two levels. At the first level of modeling, the measurement level, we model the PSD at a specific time point by a lognormal distribution. This allows us to describe the mean or mode of the distribution, but also the spread in the measured particle sizes at each time point. At the second level of modeling we describe the evolution of the mean and variance of the level-1 lognormal distribution over time. This level-2 model includes possible explanatory variables in a linear regression model, like activity indicators, to describe how both the mean and variance of the level-1 models evolve. The inclusion of covariates allows us to test quantitative hypotheses about the potential effect of a workplace activity (or intervention measure) on the size of the particles a worker is exposed to. Finally, to account for possible remaining time-dependencies between the measurements, we introduce an autoregressive component on the residuals.

Analysis of the model is performed in the Bayesian framework using a Markov Chain Monte Carlo (MCMC) algorithm. The reason for doing so is that standard multilevel models for longitudinal data analysis model the mean over time (e.g., Hedeker & Gibbons, 2006). For our application, in the Bayesian framework it is straightforward to extend this analysis to also modeling the variance parameter over time. The Bayesian framework provides an elegant solution to jointly estimate all model parameters at once, as opposed to an ad-hoc analysis in multiple, independent steps.

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