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Theoretical Investigation of the Interrelationship Between Stationary and Personal Sampling in Exposure Estimation

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In exposure estimation, personal sampling is the method of choice as it is a nearby representative of the contaminant concentration in the breathing zone. Due to the versatility of the stationary sampling in obtaining much higher sensitivity, in its adaptability to telemetering observations, it may also be an attractive sampling method for many circumstances. However, the two sampling methods differ in many theoretically important ways that go beyond the obvious differences. The theoretical investigation of the stationary and personal sampling methods vis-à-vis sampling for exposure estimation shows that the area sampling can be used to represent personal sampling under restricted conditions. Under the restricted conditions, an area of concentration within specified bounds may be determined in relation to a reasonably well-defined source. The extension of the theory to multiple or ill-defined sources pose potential complications that may be intractable through a theoretical analysis. These limitations and restrictions are inherent to the underlying premises of the two methods; therefore they are not amenable to easy correction. Even though these restrictions may suggest only a limited role for area sampling in exposure assessment, the theory shown also suggests areas of further applied and theoretical research to extend the proper use of area sampling in exposure assessment.

Keywords Exposure Assessment, Sampling Theory, Area Sampling, Personal Sampling

The determination of exposures by personal sampling is the current method of choice for exposure assessment, because, as a nearby representative of the contaminant concentration in the breathing zone, this type of sample has been demonstrated to provide more accurate estimates of the exposure.(1) Historically, stationary or area monitoring has been used to estimate environmental concentrations or of contaminant source characteristics for control purposes. The use of these area-sampling results in the characterization of exposures was successful in the determination of disease causative factors when exposures and disease rates were high in exposed populations. A change in the philosophy of exposure estimation was in large part precipitated by the development of a sampling device that was small enough to be worn by a worker. In part, this development was also made essential by the requirement of more accurate exposure determination for the understanding and/or detection of causative factors that manifested much lower disease rates.(1–4)

Clearly, in exposure estimation, personal sampling is the method of choice as it is more representative of the contaminant concentration in the breathing zone and, therefore, a better estimation of the dose. However, there are many other reasons to collect occupational and environmental samples and for many such objectives, the use of stationary sampling may be more an attractive or appropriate alternative. Citing a few examples, the stationary samplers are effective as real-time monitoring devices or as multi-point and component telemetering instruments or as “large throughput volume” sample collection equipment in the detection of very low concentrations. Results obtained from stationary sampling can provide a tempting augmentation to an exposure assessment database. In fact, examples of concentrations of airborne contaminants that were collected using area monitoring devices in the development of estimates for individual or group exposures is not uncommon.(5–7) The discussions that concern the quality and the rigor of justification for such uses are beyond the scope of this article. However, the more exposure determinant personal sampling and generally more versatile stationary sampling differ in many conceptually important ways that go beyond the obvious differences. A theoretical discussion of these two methods should provide an understanding of the underlying principles as well as practical considerations in the conversion or interpretation of the results of one method in terms of the other.

Area sampling strategies involve placing a sampler in a stationary position for the entire sampling period. Historically, in occupational hygiene, an area sampling strategy was the most common method available for collecting samples of air contaminants. In the mid-1960s, when personal-sampling pumps became commercially available, the use of area sampling strategies for exposure determination began to diminish. It is clear from the early research that area-sampling results did not provide reliable estimates of personal exposure.(3,4) Differences were typically attributed to source-worker relationships without consideration to differences in the types of data compared.(3) Additionally, decreases in use of area sampling as a method for assessing exposure came from the necessity of determining compliance with occupational exposure limits. Only one of the current compliance sampling strategies required by Occupational Safety and Health Administration (OSHA) standards (Cotton Dust, 29 CFR 1910.1043) in the work place specifies or allows for the use of an area sampling strategy.(1,5)

In some recently reported epidemiological studies, historical area sampling results have been used in various ways to quantitatively estimate worker exposures or to develop exposure categories for purposes of exposure assignment.(6–9) Typically, exposure assignment methodologies include averaging area measurements over a specific time interval and then using the derived estimates to categorize a group or individual exposure. Rice et al.(6) used this method to estimate worker exposures to silica. Departmental or individual exposures were estimated using average dust concentrations derived from area measurements. These estimates were then coupled with an individual worker's time in a department or exposure area to estimate the time-related exposure. Stewart et al.(7) used a similar strategy as a part of a hierarchy to estimate job-related acrylonitrile exposures. Also, Sexias et al.(9) used area measurements in a regression analysis to determine departmental exposure assignment, which were modified for time and department to estimate historical exposures to crystalline silica in the diatomaceous earth industry.

In environmental sampling strategies, the use of area samples is much more prevalent, and, until recently, their use in exposure assessment has not been reasonably questioned.(10,11) Although correction of area measurements with time activity patterns(12) or a combination of personal sampling with time activity patterns have been proposed,(11–13) these methodological approaches have been validated only for very restrictive conditions.(14)

To set the stage for a broader theoretical discussion of the differences between stationary (S) and mobile or personal (P) sampling, let us consider a relatively ordinary example (see Figure 1). A process includes an initial phase, T1, where a W (worker) charges the system for processing. During the charging, controls are off so that the worker can fill the hoppers with feedstock. The total time of the process from beginning of charging to end of day is 10 equal units of time (T1 through T10). After initial charging, the controls are turned back on and the operator (O) takes over the process.

An area monitor is placed to record the airborne concentration during all phases of the operation. The area monitor records the average over all periods at 10 ppm for each process. The average concentration during T1 is 100 ppm. We assume that there are five similar processes and that immediately after each charging process, W spends time in a contaminant free area ($C = 0$ ppm). Before entering the work area, O has also spent his or her time in an area free of the contaminant of interest. During the day, W has charged all five of the processes and all five of the operators have experienced exposure while working on their specific processes. In addition, during the operation of the process, the concentration of the contaminant of interest is recorded as zero. Each operator is wearing a personal dosimeter that records the actual exposure. The exposure assigned to the worker, each operator based on the area monitor, and an accurate assessment of time spent on each task would be:

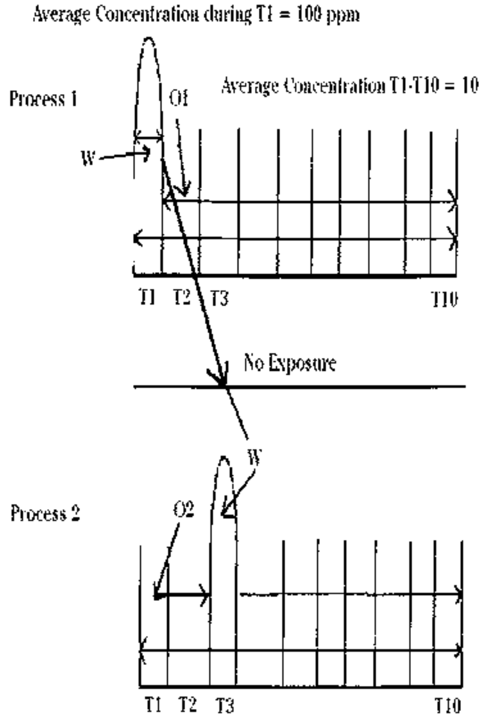

FIGURE 1
Representation of theoretical process.

Worker

$$\text{Eulerian Average} = (10 \times 0.5) + (0 \times 0.5) = 5\,\text{ppm}$$

$$\text{Lagrangian Average} = (100 \times 0.5) + (0 \times 0.5) = 50\,\text{ppm}$$

Operator

$$\text{Eulerian Average} = (10 \times 0.5) + (0 \times 0.5) = 90\,\text{ppm}$$

$$\text{Lagrangian Average} = (0 \times 0.5) + (0 \times 0.5) = 0\,\text{ppm}$$

Consequently, W's exposure would be judged one-eighteenth as much as the other workers (all operators). Clearly, W's average exposure is 50 concentration-time units and the operators are not exposed, given that when they are in the process area they have no exposure. In this example not only the exposure estimates are inaccurate, but the estimation method did not provide an accurate ranking of exposures. Obviously, using the Eulerian (fixed-point sampler) would result in a misclassification of exposure for both the worker and the operator. If an accurate time motion study were available, these exposures and their relative rankings would still be misclassified.

From a theoretical point of view, stationary and personal sampling is closely analogous to the theoretical definition of stochastic processes through two different formulations. An Eulerian formulation of a stochastic process deals with the time averages at a point in space. Therefore, any stationary sampling is analogous to a Eulerian formulation. In contrast, a Lagrangian formulation of the same stochastic process deals with the experience of a particle (or worker) in space as it travels from an initial point through time and space. Thus, the Lagrangian formulation deals with path and time dependent averages. This is analogous to personal sampling. Obviously, Lagrangian and Eulerian formulations will coincide in a field where the Eulerian average is space and time invariant. This is a very restrictive condition and essentially suggests that the Lagrangian process is defined over a well-mixed space that includes the Eulerian sampler and that this space is uniformly away from the influences of any source or sink. The conditions of interest are those that can be defined by the progressive relaxation of the space invariance. In addition, the definition of the point where the relationship between a Lagrangian and Eulerian formulation becomes untenably weak is of great import. Clearly, it would not be useful to discuss the equivalencies of processes in a strict mathematical sense. While a 10 percent absolute difference between two results may be mathematically enormous, in exposure estimation it may be accepted as negligible.

THEORETICAL CONSIDERATIONS

In the theoretical discussion, a number of formalisms will have to be used. We start with the definition of the Lagrangian and Eulerian averages. Let a concentration function be defined over a space—time domain $S(x, y)$. The Lagrangian average concentration over the path Ω is:

$$\bar{C}_L = \frac{1}{T|\Omega|}\int_0^T\int_\Omega C \cdot f(c, x, y, t)\, d\Omega(x, y)\, dt \qquad [1]$$

Where:

$\bar{C}_L$ = Lagrangian average concentration—M/L^3
C = Characteristic Concentration—M/L^3
$f(c, x, y, t)$ = Concentration distribution function over time and space
x, y = Space co-ordinates—L
T = Sampling time—T
Ω = Path of sampler travel

Similarly, the Eulerian average, $\bar{C}_E$, for a point $P(x_o, y_o)$ is defined by:

$$\bar{C}_E = \frac{1}{T}\int_0^T C \cdot f(c, x_o, y_o, t)\, dt \qquad [2]$$

We define the Eulerian sampler positioned at $P_o = (x_o, y_o)$ to be quasi invariant (δ-invariant) over a space $S_\delta \subset S$ such that $P_o \in S_\delta$ and:

$$C(P, t) = C(P_o, t)\exp(\eta), \quad -\delta < \eta < \delta\, \forall P \in S_\delta \qquad [3]$$

THEOREM I: If $\Omega \subset S_\delta$ then $\bar{C}_E \exp(-\delta) < \bar{C}_L < \bar{C}_E \exp(\delta)$

PROOF: The theorem is a direct consequence of the fundamental theorem of integration. Because $0 \leq f \leq 1$ and $C(\Omega)$ is bounded by $C(P_o, t)\exp(\pm\delta)$ substituting this information into Equation 1 will reduce it to Equation 2 with the proper multiplier.

To be able to use this theorem, it is necessary to investigate the possibilities of defining the boundary of S in relation to δ. Consider a source located at x_o, an Eulerian sampler located at a distance d_e and the point on the boundary of S located at d_b on the line connecting the sampler with the source. Consider the source emission rate $\dot{m}(t)$ and the wind velocity u. The maximum concentration at the sampler and boundary are given by the turbulent diffusion equation(10):

$$C(P_o, t) = \frac{\dot{m}(t - d_e/u)}{ukd_e^{1.5}} \quad \text{and} \quad C(P_b, t) = \frac{\dot{m}(t - d_b/u)}{ukd_b^{1.5}} \qquad [4]$$

Assuming that either the emission rate is constant or the rate of change of emission rate is small, Equations 3 and 4 may be used to determine the relationship between the boundary and δ. After a few minor algebraic manipulations, it may be shown that:

$$\delta = 1.5\ln\left(\frac{d_e}{d_b}\right) \qquad [5]$$

Equation 5 implies that for a fixed distance between d_e and d_b as the δ-invariant zone is further removed from a source, the value of δ vanishes, and commensurately, for a fixed δ, the size of the δ-invariant zone would be larger. The far boundary point of the δ-invariant zone on the source to sampler axis is calculated from Equations 3 to 5 and after minor algebraic manipulation, it may be shown to be:

$$d_f = \frac{d_e^2}{d_b} \qquad [6]$$

There is a limit to the minimum size of δ. If the concentration at the immediate source is C_s then(10):

$$(d_B)_{\min} = \left[\frac{Cuk}{\dot{m}}\right]^{\frac{2}{3}} \qquad [7]$$

In metric units, $k \approx 0.5\ \text{m}^{1/2}$ based on non-buoyant plumes.(15) The minimum boundary distance should also be used for all S that encloses a source. In the case of multiple sources, the minimum distance for each source would have to be calculated to select the dominant boundary distance, that is, the largest of the minimum boundary distances.

For a single source, the shape of the δ-invariant zone may be easily estimated. Either in a workplace or in the environment, dispersion of contaminants may be conceptually modeled by using the Gaussian turbulent diffusion equation. The application of this equation to define δ-invariant zones should not be mistaken as a rigorous concentration predictive method; rather, it is used to determine the crosswind boundaries of a δ-invariant zone. The crosswind dispersion equation has the same form of the normal distribution equation.(10, 15) By equating the laterally decayed value of concentration with $\exp(-\delta)$ along the straight line that connects the source with the Eulerian sampler, distance d measured along this line may be used to define, the corresponding cross wind distances measured along orthogonal lines:

$$\exp(\eta)\exp\left(-\frac{1}{2}\frac{d_{\text{cross}}^2}{0.16d^{1.5}}\right) = \exp(-\delta) \qquad [8]$$

Therefore:

$$d_{\text{cross}} = \sqrt{kd^{1.5}\ln\left(\frac{d_e^2}{d_b d}\right)} \qquad [9]$$

As an example, the boundary of a δ-invariant zone with specific conditions is calculated. Suppose an Eulerian sampler is located 10 meters away from a source and the δ-invariant zone is specified so that the variation within this zone is within 0.5 to 2 times the Eulerian concentration of the sampler. This implies that δ is 0.693. Using δ and the distance between the source and the sampler in Equation 5, the near source boundary d_b is calculated to be 6.3 meters. Similarly, from Equation 6 the far distance d_f may be calculated to be 15.9 meters. The crosswind distance, as calculated from Equation 9, has an approximate shape of this zone is a truncated oval. One-half of the calculated shape is shown in Figure 2. A Lagrangian average with a path included in this zone is expected to be within the limits specified for the Eulerian Average. If only one source is included in the entire space $S(x, y)$, then and the δ-invariant zone represented by a single Eulerian sampler can be expanded considerably. For this extension, consider a set of virtual Eulerian samplers with overlapping and the δ-invariant zones located at the same distance from the source. Clearly, these samplers are located on a circle around the source with radius d_e. Unless there is a specific reason to believe that the ambient wind velocity has a preferred direction, the circular band around the source with a width $(d_f - d_b)$ is a δ-invariant zone.

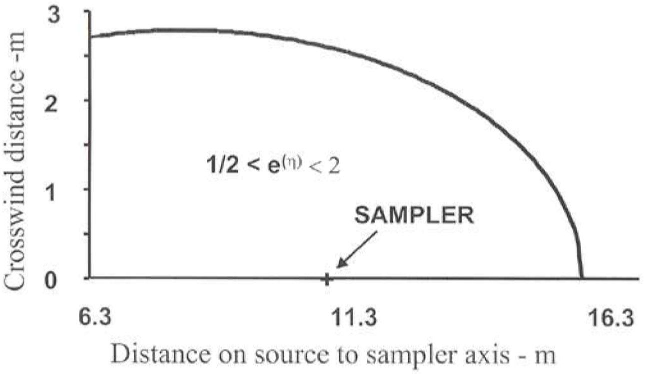

FIGURE 2
The representation of one-half of a δ-invariant zone for a sampler 10 meters from a source with $\delta = \ln 2$.

By a similar argument presented for the extension of a δ-invariant zone, multiple sources may also be modeled by determining the contribution of each source to the Eulerian sampler. However, the availability of the data that defines the contribution of each source to any point over $S(x, y)$ is not necessarily guaranteed nor is the theoretical analysis to estimate that data necessarily available. Because the representation of a combination of multiple sources by a Eulerian sampler would require a knowledge of ambient wind *vector* distribution.(10) The generalized theory of combination of multiple sources included in the space $S(x, y)$ has not been investigated. Intuitively, a scaling method based on source strengths and the distance between each source and the sampler in conjunction with the ambient wind velocity vector distribution may lead to the proper weighting factor. However, the details of the generalization of the theory presented for a single source to multiple sources are not readily apparent. It is important to point out that the correspondence of Lagrangian and Eulerian averages using δ-invariant zones explicitly implies that the Lagrangian and Eulerian averaging times are the same and concurrent. The fallacy that can be introduced by using different averaging times was illustrated by the worker example given previously.

The exposure assignment by time spent in a given zone that is characterized by an Eulerian sampler is based on the inherent and usually tacit assumption that the sampled universe in δ-invariant zone is also time invariant. There is no theoretical or practical *a priori* basis for this assumption. The validation of this assumption requires a sufficient amount of sequential observations that cover the Eulerian averaging period. This can be demonstrated easily. Consider partitioning the Eulerian average into n number of arbitrary but sequential segments:

$$\begin{aligned}\bar{C}_E &= \frac{1}{T}\left[\int_0^{t_1} C \cdot f(c, x_o, y_o, t)\,dt + \int_{t_1}^{t_2} + \cdots + \int_{t_n}^{T}\right] \\ &= \bar{C}_1 + \bar{C}_2 + \cdots + \bar{C}_n\end{aligned} \qquad [10]$$

For a non-zero Eulerian average, the only requirement for these averages is that at least one of them is non-zero. From Equation 10, it ought to be immediately obvious that without knowledge of the time distribution of the contributing concentrations, ascribing equality (either in the exact or in the weak δ sense) between a Lagrangian and an Eulerian average with different sampling times could not be justified. In fact, a careful consideration of this basic relationship leads to a rigorously derivable restriction. Consider a Lagrangian average calculated over a time $\theta <$ Eulerian averaging time. It may be shown that:

THEOREM II: If the δ-invariant zone distribution of the concentration over time is known or can be estimated for an Eulerian sampler, then the Lagrangian average in that δ-invariant zone is bounded by the local minimum and maximum of all δ-invariant zone Eulerian averages with $t = \theta$.

PROOF: This theorem is a direct consequence of the lemma which states that two bounded measurable functions g and h on the real line such that $g \leq h$ then:

$$\int g\,dt \leq \int h\,dt \qquad [11]$$

(Proof of the lemma may be found in reference 16).

DISCUSSION AND CONCLUSIONS

Of course, the implication of the last theorem is directly applicable to activity-weighted exposure estimates. By inductive generalization, it may be shown that the weighted combination of miscellaneous Eulerian averages to obtain a Lagrangian average would place the global maximum and minimum as the limits of the estimate. Within these limits, the calculated weighted average is a number without a particular significance. Therefore, this theorem both proves and defines the strong restriction on the use of Eulerian averages for Lagrangian estimates. The merging of time activity information with general air concentration information to obtain exposure estimates may result in extraordinary errors or may be surprisingly accurate. Without a careful analysis the validity of one or the other can not be assumed. However, the theorem also opens up possibilities to model exposures by determining feasible concentration distributions over time. It must be cautioned that such modeling is not a license to free and open conjecture. Thus, the extrapolation of few area sampling results over years if not decades of Lagrangian averages can not be assumed to provide an accurate estimate of exposure.

In summary, the results of this theoretical investigation demonstrate that without the required sophistication in the analysis of the properties of the sampled environments, haphazard mixing of stationary and personal type samples may result in serious errors. The usual methods of combining the stationary and personal results such as activity distribution weighing of stationary type samples are based on assumptions that may not be justifiable. The theoretical development reported also opens up possibilities for the development of potentially productive estimation methods, such as Bayesian improvement of estimates with post facto validation.

Exposure assessment, exposure estimation, and characterization of complex interactions in a complicated environment are not necessarily easy tasks and there is no reason to expect them to be so. The problems associated with the exposure estimation process can be further complicated by improvements made in the measurement techniques and "after the fact" alteration of their intended use. The allure of projecting the refinement achieved by instrumental improvements to extant data, which was obtained by less refined methods, can create a false sense of precision for that data set. The existence of data intended for uses that have little to do with exposure assessment may give a false sense of sufficiency of data. The following quotation has an important bearing on these pitfalls:

> "A serious difficulty with happenstance data is that, when we establish a significant *correlation* between y and $x1$, this does not provide evidence that these variables are necessarily *causally* related."(17)

Much can be learned through rigorous models and their independent validation based on good physicochemical principles and methods. At times, realizing that the potential errors involved in the assumed quantification of historical exposures may be too extensive, abandoning quantification may be justifiable. Under conditions where sparse area samples are the only source of data for estimating exposure, it might be more appropriate to characterize exposures by simple qualitative ranks. For reconstruction of historical exposures this may be the scientifically tenable route to take.

It is clear from the exceptional effort that usually accompanies the development of exposure estimates in the face of limited data that this process is difficult and fraught with uncertainty. Most often, the available data are so sparse as to be of limited usefulness. Although this is the typical case, it is clear from this research that making use of limited data with the stated limitations can lead to seriously misleading conclusions.

There are, however, potential solutions to these difficulties. Current and historical research in this area is limited, but there are new opportunities to shed light on these difficult problems. One such approach would be to investigate the relationship between stationary and personal samples under conditions that are representative of the historical process under study or the development of mathematical relationships under current conditions that have a definable relationship to historical conditions. Even though there are no pat or ready answers to the difficult questions associated with development of accurate historical exposure estimates, this does not mean that these issues should be ignored and, therefore, result in historical exposure estimates that have an undefined relationship to reality. The potential for creative research in the application of Bayesian techniques for data enhancement, adaptation of economic forecasting techniques to retrospective forecasting, and the treatment of sparse data by missing data modeling techniques should be avenues to be explored in the future.

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