



Exposure efficiency: an idea whose time has come?

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Abstract

Exposure efficiency, the fraction of material released from a source that is eventually inhaled or ingested, is arguably the simplest of all possible descriptions of the link between pollutant emissions and population exposures. This paper, prepared in late 1999 for the SGOMSEC Workshop, notes that several groups of researchers independently developed the concept of exposure efficiency in the late 1980s and early 1990s but argues that the potential importance of exposure efficiency in risk analysis and life cycle assessment has only recently been appreciated. The paper reviews the history of the concept; discusses and summarizes previous estimates of exposure efficiency for particulate matter and other air pollutants; presents new values for fine particulate matter emitted from power plants and mobile sources in the United States; and illustrates how preliminary estimates of exposure efficiency might be developed. The authors assert that in order for the concept of exposure efficiency to achieve its full potential exposure efficiency estimates for a wide variety of pollutants and sources must be developed and that both the results and methods must be made widely available and accessible to the community of risk assessors and life cycle analysts.

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“All models are wrong, but some are useful.”—George Box

1. Introduction

The development of sound environmental health policy requires both scientific information about the linkages between pollutant emissions and human health effects and value judgments about the importance of these health effects relative to other social concerns. Since the 1983 publication of the NAS Report “Risk Assessment in the Federal Government: Managing the Process,” the terms risk assessment and risk management have been used to differentiate these two foundations of environmental health policy (NAS, 1983). Risk assessment is commonly viewed as involving four elements—hazard identification, exposure assessment, dose–response analysis and risk characterization. In this manuscript, our focus is on exposure assessment in support of decision making and policy analysis.

Over the past two or three decades, a great deal has been learned about the fate and transport of contaminants in various environmental media; the transfer of pollutants from one medium to another; the pathways by which people are exposed to pollution; the activity patterns of individuals and the influence of these on exposure; and the distribution, metabolism and elimination of pollutants from the human body. The growth of information has been so dramatic that we seem to collectively know so much that individually we know almost nothing—i.e., we cannot see the forest for the trees.

At the same time, the world is experiencing rapid population growth, industrialization and urbanization and the environmental health problems that more often than not accompany such growth. Currently, there are more than 350 cities around the world with populations over 1 million, and 22 with populations greater than 10 million (Brinkhoff, 1999). The need for rational environmental health policy is expanding rapidly at the same time that the demand for quality and rigor in the scientific information provided in support of such policies is continually increasing.

In this paper we argue that unless approaches for more effectively organizing and utilizing scientific information are found, critical environmental health policy decisions will be delayed so long that they will be ineffective. Specifically, we argue that much of the exposure assessment needed to support environmental health policy can rely on relatively simple measures of exposure, and more generally, we believe that it might be wise to adopt approaches for risk assessment by analogy.

2. The concept of exposure efficiency

Exposure efficiency is defined as the fraction of material released from a source that is eventually inhaled or ingested (Harrison et al., 1986; Smith, 1993a,b; Phonboon, 1996).^{1,2} It is arguably the simplest of all possible descriptions of the link between source emissions and population exposures.

Of course, beneath this simplicity lies complexity which must be understood to meaningfully interpret and apply the concept. The probability that a molecule released from a source will eventually be inhaled or ingested by a human receptor is obviously dependent on features of the source, the pollutant, the medium (or media) to which it is released, and the receptor. For example, when we consider the exposure efficiency for pollutants released to the atmosphere we must account for the location and conditions of release (stack height, stack diameter, stack gas temperature and exit velocity), conditions of the atmosphere (wind speed, wind direction, atmospheric stability), properties of the pollutant (its phase (gas, vapor, particulate matter), reactivity, solubility), and of the receptors (location, population density, activity).

Clearly then, there is no universal single value of exposure efficiency, especially at the level of the individual molecule. Ultimately each individual molecule is either inhaled or not. We cannot hope to make deterministic predictions at this level of analysis. What we can estimate is the fraction of all molecules released from a specific source that will be eventually inhaled or ingested. We may also explore the influence of properties of the source, pollutant, media and receptor that influence these probabilities. And perhaps we can develop information on the dynamics of the process and the geographic distribution of results.

¹ Although our definition is restricted to terms which involve a dimensionless ratio of intake to emissions, such as Harrison or Phonboon’s “exposure efficiency” or Smith’s “nominal dose effectiveness;” a number of other terms such as exposure or dose effectiveness, exposure factor, and population dose commitment have been used to refer to closely related measures of population exposure per unit emissions.

² Authors’ note added in proof—Since this paper was written, a working group including two of the authors (Smith and Evans) of this paper have proposed that the term “intake fraction” be used rather than the term “exposure efficiency” (Bennett et al., 2002).

3. History of the concept

Our reading of the literature suggests that several research groups have independently developed the concept of exposure efficiency.

In the early 1990s, a group of us at the Harvard School of Public Health were interested in developing simple approaches for characterizing uncertainty and variability in the fate and transport of pollutants for use in risk and decision analysis. We realized that a simple measure of the ratio of population exposure to the source emissions would be useful in our analysis of the value of improved information on perchloroethylene exposures from drycleaners in the US (Thompson, 1995; Thompson and Evans, 1997; Evans et al., 2000). At about the same time, Phonboon was working with our group on an analysis of population exposures and risks from refinery emissions of benzene, sulfur oxides and particulate matter in Bangkok, Thailand (Phonboon, 1996). Again, an exposure efficiency based approach was used to characterize the population exposures needed in support of his risk assessment. As we began to talk with colleagues about this idea, we learned that several other research groups had independently come to the same realization.

In a 1986 technical report to the US EPA, Harrison, Hattis and Abbat had defined exposure efficiency as “the fraction of total production which is likely to reach people, or the ratio of human intake to the amount emitted” (Harrison et al., 1986). Their interest was in using exposure efficiency in support of the development of a “use-class based priority setting system” for toxic chemical regulation in the US. However this early work on exposure efficiency was never published in the peer reviewed literature and thus has not been widely appreciated.

In the mid 1990s, Jolliet and Crettaz became interested in improving the treatment of health risk assessment in life cycle analysis. As an element of this work on life cycle analysis, Jolliet and Crettaz developed estimates of exposure efficiency for 50 compounds. Initially they used different terminology—referring to the estimates as “fate factors,” which they defined as the factor “which enables the conversion of the emission flow into its related concentration increase” (Jolliet and Crettaz, 1996). Jolliet and Crettaz’s work is important because it was not confined to analysis of air pollutants, but examined exposure efficiency for soil, water and air. Further, at least as early as 1994, Jolliet recognized that the fate factor was proportional to the ratio of the residence time divided by the dilution volume (Jolliet, 1994).

Smith and his colleagues have been active in this area for nearly 15 years and have contributed substantially to the development and application of various measures of exposure efficiency. In the late 1980s, Smith (1987) adapted the term “population dose commitment” as

used in the radiation literature (UNSCEAR, 1977) to the field of air pollution and showed how the concept could be used to compare the dose per unit activity from various activities. Smith (1987) was the first air pollution researcher to recognize that in the late 1970s there had been efforts by Lindell (1978a,b) and Bennett (1981) to show how the principles developed for radiation protection could be applied to non-radioactive pollutants.

In a series of papers (including Smith (1988a,b), Roumasset and Smith (1990), Smith (1993a,b) and Wang and Smith (1999a,b)), conference proceedings (including Smith and Edgerton (1989)), working papers and reports (including Smith et al. (1993), Smith (1994), Smith (1995)) and book chapters, Smith and his colleagues: (i) argued that “evaluation by exposure will not only reorder the ranking of major outdoor emissions sources, but will reveal an entirely different landscape of sources; those that may significantly affect exposure without appreciably affecting ambient concentrations” (Smith, 1988a); (ii) noted the potential regulatory and economic superiority of “exposure trading” to “emissions trading” (Roumasset and Smith, 1990); (iii) showed how the concept could be applied to comparison of the global warming from different greenhouse gases (Smith and Ahuja, 1990); (iv) proposed how it might be used for regulating air quality (Smith, 1995) and (v) used the approach to illustrate the health co-benefits of greenhouse gas controls in China (Wang and Smith, 1999b).³ In her dissertation, Tsai (a student of Smith’s) used the concept to explore the relative cost-effectiveness of particle control methods in Los Angeles (Tsai, 1999).

In 1990, Roumasset and Smith used the term “exposure factor,” defined as the ratio of “total population exposure ($\mu\text{g-person-year}/\text{m}^3$)” to “total emissions (tons).” And in 1993 Smith used the terms “exposure effectiveness” to refer to “the fraction of released material that actually enters someone’s breathing zone as measured in exposure units,” and “nominal dose effectiveness” to mean “the ratio of emitted to inhaled material” (Smith, 1993a). Here nominal dose effectiveness was given in units of grams of exposure per metric tonne of emissions and is essentially identical to Harrison, Hattis and Abbat’s original definition of “exposure efficiency.”⁴

Perhaps most importantly, in 1995 Smith noted that “what is needed... are sufficient data to establish exposure effectiveness factors for major source classes”

³ Smith and Edgerton noted that Nichols (1984) had discussed the concept of exposure-based regulation of air pollution.

⁴ Smith notes that the US population exposure to particulate matter from combustion of 1 ton of coal in a (controlled) power plant is the same as the exposure due to the environmental tobacco smoke from just 10 cigarettes.

(Smith, 1995). In this manuscript we review the progress that has been made toward this goal; consider the strengths and weaknesses of the work that has been done; and suggest avenues of research that could contribute to realization of the full potential of exposure efficiency as a tool for risk assessment and regulation.

4. Approaches for estimating exposure efficiency

There are two basic approaches for the analysis of exposure efficiency. They differ primarily in the way ambient concentrations are computed. Most estimates of exposure efficiency have relied on fate and transport models to estimate the impact of specific sources on ambient concentrations. However some have relied on direct measurements of ambient concentrations or on source–receptor models to apportion measured ambient concentrations to specific sources.

We focus on the approaches which use fate and transport modeling to estimate source impacts. Among these, a further distinction can be made with regard to the approach used for modeling. Some analysts have relied on compartmental models, which represent media as well-mixed compartments and yield estimates of average concentrations within each compartment. Others have relied on more traditional site specific fate and transport models, such as the Gaussian air pollution model, which examine the spatial heterogeneity of concentration fields in some detail.

Below we briefly review the basic elements of fate and transport modeling which are necessary to understand exposure efficiency. We look only at problems involving air pollution, but the general ideas would apply to analysis of other media.

4.1. Compartmental modeling

The compartmental modeling approach may involve a single compartment or a set of linked compartments. The simplest case involves a single compartment. By considering exposure efficiency in this simple one compartment system, we can begin to understand the basic relationships that govern exposure efficiency in more complex systems.

Consider a one box model of air pollution in Boston. Represent the air over the city of Boston as one well-mixed box with a square base of area A (m^2), a mixing height L (m), and a wind blowing perpendicular to one of the sides at a constant velocity u (m/s). Into this air, a conservative pollutant is emitted at a constant emissions rate Q (g/s). Under these circumstances, the equilibrium concentration, C (g/m^3), of the pollutant in the air over Boston is:

$$C = Q/(u * L * A^{1/2}) \quad (1)$$

Once the equilibrium concentration has been determined, the exposure efficiency is simple to compute.

Exposure efficiency, ε (dimensionless), is defined as I/Q , where I is the intake of the pollutant (g/s) and Q is the emissions rate (g/s). To compute the intake, I , we must know how many people are exposed, N , and how much air they breathe. If B (m^3/s) is the nominal breathing rate, then the pollution intake of the exposed population is simply:⁵

$$I = C * B * N \quad (2)$$

Thus, the box model estimate of exposure efficiency is:

$$\varepsilon = I/Q = (C * B * N)/Q \quad (3)$$

But since C is proportional to Q , by substituting $Q/(u * L * A^{1/2})$ for C in (3) we obtain:

$$\varepsilon = (B * N)/(u * L * A^{1/2}) \quad (4)$$

This expression is attractive because it makes clear that exposure efficiency is simply the ratio of the air breathed ($B * N$) to the air available for dilution and removal of the pollutant ($u * L * A^{1/2}$). An obvious implication of this formulation is that pollutants released into small volumes (indoors) will typically have higher exposure efficiencies than pollutants released into large volumes (outdoor air).

One problem with this approach for computing exposure efficiency may now become apparent, i.e., that the estimate of exposure efficiency is related to the surface area of the box, A . It might seem that increasing the size of the box would reduce the estimate of exposure efficiency, but actually as the size of the box is increased the estimate of exposure efficiency tends to increase because the size of the exposed population is also related to the size of the box.

Consider the simplest case of uniform population density. In this case, the exposed population, N , is the product of the surface area, A (m^2), and the population density, ρ (persons/ m^2). By substituting this expression into Eq. (4), one obtains:

$$\varepsilon = (B * \rho * A^{1/2})/(u * L) \quad (5)$$

Obviously our estimate of exposure efficiency derived using this simple box model is intimately tied to our choice of the scale of analysis. This disquieting result is an artifact of our initial assumption that the pollutant is conserved. While over short distances and travel times,

⁵ Typically breathing rates are given in m^3/d , with nominal values on the order of 20. These must be converted to m^3/s by dividing by 86 400 s/d to obtain units that are compatible with those used to characterize emissions rates.

the assumption may be approximately valid for some air pollutants, it becomes less and less realistic as longer distances and travel times are considered.

Pollutants are lost from the atmosphere by deposition, impaction, decay, reaction and a host of other mechanisms. The physical and chemical mechanisms may be quite complex and highly dynamic. But to begin to understand the impact of these processes on exposure efficiency, one can consider the simple case of first order losses, characterized by a rate constant, k (1/s) and a related atmospheric half-life, $t_{1/2}$ (s).

By accounting for the atmospheric half-life of the pollutant, $t_{1/2}$ (s), we can begin to address this issue. We immediately recognize that if we are going to capture the full exposure efficiency the box must be large enough to allow most of the pollutant to deposit, decay or react. In a simple first order exponential decay process:

$$C_t/C_0 = \exp(-\ln(2) * (t/t_{1/2})) \quad (6)$$

approximately 4 half-lives must elapse before 95% of the pollutant has deposited, reacted or decayed and nearly 7 half-lives must elapse before 99% of the pollutant is lost from the air mass. This expression can be rewritten in terms of a rate constant, k , which is simply $-\ln(2)/t_{1/2}$, as:

$$C_t/C_0 = \exp(-k * t) \quad (7)$$

The mean atmospheric residence time, μ is then $1/k$ or $t_{1/2}/0.693$.

To develop any practical sense of the scale of analysis necessary to reflect the true cumulative exposure efficiency, we must consider the atmospheric half-life of the pollutant of interest and the typical wind speeds that are involved in atmospheric transport. For airborne particles, the mean residence time is a strong function of the particle size—ranging from perhaps 2–5 d for particles with aerodynamic diameters of 2.5 μm (and terminal settling velocities on the order of 2×10^{-2} cm/s) to several weeks for particles with aerodynamic diameters of 0.1 μm (and terminal settling velocities less than 1×10^{-4} cm/s) (Jaenicke, 1980).⁶ For gases, the mean residence time depends on their reactivity with other air pollutants, their solubility in water, and other physical–chemical properties. The mean residence time of sulfur dioxide in the atmosphere is thought to be between 1

and 3 d (Schwartz, 1989).⁷ Some of the volatile organic compounds, such as perchloroethylene, are quite persistent in the atmosphere, with mean residence times on the order of 100 d or more.⁸

If we consider the case of a pollutant with a half-life on the order of one day and a wind velocity of 2 m/s (about 170 km/d) we can see that ranges of 500–1000 km from the source may need to be considered to reflect actual exposure efficiencies. For pollutants with half-lives on the order of 100 d, global transport must be considered. And for pollutants with half-lives on the order of a few hours, the spatial range of interest may be relatively small.

In cases where it is necessary to consider large spatial ranges, it is important to use values of population density that are appropriate to the spatial range. The overall world population density is on the order of 10 persons per km^2 , but at a local scale is highly variable—with 60% of the land surface of the Earth unpopulated and population densities as high as 160 000 per km^2 in a portion of the Mong Kok region of Hong Kong.⁹ Most people (about 90%) live in the Northern hemisphere. On the continental scale, population densities vary from on the order of 15 persons per km^2 in North America to about 150 persons per km^2 in Asia and Europe (CIA, 1998). Within many large cities, population densities may be much higher than this. For example, in several sections of Bangkok, Thailand population densities are 20 000 persons per km^2 or higher, and throughout the Bangkok Metropolitan Area population density is typically between 1000 and 10 000 persons per km^2 . Thus, when we consider sources located in or near urban areas more of the exposure efficiency may occur close to the source than would be expected on the basis of the rate of concentration decay.

⁷ For nitrates, although both gas phase and aqueous phase reactions are possible, atmospheric formation of particulate nitrate is dominated by gas phase reactions. Secondary nitrate particles can form only at low temperatures characteristic of winter conditions. In some applications, estimated nitrate concentrations have been reduced by a factor of four to reflect that these conditions generally prevail in 3 of 12 months.

⁸ Of course, the instantaneous rate constants are highly variable and depend on a number of factors. For SO_2 , both gas phase and aqueous phase reactions are important. Peak gas phase reaction rates of 1–5% per hour are not uncommon during the daylight and in the summer. In the winter or at night, gas phase reaction rates are approximately an order of magnitude lower. Aqueous phase reactions can occur at rates as high as 10% per hour and are thought to be the dominant mode of sulfate formation, being responsible for perhaps 80% of all sulfate formation.

⁹ The population density of the Earth of about 10 persons/ km^2 is based on the entire surface area of the Earth. If restricted to the land area, the population density of the Earth is about 40 persons/ km^2 .

⁶ The terminal settling velocity of a 2.5 μm aerodynamic diameter particle is on the order of 2×10^{-2} cm/s. The terminal settling velocity of an 0.1 μm particle is less than 1×10^{-4} cm/s. However the actual dry deposition velocity of the 0.1 μm particle may be much larger than this because of losses due to impaction and diffusion.

4.2. Site-specific modeling—Gaussian models

The other modeling-based approach typically relies on Gaussian air pollution models to estimate the ambient concentrations of air pollutants near the source of interest, and couples these concentration estimates with census data on population densities within the region of interest.

In the Gaussian air pollution model, the concentration of a conservative pollutant at a point x, y, z in the downwind direction from a point source located at $0, 0, H$ is given by:

$$C(x, y, z) = (Q/u) * (1/((2\pi)^{1/2}\sigma_y)) \\ * \exp((-1/2) * (y/\sigma_y)^2) * (1/((2\pi)^{1/2}\sigma_z)) \\ * [\exp((-1/2) * ((H - z)/\sigma_z)^2) \\ + \exp((-1/2) * ((H + z)/\sigma_z)^2)] \quad (8)$$

The orientation of the x axis in this expression is defined by the wind direction. In this expression the influence of atmospheric turbulence is reflected in the σ_y and σ_z terms which are themselves functions of the distance downwind from the source, x . The specific form of the Gaussian equation given in (8) assumes that the (conservative) pollutant is perfectly reflected from the ground surface and is appropriate for continuous releases. If puffs are of interest rather than continuous releases, then it becomes important to account for dispersion in the x direction and to use values of the dispersion coefficients appropriate for puffs. Many excellent references, such as Boubel et al. (1994), provide more complete descriptions of the Gaussian model.

If, as in most exposure assessment, ground level concentrations are of interest, we can set $z = 0$ in (8) and by collecting terms, obtain a much simpler expression:

$$C(x, y, 0) = (Q/(u * \pi * \sigma_y * \sigma_z)) * \exp((-1/2) \\ * (y/\sigma_y)^2) * \exp((-1/2) * (H/\sigma_z)^2) \quad (9)$$

It is important to note that as we move downwind from the source, at some point the vertical dispersion of the plume becomes limited by the mixing depth of the atmosphere, L . This is commonly handled by replacing the Gaussian vertical dispersion term, $(1/((2\pi)^{1/2} * \sigma_z)) * \exp((-1/2) * (H/\sigma_z)^2)$, by the term, $1/L$, reflecting complete mixing in the vertical dimension.

To reflect the impact of varying wind directions, wind speeds and atmospheric stability conditions, this expression is evaluated for a variety of wind directions, wind speeds and atmospheric stability conditions. Then if long term average impacts (e.g., annual means) are of interest, the resulting concentration estimates are weighted by the relative frequency with which these various conditions occur. One additional modification

of the model is often used when long-term average concentrations are of interest—i.e., sector averaging. In a sector-averaged Gaussian model, the lateral dispersion term, $(1/((2\pi)^{1/2}\sigma_y)) * \exp((-1/2) * (y/\sigma_y)^2)$, is replaced by $1/(2\pi x/n)$, reflecting uniform dispersion across the lateral extent of each of the n sectors. Commonly sixteen 22.5° sectors are used, and so the lateral dispersion term becomes $(8/\pi x)$.

To compute exposure efficiency, it is necessary to account for the variation in concentrations experienced at various locations relative to the source and to weight these by some measure of the number of people likely to breathe the air at those locations. Typically, this is done by weighting the estimates of ambient concentration from the Gaussian model, by the population living in each region.

Using this approach the pollution intake of the exposed population, I (g/s), is computed as:

$$I = \sum_j [P_j * C_j * B] \quad (10)$$

where j is an index of receptors, P_j is the population living near receptor j (people), C_j is the average estimated pollution concentration at receptor j given by the Gaussian model (g/m^3), and B is the breathing rate (m^3/s per person). In this formulation the influence of the scale of analysis on the resulting estimates of exposure efficiency is not as obvious as it was in the compartmental modeling approach. However, the choice of scale is still important.

The Gaussian air pollution model gives valid estimates of concentration as long as one is neither too close to the source (where building wake effects can be important) nor too far from the source (where the assumptions of constant wind speed, direction and atmospheric stability begin to break down). Most authorities on the Gaussian model recommend that it be used to estimate concentrations within 50 km of the source. Clearly, this raises concern about the appropriateness of using Gaussian models to estimate exposure efficiencies for pollutants with long atmospheric half-lives. Analysis of long range transport can be accomplished using models, such as CALPUFF, which combine elements of trajectory and Gaussian models (US EPA, 1995).

5. Published estimates of exposure efficiency

Estimates of exposure efficiency have been published for a number of compounds, e.g., particulate matter, various volatile organic compounds, some semi-volatile organic compounds and a few selected metals. The estimates are difficult to directly compare because of differences in the methods that have been used, the conditions assessed, the domains of interest, the values assigned to key parameters and the divergent purposes of the underlying analyses. Most of these estimates have

been derived using Gaussian air pollution models to analyze local exposures (within 20–100 km of the source) and compartmental models to examine exposure efficiencies very close to the source (e.g., indoors) and very far from the source (e.g., regional or global). Below we briefly review the efforts of various researchers to estimate exposure efficiencies and describe the approaches used and the values assigned to key variables. Our focus in this review is on estimation of exposure efficiency for airborne particles, but we include papers which have dealt with volatile organic compounds and other air pollutants because many of the issues central to estimation of exposure efficiency are not pollutant specific.

Harrison et al. (1986) provided two sets of estimates of exposure efficiency for thirty-five compounds—including a number of volatile organic compounds, some semi-volatile organic compounds, and a few metals—under review by the US Environmental Protection Agency as potentially hazardous air pollutants under the NESHAPS program. One set of estimates examined the local exposure efficiency (within 20 km of the source) and the other looked at exposure efficiencies on a regional scale (within 1500 km of the source). The set of local exposure efficiency estimates was based on detailed atmospheric fate and transport analysis for 311 major chemical manufacturing or consuming plants; 62 categories of smaller point sources; and 77 area source categories in each of the 248 large urban areas and in 243 smaller cities (Anderson, 1980, 1983). Anderson's estimates of population exposure (person- $\mu\text{g}/\text{m}^3$) were converted to exposure efficiencies by multiplying them by the nominal breathing rate of 20 m^3/d . The set of regional exposure efficiency estimates was derived by multiplying the local exposure efficiency estimates by a factor reflecting the atmospheric half-life of each pollutant and the expected increase in the potentially exposed population as one moves from the local to regional scale. Urban population densities at the time were on the order of 1200 people/ km^2 (within 20 km of city centers) and the national average population density was approximately 20 people/ km^2 . The resulting estimates of local exposure efficiency, which varied from 1.4×10^{-7} (allyl chloride) to 1.7×10^{-5} (carbon tetrachloride), represent emissions-weighted averages characteristic of the specific mix of sources, source classes, and emissions controls prevalent in the US during the late 1970s. Differences in the rate of reaction or decay are not thought to be important determinants of local exposure efficiencies for these compounds. Under typical meteorological conditions the relatively low first-order losses would not be expected to result in appreciable (>30%) losses of any of the compounds within 20 km of the source. Thus it would seem that most of the 100-fold variation in local exposure efficiencies is due to differences in the characteristic locations and nature of emissions sources across the set of compounds consid-

ered. The regional estimates of exposure efficiency were somewhat larger, with the magnitude of the difference determined by the atmospheric half-life. The largest increases were seen for compounds with relatively long atmospheric half-lives (e.g., propylene oxide (6 d), *o*-dichlorobenzene (30 d), and chloroform (80 d)). However, the ratio of regional to local exposure efficiency never exceeded 2.15. This upper limit of the correction factor reflects the geometry of and the population densities in the two regions (i.e., local and regional) and for pollutants with half-lives longer than 6 d is insensitive to half-life. For these long-lived atmospheric pollutants most of the pollutant is still present as the air leaves the outer limit of the 1500 km radius.

Harrison, Hattis and Abbat's estimates of local and regional exposure efficiency for the three metals in the study (beryllium, nickel and manganese) treated these as fine particles with atmospheric half-lives of 4.7 d. For all three metals, the resulting estimates of local exposure efficiency were on the order of 1×10^{-5} and the estimates of regional exposure efficiency were twice this large.

Smith (1993a,b) published estimates of exposure efficiency for several sources of airborne particulate matter including—coal-fired power plants in the US and in less developed countries (LDC), vehicles, neighborhood sources, stoves vented indoors and outdoors, environmental tobacco smoke and mainstream cigarette smoke. Smith's exposure efficiency estimates for particles from coal-fired power plants in the US focused on regional exposure efficiency (within the continental US) and were derived from Rowe's estimate of the population exposure likely to result from 86 hypothetical 1000 MW coal-fired power plants uniformly spaced across the US.¹⁰ The estimated 1985 US population of each of the 243 air quality regions was used to derive population-weighted exposure estimates. Using the Pacific Northwest Laboratory long-range transport model, Rowe (1981) had estimated that in the US each English ton (2000 lb) of annual emissions resulted in approximately 100 person- $\mu\text{g}/\text{m}^3$ year of exposure to particles.¹¹ Rowe's analysis considered only primary particles, but accounted for

¹⁰ Rowe (1981) examined the contribution of local (within air quality control region) exposure to national exposure from each power plant and also evaluated plant-to-plant variability in population exposure per unit emissions, but Smith did not carry these details forward into his exposure efficiency calculations.

¹¹ Actually Rowe (1981) looked at the distribution of population exposure per unit emissions (person- $\mu\text{g}/\text{m}^3$ per English ton (2000 lb)) and stated that the median value of the distribution of such values was 92.6 person- $\mu\text{g}/\text{m}^3$ per English ton and that these values varied (5–95th percentiles of the cumulative distribution) from 16.3 to 314 persons- $\mu\text{g}/\text{m}^3$ per English ton. Smith reexpressed Rowe's estimate as 100 person- $\mu\text{g}/\text{m}^3$ per metric tonne (1000 kg).

both dry deposition (using a terminal settling velocity of 0.2 cm/s (characteristic of a 10 μm particle)) and wet deposition (by means of an empirical relationship which accounts for intensity of rainfall). Using a breathing rate of 28 m^3/d Smith was able to convert Rowe's estimate of population exposure into an estimate of exposure efficiency.¹² Smith's exposure efficiency estimate for coal-fired power plants in LDC was derived by multiplying the US value by a factor of 9 intended to account for the greater population densities typical of developing countries.¹³ His exposure efficiency estimates for indoor sources are semi-empirical. For example, Smith's estimate of the exposure efficiency for indoor emissions of environmental tobacco smoke relies on Dockery and Spengler's observation that smoking 1 cigarette per day indoors results in a 1 $\mu\text{g}/\text{m}^3$ increase in indoor concentrations of particulate matter (Dockery and Spengler, 1979).¹⁴ Smith's exposure efficiency estimates for emissions from outdoor sources vary from 1×10^{-6} for coal-fired power plants in the US to 1×10^{-5} for coal-fired power plants in LDC, 2×10^{-5} for motor vehicles and 4×10^{-5} for neighborhood sources. His estimates of exposure efficiencies for indoor sources are orders of magnitude greater, ranging from 3×10^{-3} for stoves vented indoors and indirect exposure to passive cigarette smoke to 1 for direct inhalation of mainstream cigarette smoke.

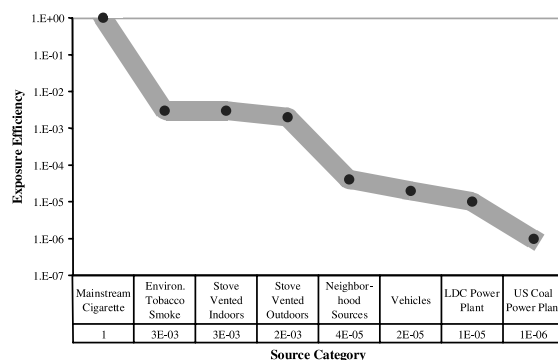
Smith's results are summarized in Fig. 1. Smith noted that indoor cook stoves are 1000 times more effective than US coal-fired power plants in delivering smoke to humans. He pointed out the potentially significant implications of this finding for developing cost-effective policies for the controlling population exposures to airborne particulate matter. Of course, the inference that these results imply a 1000-fold difference in efficiency of health risk reductions relies on the assumption that the toxicity of the particles emitted from cook stoves and from power plants is not appreciably different. It is important to note that, by themselves, estimates of exposure efficiency are incomplete proxies for risk. Differences in toxicity must be considered in any risk assessment and differences in cost of control must be addressed in any cost-effectiveness or cost-benefit analysis.¹⁵

¹² Smith noted that 28 m^3/d was toward the high end of breathing rates appropriate for adult males.

¹³ The ratio of the population densities of India and the US at the time was 9.

¹⁴ In this calculation, Smith uses an emissions factor of 24 mg/cigarette and an indoor occupancy of 2.5 persons.

¹⁵ Any decision about the appropriate degree of control for these sources would need to consider (either explicitly or implicitly) the costs and benefits of the feasible controls for each source and any important differences in the attributes of the risks involved—e.g., voluntary or involuntary, certain or uncertain, familiar or unfamiliar, etc.



Adapted from Smith (1993). Values are expressed as exposure efficiencies, derived by dividing nominal dose effectiveness (given as grams inhaled per metric ton emitted) by a conversion factor of 10^6 grams per metric ton. Smith noted that the values were intended to be illustrative and that the width of the shaded band was intended to suggest uncertainty in the estimates.

Fig. 1. Exposure efficiency estimates for particles from various emissions sources (Smith, 1993a,b).

Phonboon (1996) computed estimates of exposure efficiencies for particles, SO_2 and benzene from an oil refinery in Bangkok, Thailand. He examined both local (within 50 km) and regional (within 1000 km) scales. For the local analysis, concentration estimates were computed using ISCLT2, a sector-averaged Gaussian model.¹⁶ For the regional analysis, concentrations were estimated using a simple spreadsheet-based sector-averaged Gaussian model. In both models, first-order losses were computed using half-lives of 8 h for particles, 4 h for SO_2 and 48 h for benzene. Approximately 5 million persons live within 20 km of the source (3997 persons per km^2), 7 million live within 50 km (923 persons per km^2) and 160 million live within 1000 km (52 persons per km^2).¹⁷ One aspect of Phonboon's approach which is novel is his use of breathing rates appropriate for the

¹⁶ Meteorological data were taken from a meteorological observation station 2 km from the refinery. Observations of wind speed, wind direction and cloud cover taken each 3 h were used to construct frequency distributions of wind speed and direction. Atmospheric stability classifications were determined using Pasquill's method (US EPA, 1992). No data on mixing height were available, so Phonboon used mixing height data from a station in Florida with similar climate. This procedure resulted in estimated mixing heights of 1800 m (A stability), 1200 m (B or C stability), 1000 m (D stability), and 900 m (E or F stability). ISCLT default assumptions about pollutant half-lives were used in Phonboon's base case analysis. Population data for the Bangkok Metropolitan Area and for Thailand and other countries within 1000 km of the source were taken from the Thai Census of Housing and Population (National Statistical Office of Thailand, 1992), Population Projections for Thailand (National Economic and Social Development Board of Thailand, 1991), and The World Resources Institute (1994).

¹⁷ Note that within 1 km of the source the population density is extremely high—on the order of 25000 persons per km^2 .

body weights of the Thai population. Using Layton's method, Phonboon derived age-specific rates between 4.4 m³/d (infants) and 11.8 m³/d (young adults), which yield an average breathing rate for the entire Thai population on the order of 9.5 m³/d—about half of the nominal value of 20 m³/d (Layton, 1994). Phonboon's estimates of local exposure efficiency were 1.4×10^{-5} for both particles and SO₂ and 4.3×10^{-5} for benzene. He reported that exposure efficiency did not increase significantly beyond 50 km. This was attributed to the relatively short half-lives of the pollutants, the extremely high population densities in the immediate vicinity of the source and the relatively low stack heights at the refinery.

Phonboon was interested in the dependence of his exposure efficiency estimates on the scale of analysis and was concerned about the sensitivity of his results to assumptions about the atmospheric half-life of particulate matter. Fig. 2 illustrates that as half-life is varied from 1 to 72 h exposure efficiency estimates for particulate matter increase by about a factor of two from 7×10^{-6} to 1.6×10^{-5} . An interesting feature of Phonboon's sensitivity analysis is the marked asymmetry—i.e., exposure efficiency decreases by a factor of 2 as $t_{1/2}$ is reduced from 8 to 1 h, but only increases by 20% or 30% as $t_{1/2}$ is increased from 8 to 72 h. As noted above, in Bangkok virtually all of the exposure efficiency occurs quite near the source.

Jolliet and Crettaz have developed semi-empirical estimates of atmospheric fate factors for a number of compounds, including particulate matter. The atmospheric fate factor, F^{aa} (y/m), is the ratio of residence time to mixing height. Rather than relying on environmental fate and transport models, Jolliet and Crettaz use the ratio of the near surface atmospheric concentration (g/m³) to the emissions flux (g/m²y) to estimate atmospheric fate factors. Using emissions and concentration data for Europe, Jolliet and Crettaz's empirical estimate of the fate factor for particles is 8.5×10^{-6} y/m. Expo-

sure efficiencies are directly proportional to fate factors and may be found by multiplying the fate factor by breathing rate and population density. Using a nominal breathing rate of 20 m³/d and Jolliet and Crettaz's estimate of global population density (11 persons/km²), yields an estimate of particulate exposure efficiency of 7.5×10^{-7} .

On the basis of similar calculations for a variety of other compounds, Jolliet and Crettaz note that for compounds with atmospheric residence times greater than 60 d, the effective mixing height is approximately 10 km. However, for compounds with residence times less than 60 d, the effective mixing height is limited and can be estimated by the empirical relationship $L = 30088 \times \tau^{0.61}$ where L is in units of m and residence time τ is given in year. This yields an empirical expression in which exposure efficiency can be estimated directly from a compound's atmospheric half-life. Using an atmospheric half-life of 7.6 d for particles yielded an effective mixing height of about 4000 m and an exposure efficiency of 6.8×10^{-7} .

Thompson examined both local and global exposure efficiencies for perchloroethylene emitted from dry cleaners in the US. She estimated the exposure efficiency for each of 100 commercial dry cleaners selected randomly from the membership of a dry cleaning trade association (Thompson, 1995; Thompson and Evans, 1997; Evans et al., 2000). For each chosen site, local exposure efficiency was computed using the EPA's Human Exposure Model, HEM, version II. HEM is a sector-averaged Gaussian dispersion model coupled with block level population data from the 1990 US Census. It relies on meteorological data from the nearest stability array (STAR) site. A nominal breathing rate of 20 m³/d was used to convert ambient concentrations to intake. Sensitivity analysis was conducted to evaluate the impact of the limits of integration. Local exposure efficiencies were computed using 25, 50 and 100 km limits. The median estimate of local exposure efficiency (within 50 km) for perchloroethylene emissions from dry cleaners was 4×10^{-6} . The distribution of local exposure efficiencies (within 50 km) from the 100 individual dry cleaners was approximately lognormal with a geometric standard deviation of 2.3, a minimum value of 3.2×10^{-7} and a maximum value of 3.2×10^{-5} .

Thompson used a simple one-compartment box model to estimate global exposure efficiency for perchloroethylene emissions from dry cleaners in the Northern hemisphere. Her best estimate of the global exposure efficiency of perchloroethylene was 3.5×10^{-6} , but she provided a range of possible values from 1.9×10^{-6} to 1.7×10^{-5} to reflect uncertainty about the atmospheric half-life of perchloroethylene and other model parameters. The impact of Thompson's consideration of global transport was to increase her best estimate of the total (local + global) exposure efficiency of

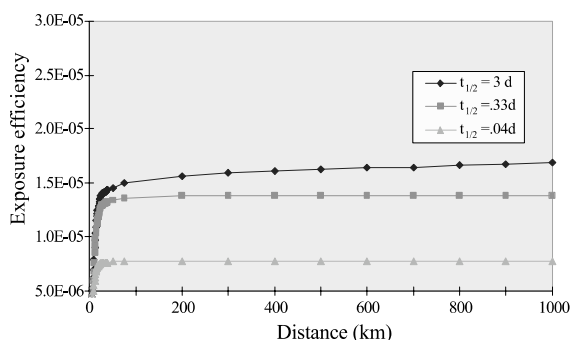


Fig. 2. Exposure efficiency estimates for particles from a refinery in Bangkok, Thailand (Phonboon, 1996). (As a function of distance from the source and assumed atmospheric half-life.)

perchloroethylene by about a factor of 2 (from 4×10^{-6} to 7.5×10^{-6}). A secondary effect was to substantially increase the estimates of exposure efficiency at the low end of the distribution and therefore to reduce the variability in total exposure efficiency among sites.

Recently, Lai, Thatcher and Nazaroff evaluated exposure efficiencies for both indoor and ambient point sources of particles (Lai et al., 1999, 2000). They used the term “inhalation transfer factor” rather than “exposure efficiency,” but as their definition of inhalation transfer factor—“the fraction of an emitted pollutant that is expected to be inhaled. . .”—makes clear, the two terms are synonymous. In contrast to most previous analyses, Lai et al. explored the sensitivity of exposure efficiency estimates to underlying variables. Extensive sensitivity analyses examined the impact of variations in ventilation rates on indoor exposure efficiencies and of variations in stack height, wind speed and atmospheric stability on ambient point source exposure efficiencies. In addition, they considered the distribution of exposure efficiencies across the affected population. However, they did not provide estimates of exposure efficiencies for either specific or typical sources or source classes.

In summary, many estimates of exposure efficiency for particulate matter and a number of other compounds in air have been published. They vary over more than six orders of magnitude. This variation reflects differences in a number of factors—including properties of the source (effective stack height, location relative to populations), the pollutant and atmosphere (atmospheric half-life, mixing height), and the population exposed (population density and breathing rate). In addition, characteristics of the analysis—such as the scale of analysis (local, regional, global), the method for characterizing the relationship between emissions and concentrations (Gaussian model, compartmental model, empirical calculations), the approach for addressing uncertainty and variability in parameter values and results—all affect the computed values.

Table 1 summarizes the estimates of exposure efficiency for ambient sources of particles that had been published prior to the SGOMSEC 14 Workshop. Some generalizations are possible. Ambient sources of particulate air pollution (such as coal-fired power plants, refineries and mobile sources) seem to have exposure efficiencies on the order of 10^{-7} – 10^{-5} . These are about 1000 fold smaller than the exposure efficiencies of indoor sources of particles. For point sources in or near major metropolitan areas, local exposure efficiency is approximately proportional to local population density. For pollutants with even moderately long half-lives (such as fine particles), consideration of regional exposure efficiency may increase total exposure efficiency by a factor of about 2 over the values that would be obtained by analysis of local exposure efficiency alone. Similarly, models based totally on regional or global estimates may

be biased downward due to failure to consider the details of exposure near the source. Finally, it is important to note that none of the particle exposure efficiency estimates published prior to SGOMSEC evaluated secondary particles formed in the atmosphere from emissions of SO_2 or NO_x .

6. New estimates of particulate exposure efficiency for power plants and vehicles

In this manuscript we present new estimates of exposure efficiency from Wolff's analysis of the impacts of fine ($<2.5 \mu\text{m}$ aerodynamic diameter) particulate matter, both primary and secondary, from coal-fired power plant and vehicular emissions in the United States (Wolff, 2000). Modern trajectory models have been coupled with census data on population density to evaluate the exposure efficiencies for primary particles and for secondary particles formed in the atmosphere from sulfur dioxide and nitrogen oxide emissions.

Wolff's fate and transport modeling for power plants and mobile sources used the CALPUFF model (US EPA, 1995). CALPUFF is based on Gaussian dispersion theory, and models continuous emissions as a sequence of discrete puffs. For long range transport, CALPUFF operates as a trajectory model, incorporating meteorological data from the nearest weather station as the plume migrates further and further downwind. Hourly meteorological data for the year 1990 were obtained from EPA. CALPUFF was used to estimate the source-related increment in ambient concentration in each of 448 geographic cells, each $100 \text{ km} \times 100 \text{ km}$, covering a region 1600 km (N–S) by 2800 km (W–E) around the source. These hourly values were averaged to determine the impact of the source on the annual mean concentration experienced in each cell. Population data for each cell were derived using ArcView 3.2 Geographic Information System and includes all of the population of the US and portions of southern Canada and northern Mexico. Future work will include more detailed analysis of the region near the source.

CALPUFF uses variable first order rate constants to model the reactions governing the transformation of SO_2 to sulfate and of NO_x to nitrate. For SO_2 , the rate constant used in CALPUFF reflects both gas phase and aqueous phase reactions. During each daylight hour of simulated transport and diffusion of the plume a new rate constant is computed using an empirical relationship reflecting the influences of the intensity of solar flux, the atmospheric stability, the concentration of ozone and the relative humidity. At night, CALPUFF utilizes a default SO_2 transformation rate of 0.2% per hour. For NO_x , during the daytime a similar approach is used in which the rate constant is determined based on the atmospheric stability and the prevailing NO_x and ozone

Table 1
Previous estimates of exposure efficiency for ambient sources of particles

Researcher	Source(s)	Half-life (d)	Breathing rate (m ³ /d)	Scale of analysis (km)			Population density (persons/km ²)				Exposure efficiency estimate		
				Local	Reg'l	Global	<20 km	<50 km	Reg'l	Global	Local	Reg'l	Global
Harrison	Mixed	4.7	20	50	1500	No	1200	—	20	—	1×10^{-5}	2×10^{-5}	—
Smith	CFPP in US	0.5–3.5 ^a	28	—	US	No	—	—	30 ^b	—	—	1×10^{-6}	—
	CFPP in LDC	”	”	—	India	”	—	—	270 ^c	—	—	1×10^{-5}	—
	Vehicles	”	”	—	n/s ^d	”	n/s ^d	—	—	—	2×10^{-5}	—	—
	Neighborhood sources	”	”	—	n/s ^d	”	n/s ^d	—	—	—	4×10^{-5}	—	—
Phonboon	Refinery—base case	0.33	9.5	50	1000	No	4000	900	50	—	1.4×10^{-5}	1.4×10^{-5}	—
	sensitivity analysis	3	”	”	”	”	”	”	”	—	1.5×10^{-5}	1.7×10^{-5}	—
	sensitivity analysis	0.04	”	”	”	”	”	”	”	—	0.7×10^{-5}	0.7×10^{-5}	—
Jolliet	Mixed	7.6	20	—	—	Yes	—	—	—	10	—	—	0.7×10^{-6}

^a Dry deposition was modeled using a terminal settling velocity of 0.23 cm/s (which would be characteristic of a particle with an aerodynamic diameter of about 10 μ m). Using the minimum and maximum (daytime) mixing heights of 200 and 1500 m, and assuming uniform vertical mixing an estimate of the atmospheric half-life of between 0.5 and 3.5 d can be derived.

^b The analysis used estimated 1985 US population statistics for the continental US, with a total estimated population of 235 million persons and a land area of approximately 3 million square miles (or 7.3 km²).

^c Based on the ratio of India's population density to the US's population density of 9.

^d These details, e.g., scale of analysis and population density in domain of analysis, were not specified by the original authors.

concentrations. At night, CALPUFF uses a NO_x transformation rate of 2% per hour. To account for the fact that particulate nitrate is only formed in cold temperatures, modeled NO_x concentrations were divided by four (i.e., 3 cold months/12 months) (Wolff, 2000).¹⁸ Hourly data for ozone and ammonia were not available so default values of 80 ppb ozone and 10 ppb ammonia were used.

In the analysis of exposure efficiencies for secondary particles it is important to carefully define all terms. In our analysis sulfate exposure efficiencies have been computed as the ratio of sulfate (SO_4) inhaled to sulfur dioxide (SO_2) emitted. Similarly nitrate exposure efficiencies have been computed as the ratio of nitrate (NO_3) inhaled to nitrogen oxides emitted, reported as equivalent NO_2 emissions.¹⁹

To account for the fact that particle deposition is a strong function of particle size, CALPUFF divides the total particle mass into a number of size-specific bins. We relied on the CALPUFF's default particle size distribution—i.e., mass median diameter of 0.5 μm and a geometric standard deviation of 2. Both gravitational settling and diffusion are modeled. Wet deposition losses due to rainout or washout were not modeled.²⁰

¹⁸ Wolff's division of the nitrate concentrations by 4 may have overcompensated for the effect of temperature on nitrate formation. Current work suggests that CALPUFF nitrate results may be used directly without application of such a correction.

¹⁹ If one were interested in knowing the fraction of sulfur molecules emitted that are eventually inhaled, it would be necessary to divide our reported sulfate exposure efficiencies by 1.5, reflecting the ratio of the molecular weights of SO_4 and SO_2 . Alternatively, if one were interested in knowing the ammonium sulfate mass that would correspond to our reported sulfate exposure efficiencies, it would be necessary to multiply the reported values by 1.375, reflecting the ratio of the molecular weights of $(\text{NH}_4)_2\text{SO}_4$ and SO_4 . Similarly, to find the fraction of nitrogen molecules emitted that would eventually be inhaled one would need to divide our reported nitrate exposure efficiencies by 1.35 and to find the ammonium nitrate mass that would correspond to our reported nitrate exposure efficiencies it would be necessary to multiply the reported values by 1.29.

²⁰ We believe that failure to model wet deposition may have yielded exposure efficiency estimates that are biased upward by 10% or 15%. Samet et al. (1997) explored the sensitivity of CALPUFF results for one power plant in Washington to the treatment of wet deposition and concluded that "ambient concentrations did not change significantly with this assumption and it can be concluded that the model is not sensitive to wet deposition." Levy et al. (2002) found that removing both wet and dry deposition in a CALPUFF analysis of power plant emissions in Illinois increased primary $\text{PM}_{2.5}$ exposure efficiencies by 16%, nitrates by 25% and sulfates by 43%.

For each source class, exposure efficiency estimates were derived for 40 specific sources, selected using a stratified random sampling approach and intended to be representative of others in the class. For coal-fired power plants, the stratification was based on geographic region, with more samples located in the regions with greater emissions.²¹ For highway segments, the stratification was based on both geographic region and urban/rural classification.²² For both source classes, regional sampling weights were based on the contribution of the region to the total national emissions of sulfates and nitrates for the source class.

Table 2 summarizes the results for the 40 power plants (Wolff, 2000). The mean exposure efficiencies for power plant emissions were estimated to be 2.2×10^{-6} for primary $\text{PM}_{2.5}$, 1.6×10^{-7} for sulfate particles derived from SO_2 emissions, and 2.7×10^{-8} for nitrate particles derived from NO_x emissions.

In addition to this strong dependence of exposure efficiency on the nature of the particle of interest, there was considerable variation in the exposure efficiency across the sources chosen to represent each class. For primary $\text{PM}_{2.5}$, the exposure efficiencies for power plant emissions varied from 2.5×10^{-7} to 6.3×10^{-6} across the 40 sources which were evaluated. Fig. 3 is a cumulative frequency distribution of the primary $\text{PM}_{2.5}$ results, plotted on log-probability paper.²³ The median and gsd of the primary $\text{PM}_{2.5}$ exposure efficiencies from this plot are 1.9×10^{-6} and 1.9, respectively. Smaller plant to plant variations in exposure efficiency were seen for sulfate and nitrate particles, with gsd's of 1.4 and 1.6 respectively.

Table 3 summarizes the results for the 40 vehicular emissions sites. The mean exposure efficiencies for mobile sources were estimated to be: 8.8×10^{-6} and 9.4×10^{-6} for primary $\text{PM}_{2.5}$ from rural and urban highway stretches; 1.4×10^{-7} and 1.2×10^{-7} for sulfate particles from rural and urban vehicular SO_2 emissions; and 2.6×10^{-8} and 2.3×10^{-8} for nitrate particles from rural and urban vehicular NO_x emissions.

Again, for these sources there was considerable variability in exposure efficiency from one highway segment to another. For primary $\text{PM}_{2.5}$ vehicular emissions exposure efficiencies varied from 3.0×10^{-6} to

²¹ The final sampling densities for coal-fired power plants were 12 plants from the South; 9 plants from the Midwest; 14 plants from the Northeast; and 5 plants in the West.

²² The final sampling densities for highway stretches were (for both rural and urban highways) 6 segments in the South; 5 segments in the Midwest; 5 segments in the Northeast; and 4 segments in the West.

²³ A Kolmogorov–Smirnov test for lognormality was computed and evaluated. At the 95% significance level, the null hypothesis could not be rejected.

Table 2

Exposure efficiency estimates for fine particles from US coal-fired power plants (Wolff, 2000)—based on results from a stratified random sample of 40 large power plants

Pollutant	Mean	SEM ^a	Minimum	Maximum
Primary PM _{2.5}	2.2E-06	1.9E-07	2.5E-07	6.3E-06
SO ₂ /sulfate	1.6E-07	5.9E-09	6.0E-08	2.2E-07
NO _x /nitrate ^b	2.7E-08	1.0E-09	7.4E-09	5.8E-08

^a SEM = standard error of the mean.

^b NO_x/nitrate values were computed by dividing CALPUFF estimates by a factor of 4 to account for the role of low temperature in formation of particulate nitrate. It is no longer clear that such a correction is necessary.

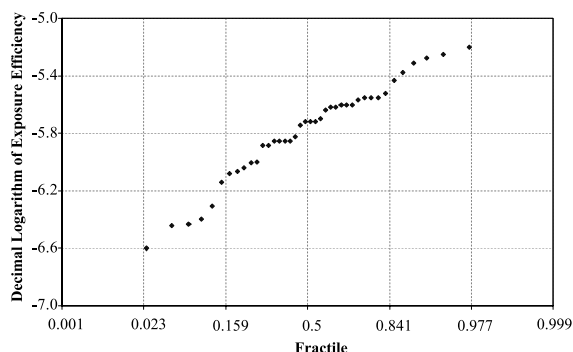


Fig. 3. Variability in exposure efficiencies for fine particles from US power plants (Wolff, 2000). (Shown as log-probability plot of results for 40 US coal-fired power plants.)

1.8×10^{-5} in urban areas, and from 1.2×10^{-6} to 1.8×10^{-5} in rural areas. Similar, but somewhat smaller variations were seen in the sulfate and nitrate exposure efficiencies.

Wolff examined the effect of the scale of analysis on the estimate of exposure efficiency. His results for one power plant are shown in Fig. 4a and b. Note that for primary fine particles, the estimated exposure efficiency reaches 50% of its final value at approximately 500 km

from the source, and 95% of its value about 1000 km from the source. For sulfates or nitrates from power plants, much of the exposure occurs at great distances from the source. For sulfates, the estimated exposure efficiency reaches 50% of its final value approximately 750 km from the source and reaches 95% of its final value at 1400 km. For nitrates, the corresponding distances are 650 km (50%) and 1350 km (95%). For emissions from automobiles, exposure is more concentrated near the source with 50% of total exposure efficiency being reached within 100 km for urban highway segments and within 350 km for rural highway segments.

Finally, Wolff used a simple rollback approach as a cross-check on his model-based estimates of exposure efficiencies for primary PM_{2.5}, sulfates and nitrates. Average US ambient concentrations of PM_{2.5}, sulfates and nitrates were divided by total US emissions of primary PM_{2.5}, sulfates and nitrates. The resulting crude estimates of exposure efficiencies—i.e., 2.7×10^{-6} for PM_{2.5}; 2.6×10^{-7} for sulfates; and 2.0×10^{-8} for nitrates—are at least qualitatively consistent with the estimates derived from source specific modeling.

To apply the exposure efficiency concept to large-scale policy decisions, the results from a limited number of sample calculations need to be generalized in such a way that they can be applied to other sources and geographic locations. This would allow for expedited risk

Table 3

Exposure efficiency estimates for fine particles from US mobile sources (Wolff, 2000)—based on results from a stratified random sample of 20 urban and 20 rural highway segments

Site and pollutant	Mean	SEM ^a	Minimum	Maximum
<i>Urban</i>				
Primary PM _{2.5}	9.4E-06	8.0E-07	3.0E-06	1.8E-05
SO ₂ /sulfate	1.2E-07	9.3E-09	3.5E-08	2.0E-07
NO _x /nitrate ^b	2.3E-08	2.0E-09	4.1E-09	4.6E-08
<i>Rural</i>				
Primary PM _{2.5}	8.8E-06	1.0E-06	1.2E-06	1.8E-05
SO ₂ /sulfate	1.4E-07	9.0E-09	4.0E-08	2.2E-07
NO _x /nitrate ^b	2.6E-08	2.3E-09	1.2E-08	5.1E-08

^a SEM = standard error of the mean.

^b NO_x/nitrate values were computed by dividing CALPUFF estimates by a factor of 4 to account for the role of low temperature in the formation of particulate nitrate. It is no longer clear that such a correction is necessary.

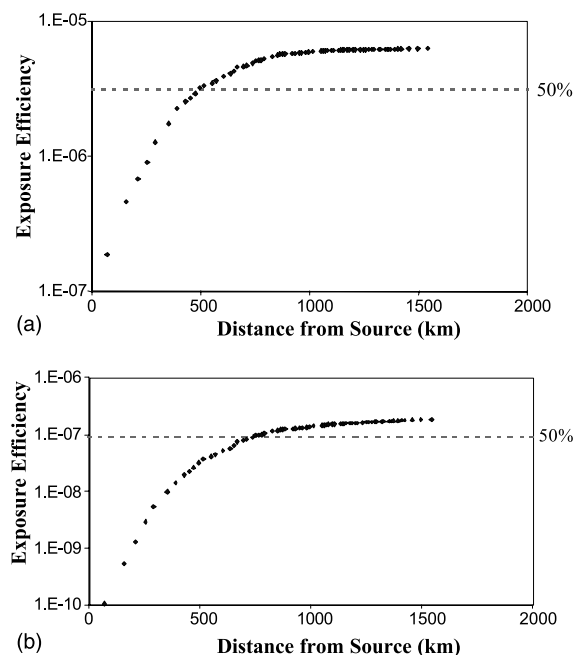


Fig. 4. (a) PM_{2.5} exposure efficiency as a function of distance from a power plant (Wolff, 2000). (b) Sulfate exposure efficiency as a function of distance from a power plant (Wolff, 2000). (Illustrative results for one power plant in West Virginia.)

assessments while maintaining the analytical strength underlying the calculation of exposure efficiency. To achieve this goal, we are currently undertaking an analysis to determine the factors which account for a large degree of the observed variation in estimates of exposure efficiency, with an eye toward developing a meta-analytic model of exposure efficiency that could be used in risk assessment in developing countries.

Based on the functional form of exposure efficiency and on basic principles governing atmospheric fate and transport, we anticipate that exposure efficiency will be strongly dependent on terms such as population density, wind speed, mixing height, pollutant half-life, and stack height. Fig. 5a plots exposure efficiency for PM_{2.5} against population within 500 km of the source. Fig. 5b is similar, but involves exposure efficiency for sulfate and plots this against population within 1000 km of the source. These figures confirm our expectation that population density is likely to be a strong predictor of exposure efficiency. Fig. 6 plots exposure efficiency for PM_{2.5} from power plants against stack height. Again, as expected, the exposure efficiency tends to drop as the stack height increases.

Preliminary analysis of annual average exposure efficiency suggests that much of the variability can be accounted for by a small number of terms. Ongoing work is aimed at finalizing the form of the regression model for annual average exposure efficiency (Levy et al.,

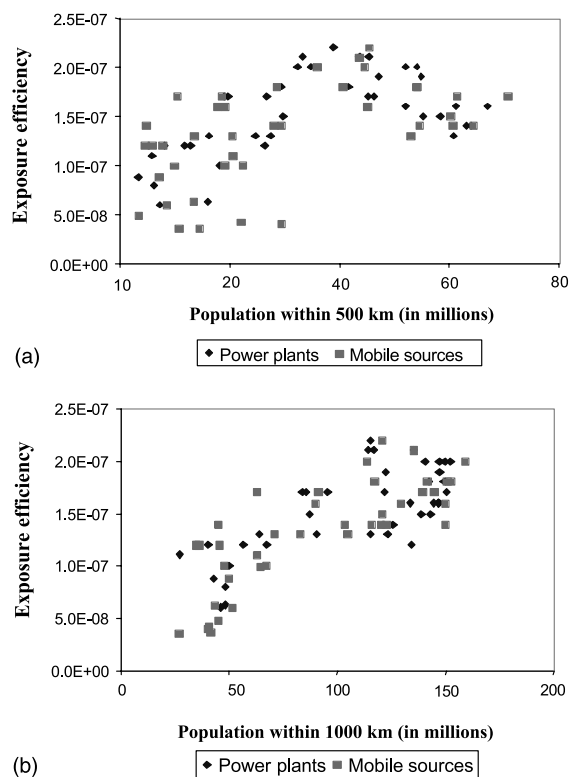


Fig. 5. (a) Dependence of primary PM_{2.5} exposure efficiency on population within 500 km of source. (b) Dependence of sulfate exposure efficiency on population within 1000 km of source (using Wolff's results for power plants and mobile sources).

in press) and exploring predictive factors for shorter time scales.

7. Discussion and conclusions

The concept of exposure efficiency has great potential for providing exposure information that is relevant for risk assessment and decision making. Exposure efficiency is a simple, transparent and potentially comprehensive measure of the relationship between emissions and human exposures. Once exposure efficiencies have been computed for a wide variety of sources, compounds and environments the concept should facilitate sequential risk assessment and decision analysis by providing preliminary estimates of the exposure likely to result from each of several major sources or source classes of interest. These exposure estimates can be readily combined with preliminary (default) dose-response coefficients (or regulatory thresholds) from data bases such as IRIS to derive provisional estimates of risk and to understand which emission sources, transport pathways, and compounds should be sub-

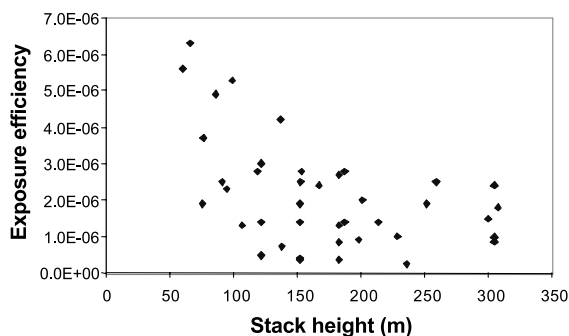


Fig. 6. Dependence of primary $\text{PM}_{2.5}$ exposure efficiency for power plants on stack height (using Wolff's results for 40 US coal-fired power plants).

jected to more detailed, site-specific, and scientifically rigorous analysis. Carrothers (2000) and Thompson and Evans (1997) provide examples of the use of exposure efficiency in support of risk and decision analysis.

Several research groups have realized the power of this concept and have made preliminary calculations of exposure efficiency for a variety of classes of sources of exposure to particulate matter—including coal-fired power plants, automobiles, cook stoves and cigarette smoke. The variation in exposure efficiency across these source classes is enormous—varying from 1 (for mainstream cigarette smoke) to 1 in 1 million for smoke from coal combustion in US coal-fired power plants. This implies that cost-effectiveness analysis based on the cost per unit of exposure reduction are likely to yield entirely different results than cost-effectiveness analyses based on the cost per unit emissions reduction.

More recently there have been efforts to compute exposure efficiency estimates that are chemical and/or particle size specific, in view of the fact that various combustion sources yield different mixes of primary and secondary particles; and that the size distributions of the primary particles emitted from these sources may vary by source class and may be influenced by the application of various control technologies. These results indicate that while exposure efficiencies for primary $\text{PM}_{2.5}$ particles may be on the order of 2×10^{-6} ; the exposure efficiencies for sulfur dioxide/sulfates are approximately an order of magnitude lower (2×10^{-7}); and the exposure efficiencies for nitrogen oxides/nitrates may be as much as two orders of magnitude lower (3×10^{-8}). Again, these substantial differences, indicate that the nature and size of the particles involved must be carefully considered in cost-effectiveness analysis.

Further, recent studies have begun to explore the facility to facility variation in exposure efficiency within source classes. Wolff's work suggests that for coal-fired power plants and interstate highway stretches in the US, these variations can be large enough (i.e., geometric

standard deviations on the order of 2) that potential differences in the cost-effectiveness of exposure controls of various sources within a source class may warrant some attention in economic analysis of regulatory options for air pollution control (Wolff, 2000).

All of this is quite promising, but in our efforts to advocate this approach we must not lose sight of the assumptions underlying the approach, the issues which remain to be addressed, and the opportunities for improving upon our current understanding of exposure efficiency.

Primary among the assumptions underlying the use of exposure efficiency in support of risk analysis and regulatory policy development, is that exposure efficiency is a proxy for health risk. For this to be true several conditions must hold—(i) health risk must be proportional to cumulative exposure (i.e., there must be no significant dose-rate dependence of risk); (ii) health risk must be proportional to $\text{PM}_{2.5}$ mass (i.e., there must be no significant differences in the toxicity of particles on chemical composition or particle size); and (iii) the social costs of health risks must be proportional to the population risk (i.e., there must be no significant non-linearity in the social utility function—individual risks matter only to the degree that they contribute to population risk).

Current estimates of exposure efficiency have been simple integrative measures—which aggregate over people, time, compounds and particle sizes. Given our current, rather crude, understanding of the relationships between exposure to particulate matter and the risks of mortality and morbidity these simple exposure efficiency estimates may be entirely appropriate. However as scientific understanding improves, we will need to consider developing measures of exposure efficiency which—(i) are size and compound specific; (ii) provide distributional characterizations of individual exposure efficiencies rather than a single cumulative value; and (iii) facilitate analysis of the fraction of the cumulative exposure efficiency which is likely to be experienced at various dose-rates.

Finally, we must recognize that very little attention has been devoted to characterizing the uncertainty in current estimates of exposure efficiency. Further work in this area is needed so that risk and policy analysts who use exposure efficiency estimates will have some sense of whether these values are known to within a factor of 10, a factor of 2, or to within 5% or 10%. Only then, will it be clear whether more refined approaches for exposure assessment in support of policy are necessary or not.

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References

- Anderson, G.E., 1980. Human exposure to atmospheric concentrations of selected chemicals. Science Applications International Corporation, Report to the US EPA under EPA contract 68-02-3066.
- Anderson, G.E., 1983. Modeling of human exposure to airborne toxic materials. In: Swann, R.L., Eschenroeder, A. (Eds.), *Fate of Chemicals in the Environment*. American Chemical Society, Washington, DC, Chapter 4.
- Bennett, B.G., 1981. Exposure Commitment Assessments of Environmental Pollutants (I) Exposure Commitment Concepts and Applications: Summary Exposure Assessments for Lead, Cadmium, and Arsenic, Technical Report, Chelsea College, Monitoring and Assessment Research Centre, London, UK.
- Bennett, D.H., McKone, T.E., Evans, J.S., Nazaroff, W., Margni, M.D., Jolliet, O., Smith, K.R., 2002. Defining intake fraction. *Environmental Science and Technology* 36, 206A–211A.
- Brinkhoff, T., 1999. Principle Cities and Agglomerations of the World, (<http://citiesin.thecountry.com>).
- Boubel, R.W., Fox, D.L., Turner, D.B., Stern, A.C., 1994. *Fundamentals of air pollution*, Third ed. Academic Press, San Diego, California.
- Carrothers, T.J., 2000. Particulate Air Pollution Policy: Assessing the Economic Value of Further Research, Doctoral Thesis, Departments of Environmental Health and Health Policy and Management, Harvard School of Public Health.
- Central Intelligence Agency (CIA), 1998. *The World Factbook 1998*, Central Intelligence Agency, Washington, DC.
- Dockery, D.W., Spengler, J.D., 1979. Indoor–outdoor relationships of respirable sulfates and particles. *Atmospheric Environment* 15, 335–343.
- Evans, J.S., Hattis, D., Thompson, K.M., 2000. Exposure efficiency. *Journal of the Air and Waste Management Association* 50, 1700–1703.
- Harrison, K., Hattis, D., Abbat K., 1986. Implications of Chemical Use for Exposure Assessment: Development of an Exposure-Estimation Methodology for Application in a Use-Clustered Priority Setting System, MIT Center for Technology Policy and Industrial Development, Report Number CTPID 86-2 to the US EPA.
- Jaenicke, R., 1980. Natural Aerosols. In: Kneip, T.J., Lioy, P.J. (Eds.), *Aerosols: Anthropogenic and Natural Sources and Transport*, 338. *Annals of the New York Academy of Sciences*, pp. 317–329.
- Jolliet, O., 1994. Critical surface–time: An evaluation method for life cycle assessment. In: Udo de Haes, H., Jensen, A., Klopffer, W., Lindfors, L.G. (Eds.), *Integrating Impact Assessment into LCA*, Proceedings of the 4th SETAC-Europe Congress, SETAC, Brussels, pp. 133–142.
- Jolliet, O., Crettaz, P., 1996. Critical Surface–Time 95: A Life Cycle Impact Assessment Methodology Including Fate and Exposure. Swiss Federal Institute of Technology, Institute of Soil and Water Management, 1015 Lausanne, Switzerland.
- Lai, A.C.K., Thatcher, T.L., Nazaroff, W.W., 1999. Inhalation transfer factors for assessing human health risks from air pollutant sources. *Indoor Air '99*, vol. 5, 193–198, Conference held in Edinburgh, Scotland, 8–13 August 1999.
- Lai, A.C.K., Thatcher, T.L., Nazaroff, W.W., 2000. Inhalation transfer factors for air pollution health risk assessment. *Journal of Air and Waste Management Association* 50, 1688–1699.
- Layton, D.W., 1994. Metabolically consistent breathing rates for use in dose assessment. *Health Physics* 64, 23–36.
- Levy, J.I., Wolff, S.K., Evans, J.S., in press. A regression-based approach for estimating primary and secondary particulate matter intake fractions. *Risk Analysis*.
- Levy, J.I., Spengler, J.D., Hlinka, D., Sullivan, D., Moon, D., 2002. Using CALPUFF to evaluate the impacts of power plant emissions in Illinois: model sensitivity and implications. *Atmospheric Environment* 36, 1063–1075.
- Lindell, B., 1978a. Dose assessment analogies. *Environ Health Perspectives* 22, 75–78.
- Lindell, B., 1978b. Source-related detriment and the commitment concept: applying the principles of radiation protection to non-radioactive pollutants. *Ambio* 7 (5–6), 250–259.
- National Academy of Sciences (NAS), 1983. *Risk Assessment in the Federal Government: Managing the Process*, National Academy Press, Washington, DC.
- National Economic and Social Development Board of Thailand, 1991. *Population Projections for Thailand: 1980–2015*, Human Resources Planning Division, Bangkok, Thailand.
- National Statistical Office of Thailand, 1992. *Population and Housing Census*, Office of the Prime Minister, Bangkok, Thailand.
- Nichols, A.L., 1984. *Targeting economic incentives for environmental protection*. MIT Press, Cambridge, MA.
- Phonboon, K., 1996. *Risk Assessment of Environmental Effects in Developing Countries*, Doctoral Thesis, Harvard School of Public Health, Boston, Massachusetts.
- Roumasset, J.A., Smith, K.R., 1990. Exposure trading: an approach to more efficient air pollution control. *Journal of Environmental Economics and Management* 18, 276–291.
- Rowe, M.D., 1981. *Human Exposure to Particulate Emissions from Power Plants*, BNL-51305, Brookhaven National Laboratory, Upton, NY, 1981.
- Samet, J.M., Jodrey, D.J., Xu, J., Wings, K.D., 1997. An assessment of the health risks due to air emissions from the centralia power plant. Submitted to Centralia Power Plant Collaborative Decision Making Group.
- Schwartz, S.E., 1989. Acid deposition: Unraveling a regional phenomenon. *Science* 243, 753–763.
- Smith, K.R., 1987. *Biofuels, Air Pollution, and Health*. Plenum, New York.
- Smith, K.R., 1988a. Total exposure assessment: Part 1, Implications for the US. *Environment* 30 (8), 10–15, 33–38.
- Smith, K.R., 1988b. Total exposure assessment: Part 2, Implications for developing countries. *Environment* 30 (10), 16–20, 28–35.
- Smith, K.R., Edgerton, S.A., 1989. *Exposure Based Air Pollution Control Strategies: Proposed Three-Way Integration*. Presented at EPA/AWMA Specialty Conference “Total Exposure Assessment Methodology”, Las Vegas, Nevada.
- Smith, K.R., Ahuja, D., 1990. Toward a greenhouse equivalence index: the total exposure analogy. *Climatic Change* 17 (1), 1–7.

- Smith, K.R., 1993a. Fuel combustion, air pollution, and health: The situation in developing countries. *Annual Review of Energy and the Environment* 18, 529–566.
- Smith, K.R., 1993b. Taking the true measure of air pollution. *EPA Journal* 19 (4), 6–8.
- Smith, K.R., Swisher, J., Kanter, R., Ahuja, D.R., 1993. Indices for a Greenhouse Gas Control Regime: Incorporating Both Efficiency and Equity Goals. In: Siddayao, C. (Ed.), *Energy Investments and the Environment*, World Bank/EDI Technical Series. Washington, DC, 1993, pp. 157–253 (Originally, Environmental Division Working Paper 91-21, World Bank).
- Smith, K.R., 1994. Looking for pollution where the people are, *Asia-Pacific Issues* #10, EWC, Honolulu, p. 8.
- Smith, K.R., 1995. The potential of human exposure assessment for air pollution regulation, WHO/EHG/95.9, United Nations Environment Program, Office of Global and Integrated Environmental Health, World Health Organization, Geneva, Switzerland.
- The World Resources Institute, 1994. *World Resources 1994–95: A Guide to Global Environment*, Oxford Press, New York.
- Thompson, K.M., 1995. Evaluation of National Exposure Assessment Information for Improving Risk Management Decisions, Doctoral Thesis, Harvard University.
- Thompson, K.M., Evans, J.S., 1997. The value of improved national exposure information for perchloroethylene: A case study for dry cleaners. *Risk Analysis* 17, 253–271.
- Tsai, F.C., 1999. Total Exposure and Significant Source Apportionment: The Relationships among Emission Sources, Concentrations, and Health Effects of Particulate Matter, Ph.D. Thesis, Environmental Health Sciences, UC Berkeley, Berkeley, California.
- UN Scientific Committee on the Effects of Atomic Radiation, 1977. *Sources and Effects of Ionizing Radiation*, UN, New York.
- US EPA, 1992. User's Guide for the Industrial Source Complex (ISC2) Dispersion Models. vol. I–III, EPA-450/4-92-008a-c, US EPA, Research Triangle Park, North Carolina.
- US EPA, 1995. A User's Guide for the CALPUFF Dispersion Model, EPA-454/B-95-006, US EPA, Office of Air Quality Planning and Standards, Research Triangle Park, North Carolina.
- Wang, X., Smith, K.R., 1999a. Near-term Health Benefits of Greenhouse Gas Reductions: A Proposed Assessment Method and Application to Two Energy Sectors of China, WHO/SPE/99-01, World Health Organ, Geneva, 1999.
- Wang, X., Smith, K.R., 1999b. Secondary benefits of greenhouse gas control: Health impacts in China. *Environmental Science and Technology* 33 (18), 3056–3061.
- Wolff, S.K., 2000. Evaluation of fine particle exposures, health risks and control options, Doctoral Thesis, Department of Environmental Health, Harvard School of Public Health.