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Abstract

Indoor pollutant concentrations depend on the indoor pollutant source strength, the total rate of pollutant removal by ventilation, air cleaning, and other removal processes, and the outdoor concentration. Pollutant control techniques can be based on modifying any of these factors except the outdoor concentration. In this paper, various pollutant control strategies are examined by reference to measured data and use of mass balance models. It is shown that the largest reductions in indoor pollutant concentration are generally achieved by limiting or reducing pollutant source strengths, and several often-practical source control measures are identified. Ventilation is identified as the predominant removal process for most pollutants and various relationships between the ventilation rate and the indoor concentration are illustrated. The impacts of other pollutant removal processes on indoor concentration are also examined. Conclusions are that efficient indoor pollutant control strategies must focus on both limiting of pollutant source strengths and ventilation and that the concept of an acceptable minimum ventilation rate should be made distinct from the concept of acceptable indoor air quality.

Introduction

Concern about indoor air quality, particularly the indoor concentrations of certain air pollutants, has increased greatly during the past several years. There are a number of reasons for this increased level of concern. The recent emphasis on energy conservation has stimulated reductions in building ventilation rates, and a consequence of reduced ventilation is increased indoor concentrations of pollutants with indoor sources. New pollutant sources are being introduced into buildings; two examples are the greater numbers and quantities of synthetic building materials being used in new buildings and the recent popularity of kerosene-fired, unvented space heaters. Undoubtedly, the increased concern with indoor air quality has also resulted from the availability of new information indicating that the indoor concentrations of some pollutants frequently and substantially exceed the outdoor concentrations.

Indoor pollutant concentrations are dependent on numerous factors. Two major factors are the indoor pollutant source strength and the ventilation rate, each of which varies substantially among buildings. For some pollutants, the indoor concentration is also affected substantially by pollutant removal processes other than ventilation, such as deposition of pollutants on surfaces and chemical reactions that occur on surfaces or within the air. The outdoor pollutant concentration is an important factor in cases where it is significant compared to the indoor concentration. Spatial variations in the indoor concentration can affect the efficiency of pollutant removal (e.g., by ventiltion) and thus the average indoor concentration. One last important variable is time; indoor pollutant concentrations often vary dramatically over time.

Techniques to control indoor pollutant concentrations can be based on any (or a combination) of the factors that affect the indoor concentration. Logic dictates, however, that emphasis be given to measures that are expected to have the largest effect on the indoor concentration and that are relatively practical and cost effective.

In this paper, measured data and simple mass-balance models are utilized to examine the potential impact of various factors on the indoor pollutant concentration. Some emphasis

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is placed on ventilation, although the pollutant source strength and removal processes other than ventilation are also treated. The implications for various pollutant control strategies are discussed.

Pollutant Source Strengths

The indoor pollutant source strength is defined here as the total rate at which pollutants are emitted, generated, and transported to indoors excluding the transport of pollutants into a building by entry of outdoor air. Experimental data indicates that variation in the indoor pollutant source strength is typically the largest cause of the wide ranges observed in indoor concentrations. This fact is illustrated by the data shown in Figures 1 and 2. Figure 1, a scatter plot of indoor radon concentration versus ventilation rate based on measurements in a large number of houses [1], shows that the indoor radon concentration varies widely, independently of variations in the ventilation rate. In fact, the distribution of indoor radon concentrations shown in the figure is broader than the distribution of ventilation rates; Nazaroff and Nero [2] present much more comprehensive data indicating that this remains true in general. Because ventilation is the only significant removal process for radon, the scatter of these data can only be explained by a broad distribution in radon source strengths. The second figure shows steady state indoor nitrogen dioxide concentration versus ventilation rate based on measurements within a single house but with various unvented gas-fired space heaters operating [3]. If no unvented heater or other unvented combustion appliance had been operating, we would expect the indoor nitrogen dioxide concentration to be less than the outdoor concentration which was typically, less than 0.1 ppm during the measurement period. Instead, the steady-state indoor nitrogen dioxide concentration varied from approximately 0.4 to 1.4 ppm depending primarily on the nitrogen dioxide source strength, i.e., the type of heater used and its operating condition. Other data are available that illustrate the importance of the source strength of other key indoor pollutants such as formaldehyde [4] and respirable particles [5]. Because of the broad distribution in indoor pollutant source strengths, maintenance of a typical ventilation rate of 0.5 to 0.9 air changes per hour [6], does not ensure that indoor pollutant concentrations will be acceptably low.

There is ample evidence that indoor pollutant source strengths can often be reduced greatly by practical measures [7]. In some instances, the source (e.g., an unvented heater) can simply be excluded or removed. When exclusion or removal is not practical, source strengths can often be reduced. For example, the emission rate of formaldehyde from pressed wood products can range by a factor of 10 due to variations in the urea-formaldehyde resins utilized and the manufacturing techniques [8,9], thus, formaldehyde source strengths can be reduced by utilizing pressed wood products that emit relatively small amounts of formaldehyde. In the case of radon, available data indicate that practical measures such as sealing penetrations between a building and the soil, and crawl space ventilation or sub-slab ventilation are often highly effective in reducing the transport of radon from the soil to within a building [7,10,11]. Decreases in indoor radon or radon progeny concentrations by a factor of five to ten are frequently reported when these measures are employed. Substantial emphasis should, therefore, be given to control measures which are based on reducing pollutant source strengths.

Mass Balance Models and Pollutant Removal Rates

An alternative strategy to control indoor pollutant concentrations is to modify the rate at which pollutants are removed from the indoor space by ventilation or other pollutant removal processes. The impact of a change in the magnitude of any particular removal process on indoor concentration can not be accurately estimated, however, without taking into account both the magnitude of other removal processes and any coupling between the pollutant source strength and the rate of pollutant removal. To allow an examination of these relationships, simple equations that relate the indoor pollutant concentration to other variables are presented in this section. These equations are based on a pollutant mass balance and are utilized in the next section to illustrate the impact of ventilation and other removal processes on indoor concentration. Each equation is based on simplifying assumptions of perfectly mixed indoor air and steady-state conditions.

Constant source strength model: If the pollutant source strength is independent of the ventilation rate and other removal rates and the assumptions stated above are met, then

the indoor concentration can be expressed by the equation

$$C_i = (S + aPC_o) / (a + K + \lambda + R) \qquad (1)$$

where:

S = pollutant source strength per unit volume indoor air,

a = air exchange rate (i.e., air flow rate/indoor volume),

P = fraction of outdoor pollutant that penetrates the building envelope or ventilation system,

C_o = outdoor pollutant concentration,

K = pollutant removal rate by deposition on surfaces and chemical reaction per unit volume indoor air,

λ = pollutant removal rate by radioactive decay, and

R = pollutant removal rate by air cleaning per unit volume indoor air.

Limited data are available for the various parameters in Equation 1. The air exchange rate of residential buildings with windows and doors closed ranges from about 0.2 air changes per hour (ach) to about 2.5 or 3 ach [6]. Information for choosing an appropriate penetration factor P and reaction constant K (also sometimes called the deposition constant or plate-out constant) is highly limited. For gases that are nearly non-reactive in indoor environments such as radon, carbon dioxide, and carbon monoxide the penetration factor and reaction constant are essentially unity and zero, respectively. Measured values of these constants for nitrogen dioxide, formaldehyde, and particles are listed in Table 1 [3,12-18]. Pollutant removal by reaction and deposition is poorly understood, but it is thought that this removal is caused by deposition on, adsorption on, or reaction with indoor surfaces and, in some cases, reactions within the indoor air. Utilization of a reaction constant in the form given in Equation 1 is an approximation. If pollutants are being removed by more than one process, each process should be accounted for independently. In the case of pollutant removal by chemical reaction or adsorption, the removal rate may not be linearly dependent on the indoor concentration as implied by Equation 1. In cases of pollutant removal by interactions with surfaces, the removal rate may depend on the nature and quantity of indoor surfaces and the amount of indoor air

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movement which affects the transport rate of pollutants to surfaces. Such removal is probably better characterized by a deposition velocity (which is a removal rate normalized by a surface area) than by a reaction constant (which is a removal rate normalized by the indoor volume); however, the majority of available data are in the form of reaction constants. Miyazaki [19], however, does provide substantial data on the deposition velocity for nitrogen dioxide -- the gaseous pollutant for which this removal process appears most significant.

Another problem with the available data on reaction constants is that they are generally from experiments in which the decay rate of pollutant concentration was measured. These experiments do not permit the process of physical adsorption of pollutants followed by later desorption to be distinguished from permanent removal processes. Thus, the average and range of pollutant removal by reaction and deposition is not well known. Despite these limitations, reaction and deposition can significantly affect indoor pollutant concentrations as shown later.

Radon progeny model: For several reasons, the impact of pollutant removal on radon progeny concentrations must be modeled by a more complex equation. The source of radon progeny is the radioactive decay of radon. Ventilation is a significant removal process for radon progeny and also affects the source strength of radon progeny (i.e., the radon concentration). In some instances, the source strength for radon itself may be coupled to the ventilation rate [1], however, for the equation presented below, a radon source strength that is independent of ventilation rate has been assumed. Another factor that must be accounted for is the removal of radon progeny by radioactive decay. Most researchers use the potential alpha energy concentration (PAEC) to indicate the total concentration of radon's short-lived progeny in units of working levels (WL). The PAEC indicates the potential alpha energy emission from all radon progeny present in the air.

An equation, similar to that presented by Rudnick et al. [20], that relates PAEC to ventilation rate and other factors is

$$PAEC = S\lambda_A /[(\lambda_R + a)(\lambda_A + a + k_A)] \times \qquad (2)$$
$$[0.00103 + (\lambda_B /(\lambda_B + a + k_B))(0.00507 + 0.00373(\lambda_C/(\lambda_C + a + k_C)))]$$

where:

S = radon source strength (pCi/h),

λ_R = radioactive decay constant for radon = 0.00758 h^{-1},

$\lambda_A, \lambda_B, \lambda_C$ = radioactive decay constants for progeny A, B, and C, respectively (λ_A = 13.7 h^{-1}, λ_B = 1.55 h^{-1}, λ_C = 2.11 h^{-1}),

a = the air exchange rate (h^{-1}), and

k_A, k_B, k_C = the first order deposition constants for progeny A, B, C, respectively (h^{-1}).

The deposition constants in Equation 2 are number-weighted average values for the airborne radon progeny which are either attached or unattached to airborne particles. Unattached radon progeny deposit on surfaces at a much greater rate than attached progeny, thus these deposition constants depend on the fractions of all progeny that are attached and unattached. Because the unattached fraction increases as the indoor particle concentration decreases, the deposition constants are highly dependent on the particle concentration. Several investigators have studied the deposition rate of radon progeny [21]. Reasonable upper and lower limits for these deposition constants are given by Offermann et al. [18] who measured values of k_A = 7.8 h^{-1}, k_B = 1.8 h^{-1}, and k_C = 0.5 h^{-1} for a low indoor particle concentration of 3000 particles/cc and values of k_A = 1.4 h^{-1}, k_B = 0.4 h^{-1}, and k_C = 0 for a high indoor particle concentration of 30,000 particles/cc. An important factor not accounted for in Equation 2 is that the indoor particle concentration and thus the deposition constants for radon progeny will generally be a function of the ventilation rate; this can be taken into account by using a more complex model [18].

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Formaldehyde model: The relationship between ventilation rate and indoor formaldehyde concentration can also be complex for the following reasons:

1. Multiple sources of formaldehyde may be present.

2. Some sources of formaldehyde may be separated from the indoor air by a permeation barrier (e.g., a carpet).

3. The emission rate of formaldehyde from many sources varies   with indoor formaldehyde concentration and the relationship between emission rate and indoor concentration varies among sources.

4. The emission rate of formaldehyde depends on the temperature and moisture content of the source which, in turn, is a function of the temperature and humidity of the surrounding air.

5. Formaldehyde may be removed by interaction with indoor surfaces, although such interaction is not considered in this paper.

The following equation similar to that presented by Matthews et al. [4], accounts for the presence of multiple formaldehyde sources:

$$C_i = C_o + (1/aV) \left(\sum_i S_i A_i \right) \tag{3}$$

where:

C_o = the outdoor formaldehyde concentration,

S_i = the source strength per unit area of source material i,

A_i = the area of source material i, and

V = the volume of indoor air.

For sources that are not separated from the indoor air by a permeation barrier, the following linear equation has been used [4] to relate the formaldehyde source strength to

the indoor formaldehyde concentration:

$$S_i = - M_i C_i + b_i \qquad (4)$$

where M_i and b_i are constants that depend on the source and must be determined experimentally. Equation 4 does not account for the substantial dependence of formaldehyde emission rate on temperature and humidity [22].

Impact of Pollutant Removal on Indoor Concentration

In this section, the mass-balance models are utilized to illustrate the impact of ventilation and other forms of pollutant removal on indoor concentration. The results shown are only valid when the indoor concentration greatly exceeds the outdoor concentration. Figure 3 shows curves of pollutant concentrations versus air exchange rate and their dependence on additional pollutant removal. Removal processes other than ventilation substantially impact the indoor concentrations when air exchange rates are moderate or low (i.e., < 0.5 ach); however, at high air exchange rates these removal processes have a relatively small effect on indoor concentrations. This figure illustrates two points. First, enhancement of pollutant removal by reaction or deposition (e.g., by selection of specific indoor surface materials or by mechanically mixing the indoor air) appears to be a potentially viable control measure for nitrogen dioxide, radon progeny and other pollutants that are reactive or subject to deposition. However, in buildings with a typical air exchange rate of 0.5 to 0.9 ach, increasing the reaction or deposition rate is unlikely to reduce the indoor concentration by more than a factor of approximately two. Second, pollutant removal rates in a well-mixed space by any process must be comparable to or greater than the ventilation rate, if a substantial reduction in indoor pollutant concentration is to be achieved. An example is provided by air cleaning devices designed to remove particles from residential air. Although some devices with sufficient particulate removal capacity are available, the capacity of the majority of devices being used is too low to have a significant effect on indoor particle concentrations [18].

A major advantage of ventilation compared to other pollutant removal processes is that it

is an effective control measure for all pollutants as long as the outdoor concentration is significantly lower than the indoor concentration. Even under these circumstances, however, a change in ventilation rate will affect the indoor concentration to a variable degree depending on the initial ventilation rate, the magnitude of other removal process, and the degree of coupling between the ventilation rate and the pollutant source strength. Figure 4 illustrates how a change in ventilation rate from a typical value of 0.5 ach affects indoor concentrations under various circumstances. A common assumption is that the indoor concentration is inversely proportional to the ventilation rate; the corresponding relationship is shown as curve C. As the rate of pollutant removal by processes other than ventilation is increased, the sensitivity of indoor concentration to a change in ventilation rate is reduced (compare curves C, E, and F or curves A and B). Because ventilation affects both the source strength of radon progeny (i.e., the indoor radon concentration) and the removal rate of radon progeny, the PAEC is even more sensitive to ventilation rate than the case of simple inverse dependence. However, in situations where the radon source strength is positively coupled to the ventilation rate, the PAEC will vary less rapidly than indicated in the figure. One additional curve shown in Figure 4 (curve D) relates air exchange rate and indoor formaldehyde concentration. The curve shown is for a building containing two formaldehyde sources (particleboard and medium density fiber-board) that are not separated from the indoor air by a permeation barrier. It is based on assumptions of no formaldehyde removal by reaction with surfaces, no change in indoor humidity or temperature, and a linear increase in formaldehyde source strength with a decrease in indoor concentration. The values chosen for M_i and b_i (see Equations 3 and 4) to generate the curve are based on data from Matthews et al. [4]. The point to note is that the coupling between formaldehyde source strength and indoor concentration can cause the indoor formaldehyde concentration to exhibit substantially less than a simple inverse dependence on ventilation rate.

The predominant trend in Figures 3 and 4 is the rapid increase in indoor pollutant concentration as air exchange rates become low. The energy required to provide ventilation increases approximately linearly with the ventilation rate, however, the benefits of an incremental increase in ventilation rate are greatest when the ventilation

rate is low. Avoiding unusually low air exchange rates is, therefore, a highly important component of any practical strategy for maintaining acceptable indoor air quality. Another implication of this trend is that moderate (i.e., practical) increases in the ventilation rate, such as by installation of readily available mechanical ventilation systems, can be expected to substantially reduce the indoor pollutant concentration only in buildings with initially low ventilation rates. In a building that is already well ventilated, it is logical to place a greater emphasis on strategies to reduce the pollutant source strength.

Localized Pollutant Removal

Some of the limitations of utilizing pollutant removal to control indoor pollutant concentrations were described in the previous section. The efficiency of pollutant removal processes can be substantially greater than indicated by the simple mathematical models in this paper when pollutant sources are localized (i.e., concentrated in space) and the removal processes are directed at the source. The most common example of this strategy is the use of local exhaust ventilation such as a range hood or a bathroom fan. Macriss and Elkins [23] found that operation of a range hood during cooking episodes (at an exhaust airstream flow rate of 93 1/s) reduced the increase in nitrogen oxides in the kitchen by only 30%. Traynor et al. [13], observed a greater effect, they reported that operation of a range hood at flow rates of 42 to 117 1/s reduced the amount of combustion products that entered the occupied space by 60 to 87%. Revzan [24] also reported high efficiencies for a range hood based on experiments with a heated tracer gas released in a manner to simulate the emission of pollutants. After conducting hour-long tests with a relatively low flow rate of air through the range hood of 60 1/s, the amount of tracer gas in the test space was as much as 77% less than theoretically predicted. The prediction was based on the flow rate of air through the hood and an assumption of perfect mixing of indoor air throughout the test, which is a convenient reference case.

The performance of other local pollutant removal processes, such as the use of bathroom fans to control moisture and local ventilation or air cleaning near sources of tobacco

smoke has not been fully documented. However, these processes are expected to be substantially more effective than providing an equivalent amount of ventilation or air cleaning for the entire occupied space.

Conclusions

In conclusion, control measures that are based on preventing high pollutant source strengths or reducing source strengths in existing buildings often deserve first consideration. It has been demonstrated that these measures are frequently capable of yielding large (i.e., a factor of five to ten) reductions in indoor concentration. In addition, source-control measures often involve acceptable initial costs, small increases in energy consumption, and minimal maintenance or other occupant participation. Some specific examples of source-control measures that may often be both effective and practical are the following: 1) exclusion of unvented combustion space heaters; 2) using only moderate quantities of formaldehyde-emitting building materials and choosing materials that emit relatively small amounts of formaldehyde; and 3) reducing radon entry by sealing leakage paths between the soil and the building interior and/or by crawl space or sub-slab ventilation.

Other basic strategies for indoor pollutant control are based on pollutant removal -- such as the removal that occurs by ventilation, air cleaning, or reactions of pollutants with indoor surfaces. In general, there is a practical upper limit on removal rates due to the associated requirements for energy, maintenance, and air movement. In addition, available data show clearly that one cannot ensure that indoor pollutant concentrations will be acceptably low by maintaining pollutant removal processes, such as ventilation, at a typical rate. Practical changes in pollutant removal rates can greatly affect the indoor concentration; however, once these removal processes reach a substantial level, further increases have only a limited effect on the indoor concentration. Active air cleaning and enhancement of natural removal processes, such as reactions of pollutants on surfaces, are potentially effective for some pollutants, but in buildings with a typical ventilation rate, even a relatively large and efficient air cleaner would typically reduce the indoor concentration by no more than a factor of two. The most important pollutant removal

process is usually ventilation and, as demonstrated in this paper, indoor pollutant concentrations generally increase dramatically as ventilation rates become very low. Therefore, avoidance of unusually low ventilation rates is essential for efficient pollutant control. The importance of avoiding low ventilation rates together with the impracticality of relying on large amounts of ventilation to control pollutant concentrations in buildings with high pollutant source strengths, may be sufficient justification for establishment of a minimum acceptable ventilation rate. It is important, however, not to equate the concept of an acceptable ventilation rate with the concept of acceptable indoor air quality. To ensure adequate indoor air quality requires that explicit attention be given to both limiting of pollutant sources and ventilation.

Acknowledgments

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Table 1. Reported values for the reaction constant and pentration factor.

| Pollutant	Reaction or Deposition Constant ± Standard Deviation (h^{-1})	Penetration Factor	Source
nitrogen dioxide	0.18, 0.29		12
nitrogen dioxide	0.20 ± 0.13		3
nitrogen dioxide	1.29 ± 0.67		13
nitrogen dioxide	1.39		14
nitrogen dioxide	0.16 to 0.5		15
nitrogen dioxide	0.83		16
formaldehyde	0.4 ± 0.24	1.0	17
particles*	0.48 ± 0:21	0.4 ±0.1	17
particles**	0.03 to 0.35		18

*   For particles with a diameter less than 0.5 um.
**   Value of constant is a function of particle size.
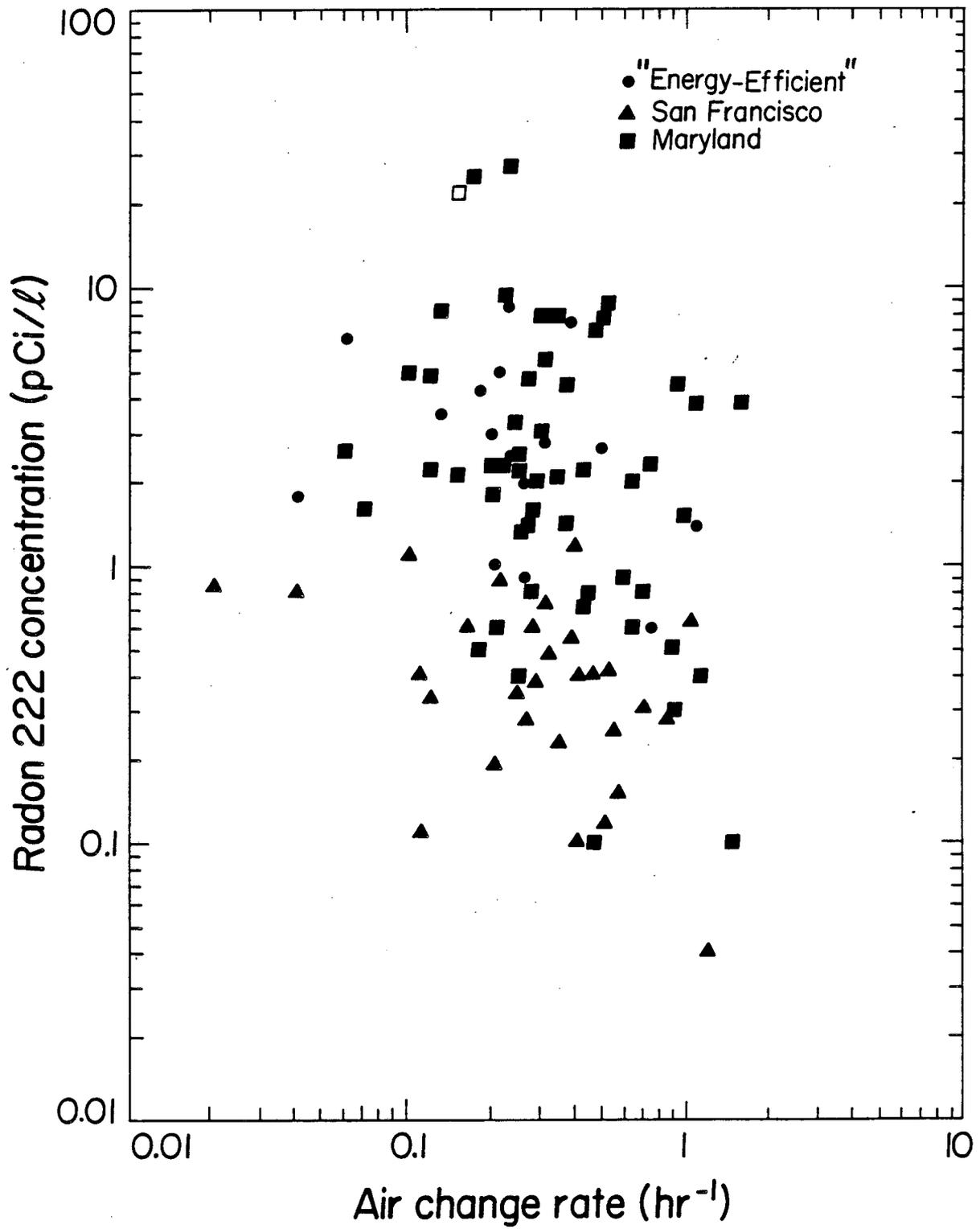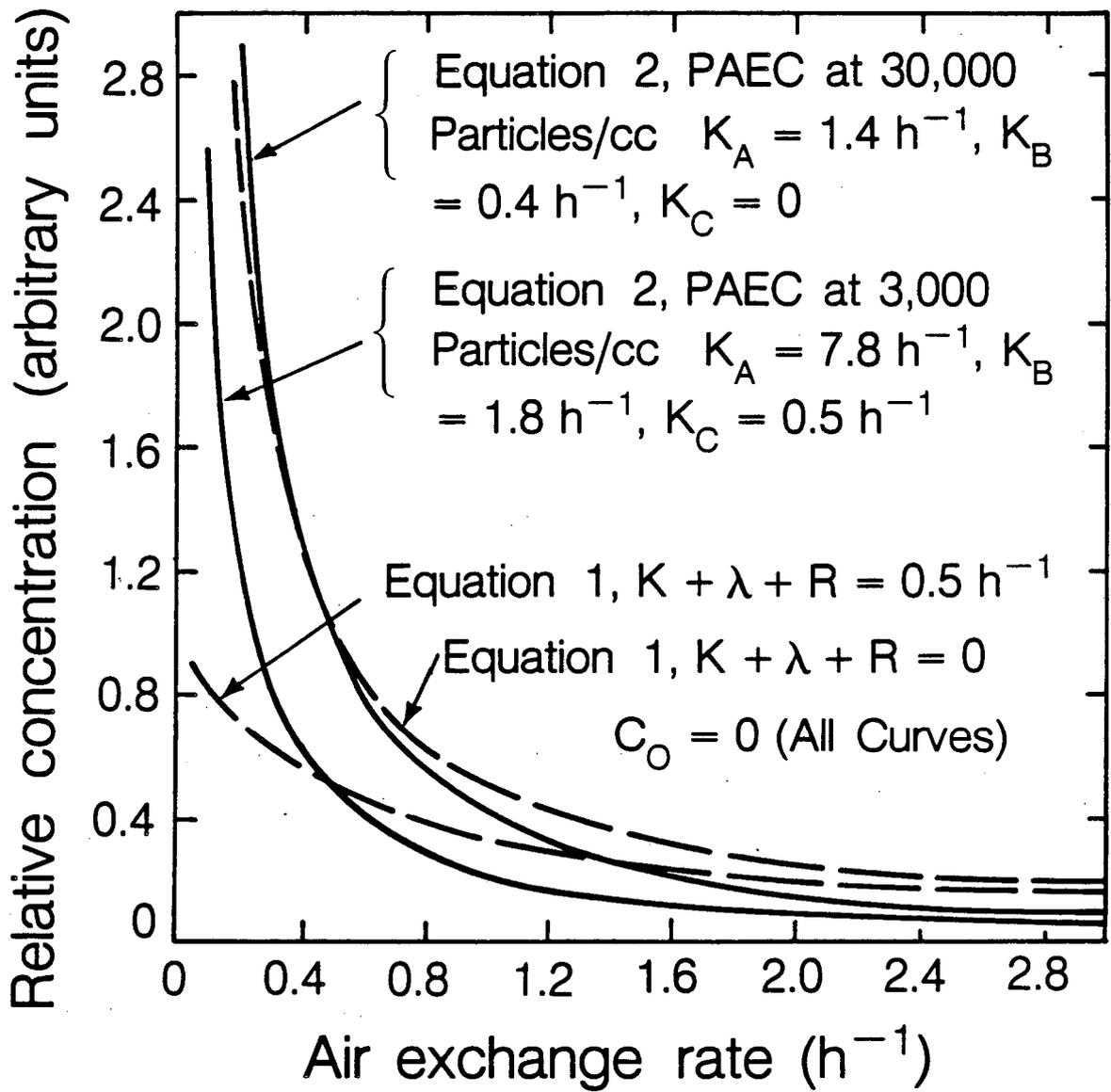
Figure 1. Scatter plot of radon concentration versus air exchange rate [1].

XBL 818-1115
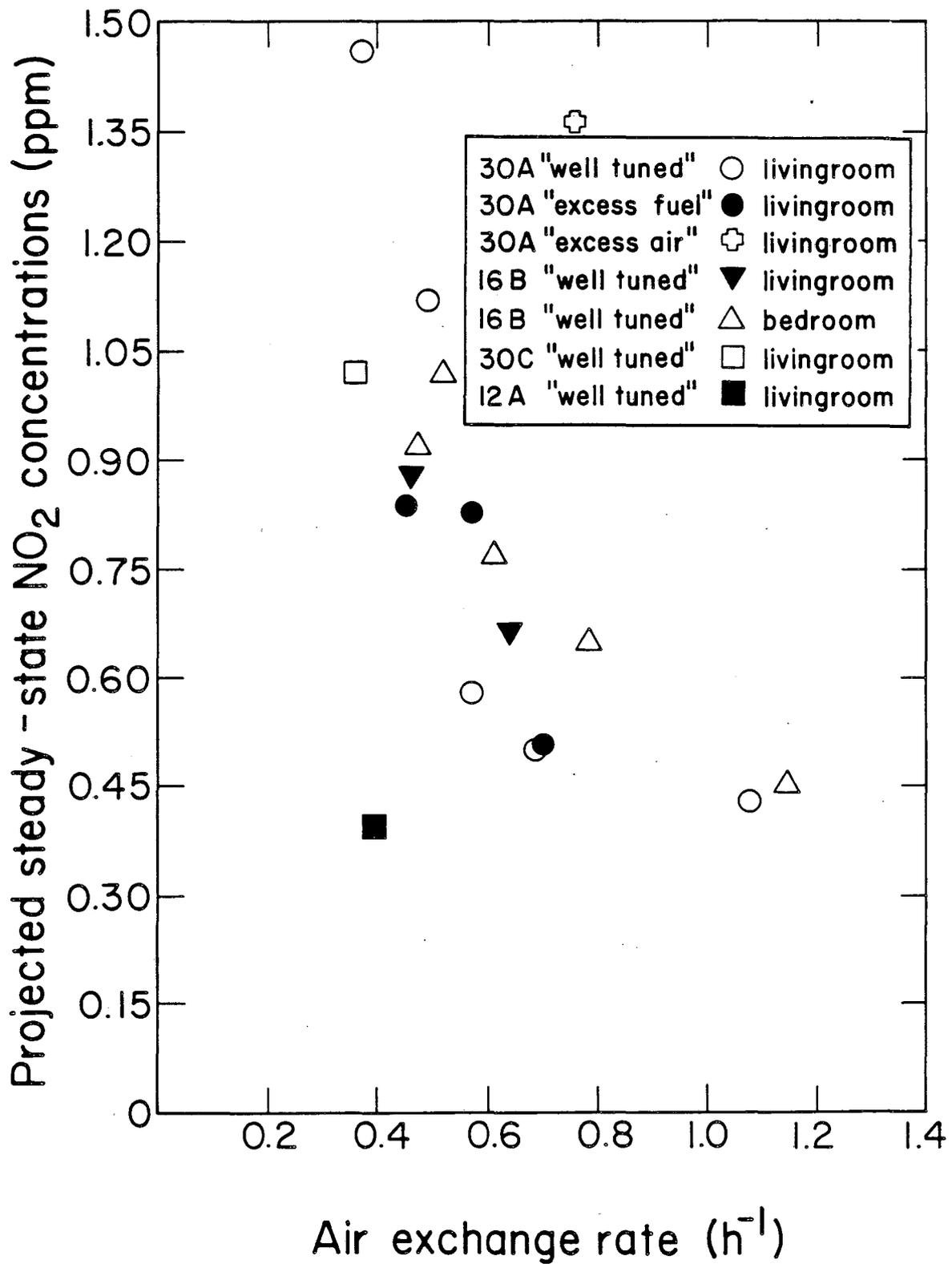
Figure 2. Nitrogen dioxide concentration versus air exchange rate in a house with various unvented gas-fired heaters [3].

XBL 834-9043

Figure 3. Relative pollutant concentration versus air exchange rate for variable rates of pollutant removal by processes other than ventilation.
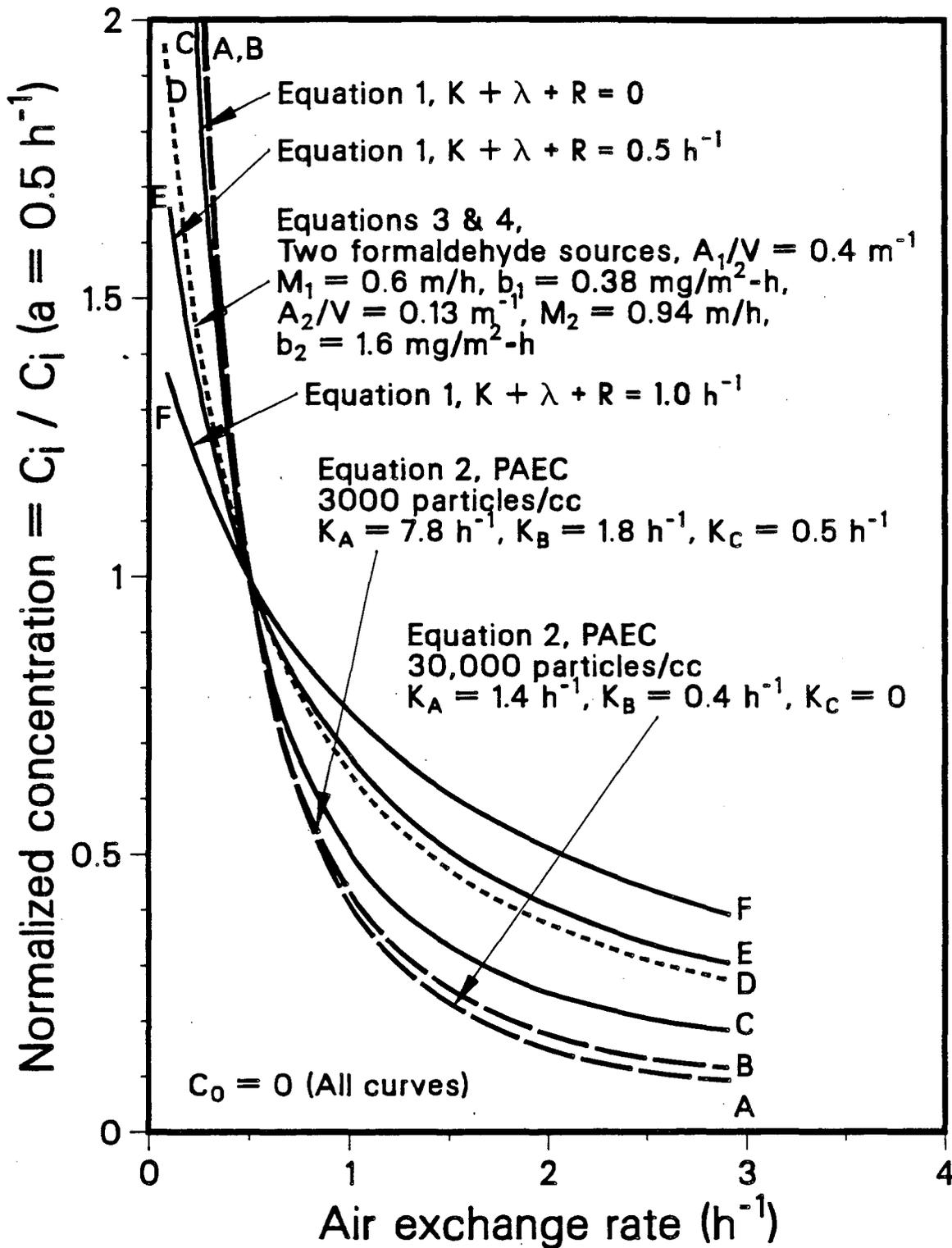
Figure 4. Predicted effects on indoor concentration of changing the air exchange rate from a typical value of 0.5 ach.

The figure plots Normalized concentration $= C_i / C_i \, (a = 0.5 \, h^{-1})$ on the vertical axis against Air exchange rate (h^{-1}) on the horizontal axis.

Curve labels and legend:

- Equation 1, $K + \lambda + R = 0$
- Equation 1, $K + \lambda + R = 0.5 \, h^{-1}$
- Equations 3 & 4, Two formaldehyde sources, $A_1/V = 0.4 \, m^{-1}$, $M_1 = 0.6 \, m/h$, $b_1 = 0.38 \, mg/m^2\text{-}h$, $A_2/V = 0.13 \, m^{-1}$, $M_2 = 0.94 \, m/h$, $b_2 = 1.6 \, mg/m^2\text{-}h$
- Equation 1, $K + \lambda + R = 1.0 \, h^{-1}$
- Equation 2, PAEC 3000 particles/cc, $K_A = 7.8 \, h^{-1}$, $K_B = 1.8 \, h^{-1}$, $K_C = 0.5 \, h^{-1}$
- Equation 2, PAEC 30,000 particles/cc, $K_A = 1.4 \, h^{-1}$, $K_B = 0.4 \, h^{-1}$, $K_C = 0$

$C_0 = 0$ (All curves)

XCG 8410-13399

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