

Does the Addition of Fly Ash to Concrete Present a Radon Hazard?

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Summary: Fly ash, a waste material from coal-fired power plants, can be added to concrete to increase durability and decrease cost. However, fly ash can contain radionuclides, including uranium-238, which will ultimately decay to form radon. In this paper, we present the results of a literature review that summarizes previous work on radon emission from fly ash-containing concrete. The results suggest no statistical correlation between fly ash content and radon emission, but a significant correlation when the specific activity of the fly ash is considered. However, when a simulation of a typical residential building is considered, fly-ash containing concrete is unlikely to represent a serious risk as it would barely elevate radon-222 concentrations in typical homes.

Keywords: radon, fly ash, concrete, residential buildings

Category:

1 Introduction

Residential radon exposure has been linked to increased lung cancer risk through several epidemiological and case-controlled studies [1-3]. The U.S. Environmental Protection Agency states that radon is the second leading cause of lung cancer for Americans, and the U.S. National Academy of Sciences reports that between 15,000 - 22,000 Americans die each year of lung cancer related to radon daughter-product exposure [4,5].

Concrete is made with cement and aggregates, both of which come from mineral deposits that may contain uranium-238. Concrete in buildings therefore contributes to indoor radon levels. The extent of radon released by concrete is characterized by its exhalation rate, which can be expressed in units of $\text{Bq}\cdot\text{kg}^{-1}\cdot\text{s}^{-1}$ or $\text{Bq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. It has been suggested that about 30% of the radon in indoor air originates in the concrete [6]; therefore knowledge of the radon emission from concrete is important for accurately assessing radiation exposure in buildings.

Many factors affect the rate of radon exhalation of concrete, including its constituent materials, age, and moisture content [7,8]. The rate of radon exhalation from concrete may also be changed by the addition of fly ash [9]. Fly ash is a waste material from coal fired power plants; when added to concrete, fly ash increases concrete durability and decreases cost. However, fly ash “typically contains elevated concentrations of trace heavy metals, hazardous organic compounds, and radionuclides,” including uranium-238 [10]. The radium-226 specific activity of fly ash can exceed that of the cement and aggregates used in concrete [11], but the effects on the overall radon exhalation rate of the concrete are unclear as a variety of different results have been

published. The work presented in this paper attempts to link the fly ash in a concrete mix to radon exhalation rates of concrete using experimental results from the literature, and then quantify whether there is an increased radon risk due to the presence of fly ash in concrete.

2 Literature Review

A wide range of radon exhalation rates for both regular concrete and fly ash-containing concrete have been reported throughout the literature due to the many differences in concrete properties including raw materials, porosity and physical structure, water-to-cement ratio, and curing conditions [12]. Studies have reached different conclusions regarding the influence of fly ash on concrete exhalation rates; some claim that fly ash causes exhalation rates to increase [11], decrease [7,9,13-16], or make no significant difference [12,17,18]. It is difficult to draw any decisive conclusions from these individual studies, given the disparity in observations. Drawing direct comparisons between results presented in different papers is challenging because of varying methodologies, sample sizes, and experimental goals. Nonetheless, an examination of existing data may shed some light on patterns of radon exhalation rates.

When examining data from the literature concerning exhalation rates of concrete it is important to keep in mind the properties of concrete that are known to affect exhalation rates. For example, low humidity conditions during curing reduce the radon exhalation rate of concrete, and high humidity, up to 75% relative humidity, increases the exhalation rate [7]. The age of concrete is another important factor, as the radon exhalation rate increases until the concrete is approximately one year old and then gradually decreases with time [7,12]. Furthermore, it is

hypothesized that the radium-226 specific activity, a measure of radioactivity, of the concrete materials has an effect on the exhalation rate [11].

Data from multiple published studies are compiled into Table 1, which reports concrete radon exhalation rates and radium-226 specific activity, as well as factors that may affect exhalation rates. Specific activity is an activity concentration per unit mass. The radium content in specific activity is equivalent to the total production rate of radon, with 1 Becquerel per kilogram ($\text{Bq}\cdot\text{kg}^{-1}$) of radium yielding radon at a rate of 1 atom $\text{kg}^{-1}\text{s}^{-1}$ [19]. We tabulate the exhalation rate on an area-normalized basis (also referred to as radon flux). For those papers that did not report an area-normalized exhalation rate, we calculated this value based on the ^{226}Ra specific activity of the concrete and an assumed slab thickness of 10 cm, based on the minimum value in the 2003 International Residential Code [20] and allowing for increased thickness near the slab edge. Based on the work of Siotis and Wrixon [11], the radon flux, J , can be calculated as:

$$J = S_{Ra} \cdot \lambda \cdot \rho \cdot \eta \cdot l_0 \cdot \tanh \frac{d}{l_0} \quad (1)$$

where S_{Ra} is the specific activity of ^{226}Ra in concrete in Bq kg^{-1} ; λ is the decay constant of radon ($2.1 \times 10^{-6} \text{ s}^{-1}$); ρ is the density of concrete (assumed to be 2500 kg m^{-3}); η is the radon emanating fraction (assumed to be 0.05); l_0 is the diffusion length in concrete (0.2 m); and d is the half thickness of the building element (assumed to be 0.05 m). With the exception of slab thickness, the radon and concrete parameters parenthetically listed above are equivalent to the values Siotis and Wrixon [11] used in their analysis..

Data from some published studies examining the effects of fly ash on concrete radon exhalation rates were not incorporated into Table 1. Generally, data were omitted if a paper neglected to mention enough defining characteristics about the concrete sample tested such as percent fly ash used, radium-226 specific activities of constituent materials, or concrete exhalation rates. Furthermore, an extensive study of the effects of fly ash by Kovler *et al.* [16] could not be included in Table 1 because the study examined cement pastes rather than concrete. Since a large portion of concrete is comprised of aggregates (approximately 80% by volume), the omission of aggregates will significantly alter the properties of the resulting material both in terms of its radium-226 specific activity and its porosity. Similarly, the work by Latona *et al.* [10] was not included because samples tested were of autoclaved aerated concrete, which has a significantly different porosity than conventional concrete used in building construction.

Table 1. Compiled radon data from the literature

Literature Source	Fly Ash (%)	Fly Ash Specific Activity ($\text{Bq}\cdot\text{kg}^{-1}$)	^{222}Rn Exhalation Rate ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}\times 10^{-4}$)
[9]	0	--	4.2
	25	244	2.8
[11]	0	--	1.0
	10	800	2.1
	20	800	3.6
	0	--	1.6
	10	330	6.8
[12]	20	330	2.8
	5	10.4	1.3
	10	11.2	1.4
	15	12.0	1.5
	20	12.8	1.6
	25	13.6	1.7
	30	14.4	1.9
[14]	25	15.2	2
	0	--	1.7
	15	290	2.3
	25	290	2.8
	35	290	3.3
	15	87	1.8
	25	87	1.8
	35	87	1.9
	15	165	2
	25	165	2.2
35	165	2.4	
[13]	0	N/A	6.9
[17]	0	N/A	6.1
[21]	0	N/A	10.8
	0	N/A	6.9
[7]	0	N/A	12.8
	0	N/A	1.5
	18	N/A	2.2
	0	N/A	1.4
	22	N/A	2.6
	0	N/A	1.5
	18	N/A	2.2
	0	N/A	1.4
[15]	22	N/A	2.6
	0	N/A	12.6
	5	N/A	12.7
	10	N/A	13.1
	15	N/A	13.4
	20	N/A	13.8
[18]	25	N/A	14
	0	N/A	98.3
	15	N/A	105.0
[22]	0	N/A	80
	25	N/A	80
	25	N/A	70

Ideally, one would combine all of the experimental data in Table 1 into a comprehensive meta-analysis or joint statistical model. However, such an approach rests on the assumption that factors are not confounded within experiments. For example, if all of the highest values for radon exhalation rate come from one study where only one cement was used which had a high ^{226}Ra specific activity and one fly ash was used which also had a high ^{226}Ra specific activity, it would be impossible to distinguish between the effects of fly ash or cement ^{226}Ra specific activity on exhalation rates. In the present data set, the data are not ideally randomized and do not adequately span the necessary range of inference. Furthermore, the data set is not ideal because there are unreported variables in almost every study. Therefore, analysis is limited to simple linear regression methods. However, a carefully performed linear regression analysis should provide limited information on the effects of single variables on radon exhalation rates in concrete.

As discussed earlier, there is disagreement in the literature as to whether or not the addition of fly ash to concrete increases exhalation rates. Plotting the exhalation rate from the data in Table 1 versus the percentage of fly ash provides insight into these inconsistencies, Figure 1. Across the data in the studies examined, increasing the amount of fly ash does not appear to influence radon exhalation rates as the slope is not statistically significantly different than zero, $P > 0.5$. This is consistent with the conclusions of some of the individual studies and inconsistent with others. It is likely that variables other than fly ash amount cause this discrepancy in conclusions. Additionally three studies [18,22,23] measure exhalation rates from concrete containing aggregate from Hong Kong with very high specific activity [24,25]. Given that aggregate is a much larger mass fraction of the concrete than the fly-ash containing cement, these concretes have exhalation rates that are 4 to 5 times larger than the next nearest exhalation rates and over an order of magnitude greater exhalation rates than most of the concretes in Table 1. The effect of the aggregate overwhelms any effect due to fly ash and thus these concretes are excluded from the subsequent analysis. Additionally, one concrete sample from Siotis and Wrixon [11] has a very high exhalation rate and was excluded from the rest of the analysis as it likely represents measurement variability or radon emission due to a concrete constituent besides fly ash. There are two regression lines shown on Figure 1, the thin dotted line includes all data that reported exhalation rates and fly ash contents, and the thick solid line excludes the data as described above. Neither the full, nor the refined data set suggests a statistical correlation.

The relationship between fly ash ^{226}Ra specific activity and concrete radon exhalation rates is shown in Figure 2. In this case, it is clear that the ^{226}Ra

specific activity of the fly ash has a significant effect on the concrete radon exhalation rate. There is a significant linear relationship between ^{226}Ra specific activity of fly ash and concrete radon exhalation rate (slope = $2 \times 10^{-7} \text{ kg m}^{-2} \text{ s}^{-1}$, $R^2 = 0.47$, $P < 0.001$). We are more confident that this is a significant relationship because fly ash ^{226}Ra specific activity is not seriously confounded with other variables in the data set. While fly ash ^{226}Ra specific activity explains approximately half of the variation in the data, the remainder of the variation in the data might be explained by variables such as the quantity of fly ash used, the mass fraction of the cement in the concrete, the sample age, the moisture content of the concrete, the radium contents of the other constituent materials, and the testing methods used.

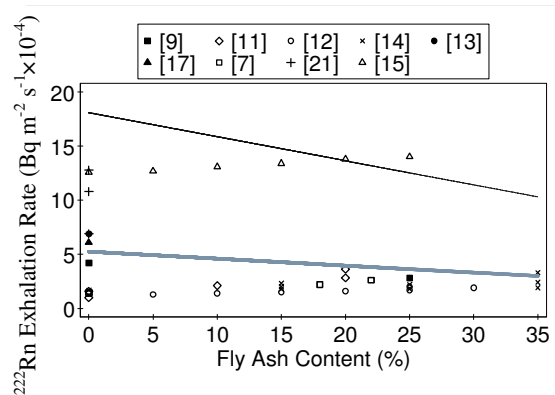


Fig. 1. Concrete radon exhalation rate as a function of fly ash content for all applicable studies described in Table 1. References [18,22,23] excluded from plot but not from data analysis.

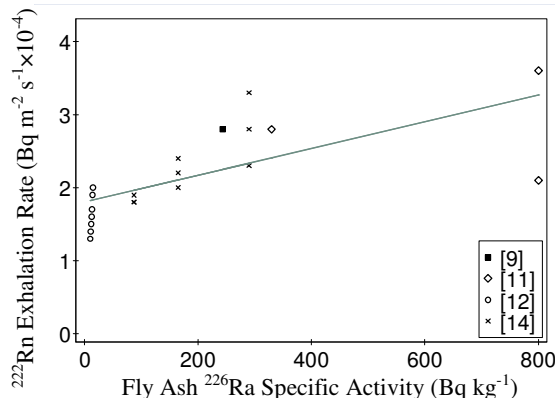


Fig. 2. Concrete radon exhalation rates versus fly ash ^{226}Ra specific activity for four studies that report both variables.

An even clearer correlation is found when radon exhalation rate is regressed on the product of fly ash content and ^{226}Ra fly ash specific activity (Figure 3). This product is a measure of the total specific activity associated with the fly ash in the concrete. Although only the same four studies present data that allows for calculation of this product, it explains most of the variation of the data (slope = $1.4 \times 10^{-6} \text{ kg m}^{-2} \text{ s}^{-1}$, $R^2 = 0.85$, $P < 0.0005$).

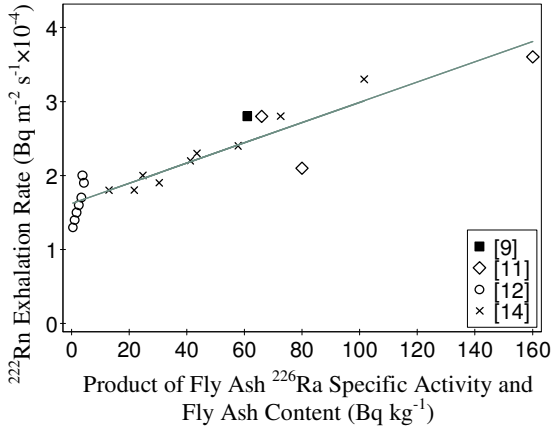


Fig. 3. Concrete radon exhalation rates versus product of fly ash ^{226}Ra specific activity and fly ash content.

Based on the simple analysis described above, it can be stated that fly ash increases the radon exhalation rate of concrete, with the effects being highly dependent on the ^{226}Ra specific activity of the fly ash used. One of the reasons that this effect has not been extensively acknowledged previously is because many studies only examined one type of fly ash and didn't specifically investigate the effects of ^{226}Ra specific activity of the constituent materials on exhalation rates.

3 Model and Input Parameters

A goal of this investigation is to determine whether fly ash-containing concrete poses a health risk when used in typical residential buildings. To evaluate this we simulated a residence using a well-mixed reactor model. In addition to well-mixed conditions, we also assumed steady-state conditions, limited removal of ^{222}Rn and daughter products because of attachment indoor aerosols, and limited interference with radon emission because of floor and wall coverings. These are reasonable assumptions given the work of Siotis and Wrixon [11] and Nazaroff and Nero [19]. We also assumed that the only radon source was a 10 cm slab; any additional sources will lead to higher radon concentrations.

The steady-state radon concentration in the room, C_{SS} , is calculated as:

$$C_{SS} = \frac{AJ}{Q} \quad (2)$$

where A is the surface area of the slab in m^2 and Q is the ventilation rate in $\text{m}^3 \text{s}^{-1}$.

In order to determine the model parameters for Equation 2, we used values from the literature to determine the radon exhalation rate and the other parameters. We are primarily interested in airborne concentrations of ^{222}Rn and are not considering the formation and fate of daughter products. This is a reasonable assumption given that the air exchange

rate ($5.5 \times 10^{-5} \text{ s}^{-1}$) is an order of magnitude greater than the decay constant ($2.1 \times 10^{-6} \text{ s}^{-1}$) [19].

The volume of a typical home in the U.S. was selected by considering the typical home area from the 2001 American Housing Survey [26] of 157 m^2 and multiplying it by an assumed ceiling height of 2.4 m, for a volume of 377 m^3 . For purposes of assessing the concrete area for radon emissions, we considered single story slab-on-grade construction and thus the entire floor area was associated with concrete. Given that radon concentrations are typically higher in basements, we also considered the case of a basement with one quarter the total volume of the house (94 m^3) and one half of the concrete surface area (79 m^2).

The air exchange rate (λ) was determined from a study of 2,844 American homes by Murray and Burmaster [27]. We considered their 10 – 90% range of air exchange rates of 0.2/hr and 1.3/hr, with a typical value of 0.5/hr.

The radon flux was based on a comprehensive review of exhalation rates in the literature and is discussed in more detail in the Literature Review section. Based on our literature review, we considered three cases for the exhalation rate: the maximum reported radon flux $5.0 \text{ Bq m}^{-2} \text{ hr}^{-1}$ [15] and the median and mean values of $1.8 \text{ Bq m}^{-2} \text{ hr}^{-1}$ and $0.79 \text{ Bq m}^{-2} \text{ hr}^{-1}$ from all concretes that contain fly ash in Table 1.

4 Results

Figure 4 shows the indoor steady-state radon concentration as a function of air exchange rate for the three different radon flux conditions in both basement and whole house volumes. The air exchange rate has a strong influence on air concentrations with air exchange rates above 1 h^{-1} resulting in ^{222}Rn concentrations of below 5 Bq m^{-3} in both the house and the basement for all exhalation rates. For air exchange rates below 0.25 hr^{-1} , the radon concentrations are close to 20 Bq m^{-3} for the Basement Case and close to 10 Bq m^{-3} for the Whole House Case for the highest exhalation rate and proportionally lower for the smaller fluxes.

To put the curves in Figure 4 in context, typical outdoor ^{222}Rn concentrations in the United States range from $4 - 15 \text{ Bq m}^{-3}$, about 20% of the average indoor concentration of 48.1 Bq m^{-3} [28]. Thus, the contribution of fly ash-containing concrete to indoor radon concentration is approximately equivalent to that coming from outdoor air when considering a mean air exchange rate of 0.5 hr^{-1} . The United States Environmental Protection Agency has set an action level of 148 Bq m^{-3} [5], with other nations selecting different levels. It is recommended that homes with larger concentrations be fixed, and even homes with a $74 - 148 \text{ Bq m}^{-3}$ concentration may require remedial action, depending on the homeowner's risk tolerance.

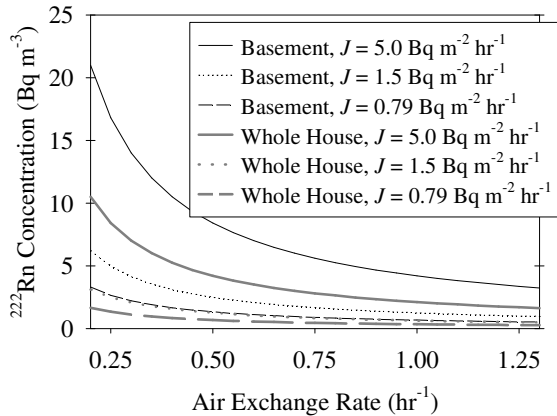


Fig. 4. Indoor Radon concentrations that would result solely from the use of fly ash-containing concrete.

The results in Figure 4 indicate that for very tight homes, the use of high ^{222}Rn specific activity fly ash containing concrete will elevate basement ^{222}Rn concentration by approximately 20 Bq m^{-3} . Although this worst-case scenario does represent an increased risk, it is small and unlikely to be statistically significant, given typical occupancy patterns and a recent review of the health literature [3]. Given that many concretes have considerably lower exhalation rates than this maximum value and many homes have higher air exchange rates, the typical health risk from the use of concrete containing fly ash is likely very small. However, there are some concretes that have very high exhalation rates, such as those tested by Yu *et al.* [18,22,23], because they contain aggregate with high specific activity. Use of concretes such as these, whether they contain fly ash or not, would elevate the radon risk in many homes.

Given the increased focus on green building, material reuse, and alternative materials, there will likely be increased demand for concrete that contains fly ash. As one example, the Leadership in Energy and Environmental Design (LEED) Green Building Rating System gives credit for the reuse of material in construction [29]. The use of fly ash in concrete would contribute to fulfilling Credits 3.1 - 4.2 in the Materials and Resources section of the LEED Rating System [29]. This analysis suggests for the concretes and scenarios considered, the use of fly ash is unlikely to present a significant health risk. Having said that, the use of a concrete with a higher ^{226}Ra specific activity and higher fly ash content than those summarized in Table, a larger concrete surface area or a smaller volume than modeled here may present a health risk, particularly in a building with a very low air exchange rate. However, for most buildings and scenarios, this risk seems small compared to other indoor air quality issues, including the indoor radon contribution from soil.

Our review of the literature suggests a wide range of radon exhalation rates from fly ash-containing concrete. The addition of fly ash to concrete does not necessarily increase the amount of radon exhalation

for that concrete. In some cases the exhalation rates rise slightly, in others they decrease, and it is not predictable based on fly ash percentage alone. When the specific activity of the fly ash is considered, there is a significant statistical correlation with radon exhalation rates. The predictive power increases further when the product of fly ash content and fly ash specific activity is considered.

A worst-case analysis suggests that the radon related to emission from concrete can be a measurable component of the overall radon concentration in a home with a very low air exchange. However, occupants of a residence with a typical air exchange rate are unlikely to see a significant increased health risk from fly ash-containing concrete. However, the use of a higher amount of fly ash with a higher ^{226}Ra specific activity than those in the studies cited here, may lead to a significant health risk. A more complete dataset of exhalation rates, particularly with measurement of the specific activities of all concrete components, would allow for more complete understanding of radon exhalation rates in concrete. In order to achieve this, at least one study should be performed that explores a full range of values in all of the variables of interest in a well-randomized, orthogonal study. Such a study, particularly with a field component, could provide confirmation of the findings of insignificant health risks.

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