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BEHAVIOR OF RADON PROGENY NEAR OUTDOOR SURFACES IN CONTRASTING TERRAINS

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ABSTRACT

Federal legislation expresses a long-term goal of reducing indoor radon to levels comparable to those outdoors. A thorough understanding of the behavior of radon and its progeny in the outdoor environment is therefore important. We report near-surface measurements of radon, attached-to-aerosol progeny, and unattached-to-aerosol progeny in the contrasting environments of the forested hill country of Oak Ridge, TN and the desert sands of White Sands National Monument, NM. Vertical progeny gradients are greater at White Sands due to a smoother terrain, and dose levels tend to be lower due to lower radon flux from the gypsum sand. Both sites show a significant diurnal variation of dose rate with a maximum occurring usually in the early morning. Correlation of dose rate variation with radon variation is surprisingly small suggesting the importance of other factors such as progeny deposition and aerosol concentration in controlling outdoor dose.

INTRODUCTION

Federal legislation expresses a long-term goal of reducing indoor radon to levels comparable to those outdoors. In an earlier paper (Wasiolek and Schery, 1993), based on measurements at Socorro, NM, we pointed out that outdoor radon progeny doses typically exceed background from the nuclear power industry and are comparable to doses from medical procedures. In this paper, we discuss measurements of radon, attached-to-aerosol progeny, and unattached-to-aerosol progeny at two new sites with contrasting terrains and climates. The first is the Walker Branch Watershed site near Oak Ridge, TN and the second is White Sands National Monument near Alamogordo, NM. The focus is on what factors control radon, its progeny, and dose from radon progeny near the breathing level where nearby surfaces (ground, foliage, etc) play an important role in controlling disequilibrium. While there exist many measurements of radon in the outdoor environment, previous measurements of attached and unattached progeny, which are necessary for estimating dose and understanding removal mechanisms for progeny, are quite limited.

METHOD

Experimental Measurements

Measurements were taken from 21 July 1992 to 28 July 1992 at the Walker Branch Watershed near Oak Ridge, TN (Oak Ridge site) and from 28 May 1993 to 29 May 1992 at the White Sands National Monument (White Sands site). The Oak Ridge site, operated by the National Oceanic and Atmospheric Administration, is located at latitude 35°57'30" North and longitude 84°17'15" West. The field site is at the top of a ridge at an altitude of 365 m in undulating terrain. The soil is a cherty silt loam and the site and surrounding area are covered by a forest of oak, hickory, and loblolly pine with a mean canopy height of about 23 m. The climate is subtropical-humid and average yearly rainfall is about 130 cm. Using the guidelines of McRae et al. (1982), a terrain roughness length of about 200 cm was estimated for this site. Further information can be found in Baldocchi and Meyers (1991). At White Sands the measurements were taken in an unoccupied picnic area located at about 32°49' North by 106°16' West at an altitude of about 1250 m. The surface for several kilometers in all directions is covered by unvegetated gentle dunes of gypsum sand. The climate is semi-arid with the average yearly rainfall for this region of NM about 25 cm. The estimated roughness length is about 8 cm.

Basic data sets collected at each site consisted of activity concentration measurements of attached and unattached progeny at 0.2 m and 2.2 m above ground, radon and aerosol concentration at 1 m, and wind speed and wind direction several meters above ground. Radon was measured with a 36-L two-filter system. Attached and unattached progeny were measured with high volume air samplers using about 400 L min^{-1} flow with filters and screens of 10-cm diameter. Four-hundred-mesh screens provided measurements of unattached progeny with a 50% penetration diameter of about 1.4 nm. Gelman type A/E glass filters provided the measurements of the attached progeny. Both filters and screens were counted for total alpha particles on commercial scintillation counters where individual progeny could be deduced from the rate of reduction of the count rate with time (Knutson, 1989). Sampling intervals were usually 18 min for progeny and 120 min for radon at the Oak Ridge Watershed and 30 min at White Sands. Flux density measurements were made with a 30-cm high 50-cm diameter closed accumulator using a small commercial ionization chamber inside (Femtotech, R210F). A commercial unit (Rich 200) was used to make condensation nuclei measurements. The absolute accuracy of the progeny measurements was estimated to be better than ±10%; relative accuracy for a high/low pair of measurements should be better than ±3%. The absolute accuracy of radon measurements should be better than ±15%. Further details of the experimental procedures can be found in Schery and Wasiolek (1993) and Wasiolek and Schery (1993).

Data Reduction

For assistance in interpretation, much of the data is reported in terms of quantities derived from the basic measurements. Atmospheric stability is based on standard deviation of the wind direction using guidelines of USEPA (1986). This procedure yields Pasquill stability classes ranging from {1} = extremely unstable to {7} = extremely stable. Radon and individual progeny are determined in units of Bq m^{-3}. A collective measure of all progeny can be reported using Potential Alpha Energy Concentration (PAEC). The relation between PAEC in nJ m^{-3} and progeny concentrations in Bq m^{-3} used in calculations was

$$PAEC = 0.579\, C(^{218}Po) + 2.86\, C(^{214}Pb) + 2.10\, C(^{214}Bi). \quad (1)$$

To aid in the interpretation of the data we report the ratio of total PAEC at 2.2 m to that at 0.2 m, denoted by RATT. We also report a similar ratio, but for unattached progeny only, denoted by RATU. The equilibrium factor F2M, a measure of the extent of disequilibrium between radon and its progeny, was calculated for a 2.2 m height using F2M = $0.178*PAEC/C(^{222}Rn)$ where the radon concentration at 1 m was considered a sufficient estimate of that at 2.2 m. The symbol FU2M denotes the ratio of unattached PAEC at 2.2 m to total PAEC at 2.2 m. The notation $^{214}Bi/^{214}Pb$ is for the total activity ratio of $C(^{214}Bi)/C(^{214}Pb)$ at 0.2 m. The notation $^{214}Pb/^{218}Po$ is for the corresponding activity ratio of $C(^{214}Pb)/C(^{218}Po)$ at 0.2 m. Condensation nuclei concentration (particles per unit volume), radon concentration, stability index, wind speed, and roughness length are denoted with CN, RN, STAB, WS, and ROUGH. Calculation of dose from radon progeny is an evolving subject with variable results depending on the dosimetry model used. Here we estimate effective dose equivalent rate at 2.2 m, denoted DOSRA2M, using adaptation of the approach of NRC (1991) which assigns a higher weighting to the effects of unattached progeny versus attached progeny. Assuming a quality factor of 20 and an organ weighting factor of 0.06 gives for the dose rate in nSv h^{-1}

$$DOSRA2M = 27.7*PAECU + 2.2*PAECA \quad (2)$$

where PAECU is the PAEC due to unattached progeny and PAECA is PAEC due to attached progeny. Statistical screening of the data was carried out with the statistical package NCSS (Hintze, Kaysville, Utah 84037).

RESULTS

Table 1 summarizes the gross statistics for the two sites. There were 56 measurement sets for the Oak Ridge site and 20 measurement sets for the White Sands site. Data were not always available continuously at both sites due to equipment failures and operational constraints, but there is approximately equal representation of all times of day and night. The symbol σ_m stands for standard deviation of the mean so that any listed means that differ by more than this value have a statistically significant difference. Using this criterion, almost all mean results for White Sands versus Oak Ridge show a statistically significant difference except for the unattached fraction FU2M and the stability index STAB. The average dose rate at White Sands (54 nSv h^{-1}) was significantly less than that at the Oak Ridge site (133 nSv h^{-1}). The average gradient at White Sands for unattached progeny (RATU) was decreasing downward (indicating a downward directed progeny flux) while at the Oak Ridge site there was no detected direction of the mean flux within sampling error.

Fig. 1 presents dose rate at 2.2 m (DOSRA2M) versus (local) time of day for the Oak Ridge site. Data taken at the same time of day but on different days will thus appear with the same abscissa. A notable diurnal trend is observable with a maximum occurring in the early morning and a minimum occurring in the late afternoon. The data for White Sands (not shown) showed a similar trend. Fig. 2 is a similar plot for the equilibrium factor at 2.2 m at the Oak Ridge site. A diurnal pattern is also evident but not as strong as in Fig. 1. Table 2 shows single-parameter correlations between dose rate and selected variables at both sites. The strongest consistent correlation with does rate at both sites is PAEC, time of day, then stability index. The correlation with radon concentration at both sites is surprisingly weak. Table 3 shows some correlation results at the Oak Ridge site for some of the underlying dependent and independent variables that relate to the behavior of radon and its progeny near the earth's surface. Most correlations seem weak with the exception of increasing radon with stability (Fig. 3) and increasing ratio $^{214}Bi/^{214}Pb$ with increasing wind speed (Fig. 4).

Flux density measurements were not taken at the Oak Ridge site. Based on soil type, regional studies, and average ground level radon concentrations one would expect an average flux density close to the average for temperate continental land of about 20 mBq m^{-2} s^{-1}. Measurements of flux density at the White Sands site proved below the limit of detection, about 5 mBq m^{-2} s^{-1}. It is reasonable to conclude that the local flux density is significantly lower at the White Sands site, although radon travels horizontally for great distances and outside the dune area flux densities in NM are close to the continental average (Wilkening et al, 1972).

DISCUSSION

We first comment on some of the trends common to both sites. Both sites show a significant diurnal trend for dose rate with a tendency for a maximum in the early morning and a minimum in the late afternoon. This trend is not unexpected and has been documented before for radon gas, although not for dose calculated in terms of both attached and unattached progeny. The explanation is that, barring special events such as passage of a weather front, ground level air is usually most stable at night with less vertical mixing. Hence there is more likely to be a build up of radon and progeny near ground at this time. Supporting this interpretation is the positive correlation of dose rate with atmospheric stability as indicated in Table 2. A somewhat surprising result is the modest correlation of dose rate with radon as evidenced in Table 2. Since radon is the progenitor of progeny, at some point absence of radon must force absence of progeny. However, dose rate is also controlled by removal processes of progeny such as dry deposition, and the distribution between attached and unattached states which depends strongly on aerosol concentration. Progeny (and hence dose) can have a significant gradient outdoors even when radon, which does not experience plate out, is relatively well mixed. Apparently these processes are playing a part and complicating the picture of dose rate variation with position and time. One implication of this result, suggesting further investigation, is that outdoor measurements of radon in combination with a fixed equilibrium factor or constants for conversion to dose may not be a reliable indication of outdoor dose. Measurement of total PAEC is fairly easy and more directly correlated with dose rate (see Table 2). This correlation is not surprising in light of Equ. 2. Apparently the partition between the attached and unattached states is not varying wildly. In any case, measurement of total PAEC seems a superior indicator of dose compared with simple measurement of radon gas.

On theoretical grounds one would expect a negative correlation between dose rate and aerosol concentration since increased aerosol should reduce the fraction of unattached PAEC with its higher dose conversion factor. The lack of such correlation in Table 2 is puzzling and requires further investigation. The results for White Sands might be discounted because of the relatively small sample size. However, there was an adequate sample size for the Oak Ridge site and its correlation is too small to be significant. Preliminary multi-variable analysis suggests that a paracorrelation may be present. For example, if the effect of total PAEC is removed, a stronger negative correlation between dose and CN results. Our data show somewhat less volatility in the equilibrium factor compared with dose rate or radon. This result has been observed before and is supported by theoretical modeling as long as the aerosol concentration does not get small (CN $<< 5000$ cm^{-3}, Schery and Wasiolek (1993)).

Table 3 indicates that of the studied variables the most significant correlations were between radon and stability (positive) and the activity ratio $^{214}Bi/^{214}Pb$ and wind speed (positive). A similar result (not shown) was found for the White Sands site. The result for radon with stability is expected since greater stability decreases the vertical mixing of radon reducing the dilution of this gas which has soil as its source. The explanation for the progeny ratio is more complex and is related to an increase in deposition velocity to nearby surfaces. This effect has been successfully modeled in a recently developed two-particle group size model for the vertical distribution of radon and its progeny (model RPOUT, Schery and Wasiolek, 1993). The remaining correlations are weaker and are not necessarily adequately understood. Suffice it to say that the concentration of radon progeny in the outdoor environment depends on a complex interplay of factors (radon concentration, wind, stability, aerosol concentration, surface roughness) under atmospheric conditions that introduce fluctuation due to turbulent conditions.

One of the most striking differences between the two sites is the much higher average dose rate at the Oak Ridge site by a factor of about 2.5. Continuous year-round exposure at the Oak Ridge site at the July outdoor rate would result in a yearly dose of about 1 mSv. This is comparable to the average yearly dose from medical procedures (UNSCEAR, 1988). This result would vary, of course, depending on the chosen dosimetry model. The most important factor for lower dose at White Sands is almost undoubtedly the lower parent concentration of radon mostly due to lower flux density from ground. Average stability and CN concentration at both sites are comparable and not likely to be a factor. The wind speed is modestly higher at the White Sands site, and the resulting higher deposition velocity (predicted by RPOUT) could also be a factor reducing progeny concentrations and dose.

From the pure physical science point of view, one of the more interesting results is the lack of a downward directed unattached progeny gradient at the Oak Ridge site (mean RATU in Table 1) versus a clearly downward directed gradient for White Sands. The model RPOUT provides an explanation for this because surfaces provide a good sink for unattached particles (good sticking). For White Sands this sink is concentrated at the ground resulting in a gradient above. At the Oak Ridge site, since measurements were inside the forest canopy, surfaces are more uniformly distributed around the measurement point resulting in less anisotropy. The gradient for total progeny at both sites is small (RATT ~ 1 in Table 1). The explanation in the context of RPOUT is that at these sites total PAEC is primarily attached PAEC. Attached PAEC does not readily diffuse through laminar air layers near surfaces resulting in nearby gradients. The lower equilibrium factor at the Oak Ridge site (F2M in Table 1) might be due to the greater nearby surface area (greater roughness length) resulting in greater removal of radon progeny. The progeny/parent activity ratio for $^{214}Pb/^{218}Po$ is significantly greater than one for the White Sands site. RPOUT predicts how this can occur. It is involves a greater unattached fraction for ^{218}Po compared with ^{214}Pb resulting in greater removal of ^{218}Po to a nearby surface.

CONCLUSIONS

1. The average dose rate at the Oak Ridge site was 133 nSv h^{-1} compared with 54 nSv h^{-1} for the White Sands site. The major explanation for the difference is probably the lower radon flux from the gypsum sand at White Sands.

2. At both sites there was a strong diurnal pattern in dose with a maximum common in the early morning and a minimum common in the late afternoon. An important factor causing this is greater vertical mixing of air that frequently occurs in the late afternoon due to greater atmospheric instability from solar heating of the ground.

3. The dose rate showed a much stronger correlation with the total potential alpha energy concentration (PAEC) than with radon gas itself. This result stresses the importance of other factors besides radon in controlling dose (removal processes, aerosol concentration, etc.) and suggests that simple measurements of outdoor radon combined with a fixed conversion factor may not be an adequate predictor of dose. Measurement of total PAEC may therefore be a better indicator for dose rate.

4. The unattached progeny showed almost no vertical gradient at the Oak Ridge site but a significant downward directed gradient at the White Sands site. The probable explanation is the anisotropy for the White Sands site with lower roughness and a uniform surface at the lower boundary, in contrast to a measurement position inside the forest canopy at the Oak Ridge site with surrounding trunks and leaves above and below.

5. The mean equilibrium factor for the Oak Ridge site (0.58) was lower than that for the White Sands site (0.78) probably due to the larger available surface from the forest increasing dry deposition (plate out).

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Table 1. Summary of measurements at Oak Ridge (OR, 56 samples) and at White Sands (WHS, 20 Samples).

	Units	Median		Mean		σ_m		Range	
		OR	WHS	OR	WHS	OR	WHS	OR	WHS
DOSRA2M	nSv h^{-1}	95	52	133	54	13	4	39-433	18-91
RN	Bq m^{-3}	8.1	2.6	8.4	3.1	0.3	0.4	4.1-14.0	1.0-7.6
F2M		0.54	0.71	0.58	0.78	0.03	0.09	0.26-1.35	0.26-1.61
FU2M	%	7.2	10.9	10.1	11.0	1.1	0.5	1.3-36.6	6.2-15.7
RATU		0.88	1.31	0.99	1.40	0.05	0.18	0.26-2.42	0.60-4.48
RATT		1.00	1.05	0.98	1.05	0.01	0.02	0.73-1.09	0.87-1.25
$^{214}Bi/^{214}Pb$		0.87	0.99	0.85	1.00	0.02	0.02	0.51-1.01	0.76-1.29
$^{214}Pb/^{218}Po$		0.95	1.29	1.05	1.65	0.06	0.29	0.40-2.40	0.43-6.33
CN	10^3 cm^{-3}	6.1	6.0	7.2	6.3	0.6	0.6	1.1-25.0	2.5-13.0
STAB		2.5	1.5	2.5	3	0.2	0.5	1-5	1-6
WS	m s^{-1}	0.34	1.38	0.31	1.63	0.02	0.29	0.20-0.67	0.39-6.58

Table 2. Correlation results for DOSRA2M at Oak Ridge and White Sands.

	STAB	WS	CN	RN	TIME OF DAY	PAEC
DOSRA2M (Oak Ridge)	0.29	-0.20	-0.02	0.13	-0.44	0.73
DOSRA2M (White Sands)	0.41	-0.01	0.41	0.25	-0.41	0.97

Table 3. Correlation results for underlying variables at Oak Ridge.

	CN	STAB	WS
RN	-0.09	0.47	-0.28
FU2M	-0.11	0.25	-0.22
F2M	0.18	0.00	-0.02
RATU	0.01	-0.19	0.11
RATT	0.14	0.03	0.31
$^{214}Pb/^{218}Po$	0.18	-0.22	0.25
$^{214}Bi/^{214}Pb$	0.24	-0.12	0.46

Fig. 1 presents dose rate at 2.2 m (DOSRA2M) versus (local) time of day for the Oak Ridge site. Data taken

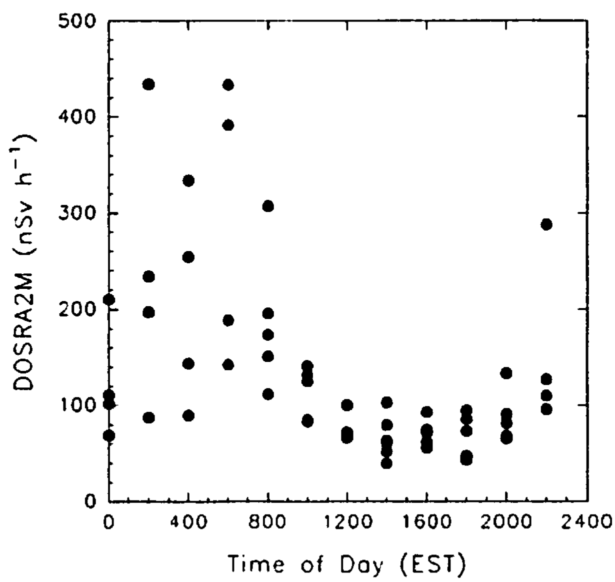

Figure 1. Dose rate at 2.2 m versus time of day for the Oak Ridge site.

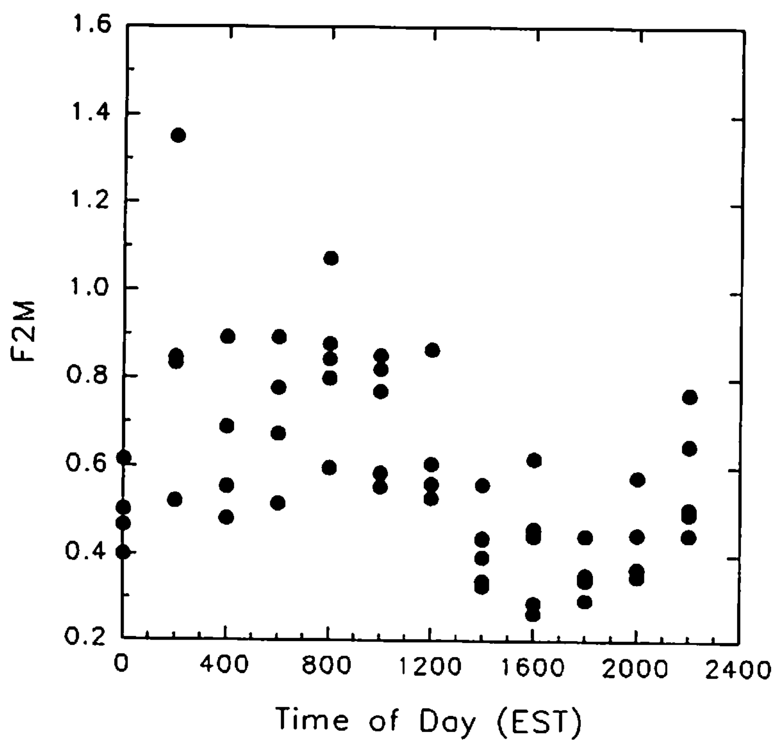

Figure 2. Equilibrium factor versus time of day for the Oak Ridge site.

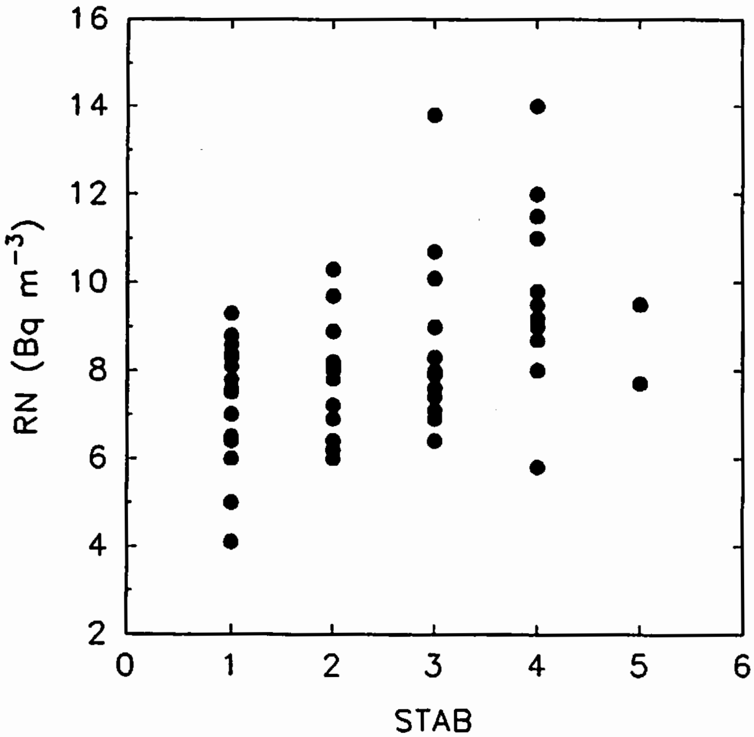

Figure 3. Radon concentration at 1 m versus Pasquill stability index for the Oak Ridge site.

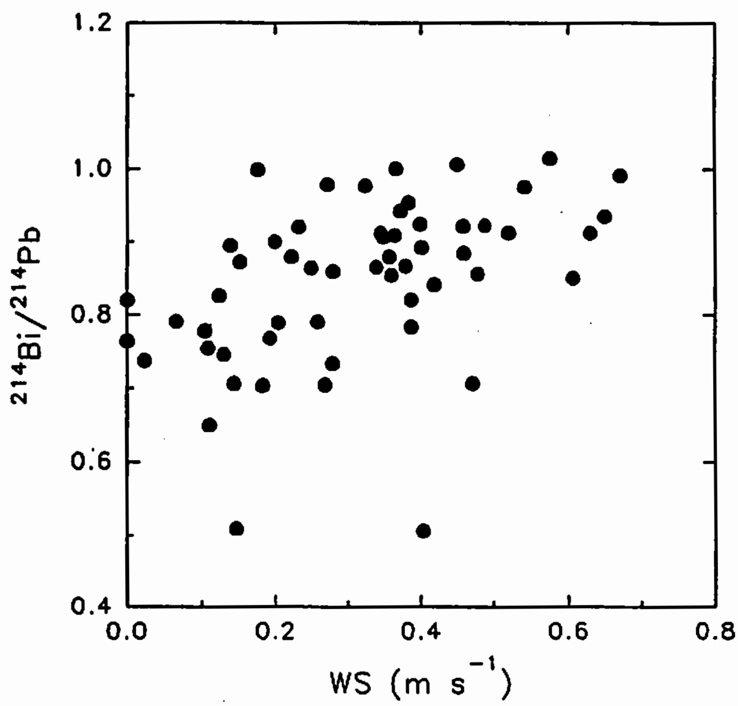

Figure 4. The activity ratio ^{214}Bi/^{214}Pb versus wind speed at the Oak Ridge site.