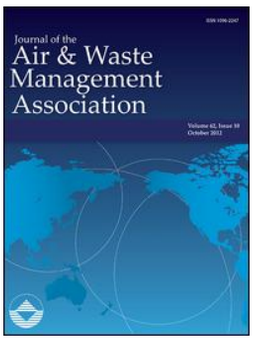

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Measurement of the gross alpha activity of the fine fractions of road dust and near-roadway ambient particle matter

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ABSTRACT

Traffic-related air pollution, including direct exhaust emissions and road dust (RD), impacts individuals living near busy roads. We recently conducted a study to investigate the sources and composition of tailpipe and non-tailpipe traffic emissions, where we collected and analyzed samples of ambient air fine particulate matter ($PM_{2.5}$) and fine RD ($RD_{2.5}$) at different distances from major roadways. We analyzed a subset of the samples, including those collected at the roadside and local background, for their alpha activity level. Subsequently, we investigated whether there is a distance-related decay in the alpha activity in $RD_{2.5}$ or $PM_{2.5}$ similar to those observed for traffic-related species in $PM_{2.5}$ and $RD_{2.5}$. We found that the alpha activity of ambient air $PM_{2.5}$ (Bq/mg) was more than an order of magnitude higher than the activity level of the corresponding $RD_{2.5}$ sample, suggesting that $PM_{2.5}$ may be more toxic than $RD_{2.5}$. Using mixed effects regression models, we found that ambient $PM_{2.5}$ alpha activity was significantly higher during the cold months than during warm months, and that the background was higher than the roadside (though not significantly). In contrast, the $RD_{2.5}$ alpha activity was significantly higher at the background site compared to the roadside but was not significantly affected by season. In addition to sampling position, both Zn and elemental carbon (EC) were significant predictors of $RD_{2.5}$ alpha activity. In addition, the roadside $RD_{2.5}$ activity levels were found to be higher at highways as compared to secondary roads. While traffic-related emissions do not appear to be significant sources of either

ambient $PM_{2.5}$ or $RD_{2.5}$ alpha activity, the $RD_{2.5}$ results suggest that traffic-related particles may contribute to $RD_{2.5}$ alpha-activity.

Keywords: Air Pollution; alpha activity; Road Dust; particle concentrator; particle radioactivity

INTRODUCTION

Traffic-related emissions, which impact individuals living near and commuting on busy roads, are a significant source of air pollution (Zhang and Batterman 2013; Hankey et al. 2017; Huang et al. 2018; Hamra et al. 2015; Puett et al. 2014; Kim et al. 2005). Exposure to traffic-related ambient fine particle matter ($PM_{2.5}$) or residential proximity to road have been associated with a wide range of adverse health effects, including respiratory and cardiovascular symptom exacerbation and birth outcomes (Boothe and Shendell, 2008; Carlsten et al., 2011; Gan et al., 2011; Gauderman et al., 2005, 2007; Hoffman et al., 2012; Jerrett et al., 2008; McConnell et al., 2006, 2010; Van Hee et al., 2010; Wilhelm et al., 2012; Wilhelm and Ritz, 2003; Williams et al., 2009; Zanobetti et al., 2010; Zeka et al., 2006).

Traffic-related air pollution is a complex mixture, which includes particulate matter (PM) originating from tailpipe emissions of vehicles and both direct and indirect non-tailpipe emissions. Tailpipe emissions contain elemental and organic carbon and various trace elements associated with incomplete combustion of fuel and oil additives. Non-tailpipe emissions include particles from wear of brakes, tires, engines and road surfaces, and resuspended road dust. Road dust itself commonly contains high concentrations of trace contaminants, which originate from tailpipe and non-tailpipe emissions, as well as crustal material (soil and sand), road salt, vegetation debris, etc. (Apeagyei et al. 2011; Gunawardana et al., 2012; Hwang et al., 2018). For example, Zn, Cu, Pb, Ni, Cr and Cd primarily originate from non-tailpipe emissions; Si, Ca and Al primarily originate from soil; Na and Cl mainly originate from ocean and road salt (Gunawardana et al., 2012; Ho et al., 2003; Johansson et al., 2009).

There are currently not sufficient available measurements of ambient particle alpha activity available to allow epidemiological studies of the health effects of environmentally relevant exposure levels in populations. However, it is known that alpha activity causes DNA damage at the cellular level (Chauhan et al., 2012). In addition, exposure to alpha emitters at low dosage levels causes significantly more chromosome damage in liver cells than either beta or gamma radiation (Brooks, 1975). A study by Little et al. (1975) supports the hypothesis that alpha activity exposure from ^{210}Po associated with cigarette

smoking may be a significant contributing factor in development of human lung cancer. Furthermore, recent studies have shown associations between a variety of human health effects and Particle Radioactivity (PR) measured as gross beta- and gamma-activities. These include association with total and cardiovascular mortality (Blomberg et al., 2018), effects on blood pressure (Nyhan et al., 2019), oxidative stress (Li et al., 2018), and lung and cardiac function (Vieira et al., 2019). In the absence of anthropogenic release, PR is attributed to predominantly to radionuclides of terrestrial origin, specifically radon progeny (Ceballos et al. 2016). Radon is a noble gas, a decay product of ^{238}U and ^{232}Th, which are both naturally occurring radioactive elements found in earth's crust. After formation, radon diffuses from the soil into ambient air, where it decays to radioactive progeny. The progeny quickly form charged clusters that attach to airborne accumulation mode particulate matter. Other natural sources of particle radioactivity include cosmogenic radionuclides such as ^{7}Be, and terrestrial radionuclides such as ^{40}K, from resuspension of soil (Blomberg et al. 2020).

In other publications from our current study, which was designed to investigate the sources and composition of tailpipe and non-tailpipe traffic emissions, we report that trace elements associated with traffic emissions show distance-related decreases in concentration for both ambient particles and road dust (Huang et al. 2020; Moreira et al. 2020; Silva et al. 2020). In this paper, we measure the gross alpha radioactivity of ambient fine particles and road dust at different distances from the road, and investigate whether there are decay patterns for PM gross alpha activity that are similar to those of the traffic-related elements we have reported previously (Huang et al., 2020; Silva et al., 2020). To the best of our knowledge, this is the first study to investigate the alpha activity of near roadway ambient PM or RD.

METHODS

1. Study design

We conducted a field study from June 2018 to December 2019 in the Greater Boston metropolitan area to examine the relationships between road proximity and traffic-related composition of the coarse and fine fractions of particle matter in ambient air (PM) and road dust (RD), at different distances from major roadways. On each sampling day, RD and ambient PM samples were collected at three distances from the road, one at the roadside (0-25 m), one at an intermediate distance (50-200 m) and one at a background site (500-1000 m) away from the roadside. Major roadways sampled included multi-lane divided state and interstate highways (with [A1] or without [A2] limited access via onramps and exit

ramps) and busy state secondary and connecting roads [A3]. Background and intermediate samples were collected the same day at locations on adjacent roadways within the target distances from the roadside site and were almost entirely on residential roads. For this paper, we measured the gross alpha radioactivity in 52 samples of fine ambient PM ($PM_{2.5}$) and the fine fraction of RD ($RD_{2.5}$) collected at paired roadside and background sites for 20 major roads in the greater Boston area Samples were collected twice at six of the sites, during cold and warm seasons. Sampling locations are shown in the Supplementary Material, Figure S1; road parameters and site locations are also included in the Supplementary Material, Table S1.

2. Sample Collection

For this study, we designed a Mobile Particle Concentrator Platform (MCMP). The MPCP was equipped with fine (0.2 to 2.5 μm) and coarse (2.5 to 10 μm) particle concentrators and a Road Dust Aerosolizer sampler (RDA), which was designed and constructed to simultaneously re-suspend road dust and separate it into fine and coarse fractions. Methods are described in detail elsewhere in this special issue (Martins et al. 2020) and summarized below.

a. Ambient $PM_{2.5}$

We used a modified Harvard Ambient Fine Particle Concentrator (HAFPC), originally a 5,500 LPM three-stage fine particle concentrator that has been described in detail in previous studies (Lawrence et al. 2004; Sioutas et al. 1997). The HAFPC was modified to require less power to be accommodated by the mobile platform. The modified system used two parallel two-stage concentrators whose outputs were combined, followed by separate collection of samples on Teflon and Quartz fiber filters, with flows of 45 LPM for each. For this study, we analyzed the samples collected on Teflon. Samples were collected for one hour at a total intake flowrate of 2,200 LPM; ambient concentrations were calculated using the concentrator enrichment factor as described elsewhere (Martins et al. 2020).

b. Road dust fine fraction ($RD_{2.5}$)

We developed a novel Road Dust Aerosolizer (RDA) sampler to re-suspend and collect $PM_{2.5}$ from the road surface ($RD_{2.5}$). The RDA sampler aerosolizes road dust at a very high flow rate from the surface of the roadway and simulates road PM resuspension into the air, which results in a more realistic measurement of PM composition and size, as described in our methods paper published in this special issue (Martins et al., 2020). RD was sampled for approximately 5 minutes, during which 12 m2 of road

surface was vacuumed. The $RD_{2.5}$ fraction was collected using a 30LPM cascade sampler (Demokritou et al. 2004). Like the HAFPC above, the RDA sampler collected samples on both Teflon and Quartz fiber filters, and the Teflon filters were analyzed for this study.

3. Sample radioactivity analysis

The analysis of PM gross alpha activity is described in detail elsewhere (Kang et al. 2020; Liu et al., 2020) and summarized here. We measured PM alpha activity in 52 samples of ambient $PM_{2.5}$ and $RD_{2.5}$ collected at paired roadside and background sites for 20 major roads in the greater Boston area using a low background gas proportional counter (LB4200, Canberra Industries, Inc., Meriden, CT) with a counting time of 600 min per sample. The instrument was calibrated using a 0.0518 μCi NIST traceable ^{210}Po source, and measurements were blank corrected. The background level was below 0.1 cpm and the minimum detectable activity (MDA) level for alpha averaged 0.071 Bq/mg. Most of the measured alpha activities in samples were above the detection limit.

The total activity on a filter at any time after collection is a function of radionuclide type and concentration as well as filter loading. After 1.5-2 years of storage, the total alpha activity is likely dominated by ^{210}Po decay, which is the decay progeny of ^{210}Pb (a beta-emitter) whose half-life is the longest of the radon decay radionuclides (22.3 years). Therefore, long-lived alpha activities in the air at the time of sampling can be estimated using the following equation modified based on Sheets and Thompson's equation [1]:

$$\mathrm{C} = \frac{A_t \cdot e^{\lambda t}}{M} \qquad [1]$$

Where A_t is the alpha activity on the filter at time of counting (Becquerel, Bq), λ is the decay constant for ^{210}Pb (8.51×10^{-5} day^{-1}), t is the time duration from the end of PM sampling to the start of alpha counting (day), and M is the mass loading of the sample collected on the filter (mg).

Although we measured both alpha- and beta-activity in these samples, the beta-activity levels were low, below the MDA (average of 0.170 Bg/mg) in 70% of $PM_{2.5}$ and 93% of the $RD_{2.5}$ samples. Background correction of measurements using blank filters generates some negative counts. For this reason, in this paper we focus primarily on the alpha activity of the samples. Gross beta activity measurements for these samples are included in the Supplementary Materials, Figure S2.

4. Other Data

Other data used in this paper include the concentrations of trace elements as well as elemental and organic carbon and their thermally evolved fractions, measured during this study and reported elsewhere in this special issue (Huang et al., 2020; Moreira et al., 2020; Silva et al., 2020). We analyzed our collected ambient $PM_{2.5}$ and $RD_{2.5}$ samples for trace elemental composition using X-Ray Fluorescence (Silva et al., 2020 and Huang et al., 2020, respectively). We analyzed elemental carbon (EC) and organic carbon (OC) using thermal optical reflectance (TOR) (Moreira et al., 2020, in this special issue). We used the IMPROVE TOR method (Chow et al., 2004), which distinguishes EC/OC as thermally evolved fractions, including 3 EC fractions (EC1, EC2 and EC3), 4 OC fractions (OC1, OC2, OC3 and OC4), a pyrolyzed carbon fraction (OP), and an optical EC fraction (Kang et al., 2010). The different characteristics of the carbonaceous species composing these thermally evolved fractions may offer insight into their origins (Habre et al. 2014; Kim et al. 2004; Kim and Hopke 2005; Kim et al. 2011; Lim et al. 2012; Sahu et al., 2011; Yu et al. 2002).

5. Statistical analysis

All statistical analyses were performed using SAS (version 9.4, SAS Institute, Cary, NC). Differences between background and roadside samples were tested using t-tests and paired t-tests, where appropriate. Because our study featured repeated measurements (i.e., multiple distances from each major road and measurements at the same sites during different seasons), we used a linear mixed-effect regression model to estimate the relationships between the alpha activity and study variables (e.g., season, proximity to roadway, road type), including PM composition. Models for $PM_{2.5}$ and $RD_{2.5}$ were run separately, with a random intercept for each major road. Slope coefficients for each parameter and species included were treated as fixed effects. Parameters investigated included the dichotomous variables reflecting distance from road (roadside versus local background), season (warm versus cold) and road type (A3 versus combined A1 and A2). Species investigated included the trace elements and carbon fractions measured for each sample. We considered results significant when $p < 0.05$ for the t-tests and mixed effects model.

RESULTS AND DISCUSSION

Results of alpha activity measurements are shown in Figures 1 and 2 for ambient $PM_{2.5}$ and $RD_{2.5}$, respectively. The alpha-activity of $PM_{2.5}$ and $RD_{2.5}$ is reported in Becquerels per milligram of mass (Bq/mg). The alpha activity of $PM_{2.5}$ is also shown in Bq/m^3 (Figure 1b). The one-hour alpha activity concentrations measured in this study are consistent with those reported in a recent study in greater Boston, where the 24-hour average during 2005-2006 was 0.0019±0.0011 Bq/m^3 (range, ND to 0.0076 Bq/m^3) (Liu et al., 2020).

Overall, ambient $PM_{2.5}$ alpha activity per unit mass was more than an order of magnitude higher than $RD_{2.5}$ as shown in Table 1. The ratio of $PM_{2.5}$ to $RD_{2.5}$ alpha activity was higher for roadside samples compared to the background, but this difference was not statistically significant (paired sample t-test, p-value 0.20).

Both near road and background ambient $PM_{2.5}$ alpha activity levels were significantly higher during the cold season (October-January) than during the warm season (June-September), averaging 0.236±0.112 and 0.172±0.052 Bq/mg, respectively (t-test, p-value 0.013). $PM_{2.5}$ alpha activity levels were also lower at the roadside sites than background sites during the cold season (0.183±0.068 and 0.226±0.109 Bq/mg, respectively; p-value 0.025), but similar during the warm season. There were no significant differences in $PM_{2.5}$ alpha activity levels observed in roadside samples for highways (combined A1 and A2 roads) compared to secondary (A3) roads. This suggests that traffic-related emissions to not contribute significantly to the $PM_{2.5}$ alpha activity.

$RD_{2.5}$ alpha activity levels were higher at the background than at the roadside, 0.012±0.006 and 0.009±0.004 Bq/mg, respectively. During the cold season, this difference was even more pronounced, with levels of 0.014±0.006 and 0.008±0.004 Bq/mg for background and roadside, respectively. During the warm season, the background (0.011±0.006 Bq/mg) and roadside (0.009±0.003 Bq/mg) $RD_{2.5}$ alpha activities were more similar. These differences were statistically significant both overall (paired sample t-test, p-value 0.015) and during the cold season (paired sample t-test, p-value 0.005). This suggests that traffic-related emissions are not a significant source of $RD_{2.5}$ alpha activity. Although it did not reach statistical significance, the $RD_{2.5}$ alpha-activity in roadside samples was observed to be higher in the warm season than the cold season, 0.010±0.003 and 0.008±0.004 Bq/mg, respectively.

$RD_{2.5}$ roadside alpha-activity levels were significantly higher for highways (combined A1 and A2 roads, 0.010±0.003 Bq/mg) than for busy state secondary and connecting roads (A3, 0.007±0.004 Bq/mg)

where traffic density and speed are generally lower (Table S1, supplementary material); t-test p-value 0.022. At background sites, alpha activity levels were similar for highways and secondary roads.

The results of the mixed effects regression analysis for $PM_{2.5}$ and $RD_{2.5}$ are presented in Table 2. The only significant predictor of the $PM_{2.5}$ alpha activity levels was season, with higher activities observed during the cold months. Position was borderline significant as a predictor, with lower activities at the roadside compared to the background. None of the trace elemental species or carbon fractions were significant predictors for the ambient $PM_{2.5}$ alpha-activity. This is in contrast with previous findings from our group, which show strong associations of alpha activity with both $PM_{2.5}$ and Sulfur (S), a known regional and secondary $PM_{2.5}$ component (Kang et al., 2020; Liu et al., 2020). However, these previous studies used large datasets. Our results agree with a previous study from our group, which found that particle radioactivity in Boston was highest in February (Blomberg 2020). This suggests that the alpha activity measured in our study is similarly due to long-lived radon decay products.

The $RD_{2.5}$ alpha activity is significantly predicted by Zn, EC1 and position, with activity levels higher at the background than roadside sites. The Zn and EC1 both have positive slope estimates, indicating that higher concentrations of Zn and EC1 are both associated with higher alpha activity. In our previous papers on the concentrations of trace elements and carbon fractions in $RD_{2.5}$ published in this special issue, both Zn and EC1 showed a decay with distance from the road (Huang et al. 2020, Moreira et al. 2020). These species are both related to traffic. EC1 is associated with diesel combustion in vehicles and engines (Kim et al. 2004; Kim and Hopke 2005) and Zn is associated with tire wear, brake wear, and engine oil (Garg et al 2000; Hjortenkrans et al 2007; Lough et al 2005). The positive associations observed between traffic-related species and alpha activity in $RD_{2.5}$, together with roadside activity levels that are higher at highways than beside secondary roads, may suggest that traffic-related emissions may contribute to $RD_{2.5}$ activity, though not a significant source. However, as the concentrations of road dust gross alpha activity are significantly higher in $PM_{2.5}$ than in $RD_{2.5}$, deposition of long-lived radon progeny attached to ambient $PM_{2.5}$ is likely to be a contributor.

Our field study was not designed to investigate the alpha activity of $RD_{2.5}$ or ambient near-road $PM_{2.5}$, but rather to investigate direct and indirect traffic-related PM emissions, as reported in several papers in this special issue (Huang et al. 2020, Martins et al. 2020, Moreira et al. 2020, Silva et al. 2020). However, for this paper, we measured gross alpha activity in a subset of samples collected from the original study. Our current study, to the best of our knowledge, is the first to investigate the alpha activity of either road dust or near-roadway ambient $PM_{2.5}$. With the relatively small sample size, we were unable to

include many variables in our analysis. However, we found that the alpha activity of ambient $PM_{2.5}$ is more than an order of magnitude higher than that of $RD_{2.5}$ (Table 1), suggesting that $PM_{2.5}$ may be more toxic than $RD_{2.5}$. In addition, we observed that both EC1 and Zn, traffic-related species, are both significant positive predictors of alpha activity in $RD_{2.5}$, which may suggest that there could be some traffic-related contribution.

CONCLUSIONS

In the first study investigating the alpha activity of near roadway traffic-related PM and RD, we found that ambient $PM_{2.5}$ alpha activity levels in the greater Boston area were more than twenty times higher than their corresponding samples of $RD_{2.5}$. This suggests that ambient $PM_{2.5}$ may be considerably more toxic than road or soil dust.

Using linear mixed effects regression models, we found that $PM_{2.5}$ alpha-activity is influenced significantly by season, with lower levels during the warm season, consistent with observations from previous studies. Distance from roadway is a borderline significant predictor of $PM_{2.5}$ alpha activity, with higher levels at background locations than roadside. Our results indicate that traffic-related sources do not directly contribute significantly to $PM_{2.5}$ alpha activity. In contrast to previous studies, neither S nor $PM_{2.5}$ concentrations were associated with $PM_{2.5}$ alpha activity; however, this may be due to the relatively small sample size we analysed.

Alpha-activity of $RD_{2.5}$ was also higher at the background locations than the roadside, but in contrast to $PM_{2.5}$, was not significantly affected by the season. We found that the roadside alpha activity of $RD_{2.5}$ was higher at highways than secondary roads. Significant predictors for $RD_{2.5}$ alpha activity using mixed effects models included sample location (roadside or background), Zn, and EC1. Together, these results may suggest that although traffic-related emissions are not a significant source pf $RD_{2.5}$ alpha activity, they may contribute and thus may impact the hazards of $RD_{2.5}$ exposure.

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Numerous studies have reported the effects of traffic-related particulate matter (PM) on human health, and there is growing interest in the health effects of exposure to environmental exposures to PM alpha activity. This is the first study to report on the alpha activity of road dust (RD) or near-roadway ambient PM. We found that the alpha activity of ambient PM is an order or magnitude than that of RD, suggesting that ambient PM may be more toxic. In both PM and RD, the alpha activities were higher at background sites than at the roadside, indicating that traffic-related emissions are not a significant source of PM or RD radioactivity. The alpha activity of PM was related only to season, with higher levels during colder months. In contrast, RD alpha activity was not influenced by the season, however it was associated traffic-related species (EC and Zn), which may suggest some traffic-related contribution.

SUPPLEMENTARY MATERIAL

Measurement of the gross alpha activity of the fine fractions of road dust and near-roadway ambient particle matter

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SUPPLEMENTARY MATERIAL

Figure S1. Sampling locations within the greater Boston metropolitan area

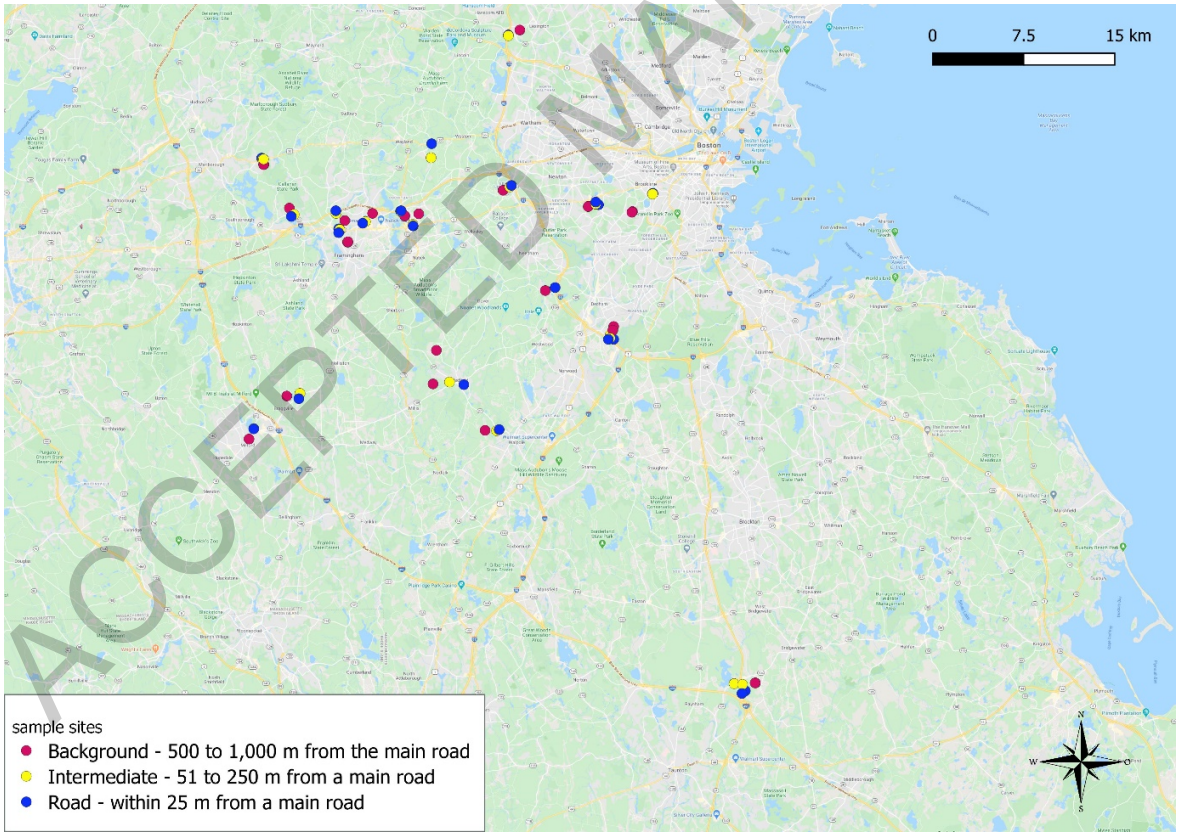

Table S1 - Sampling location details including road parameters and meteorological conditions

Date	Location	Type	Daily Count*	Speed Limit	# Lanes	Distance (m) to Road			Temp (°F)	%RH	WS	WD
						Road	Intermed.	Backgr.				
6/7/2018	9E Framingham	A2	87,209	50	2+2	6	107	1010	69.6	59.6	2.31	SW
6/8/2018	Marlborough 20	A3	18,304	40	2+2	12	110	550	76.7	49.6	0.64	WSW
6/18/2018	I-95 Lexington	A1	182,647	55	4+4	10	65	800	85.1	58.8	6.36	SSE
6/22/2018	Hammond pond	A3	29,208	40	2+2	2	81	680	41.2	45.6	1.15	--
6/28/2018	I-95 Dedham	A1	168,126	55	4+4	10	87	1010	77.7	32.8	3.81	SE
7/9/2018	I-90 Natick	A1	114,872	65	3+3	25	190	576	84.6	46	1.25	NW
7/13/2018	I-95 Newton	A1	174,037	55	4+4	15	180	854	82.0	48.6	3.47	SW
7/20/2018	Wayland 20	A3	22,352	35	1+1	7	68	320	81.9	44.4	1.07	ENE
7/30/2018	Rt 30 Framingham	A2	19,326	40	2+2	12	44	430	80.1	52	1.69	WSW
8/7/2018	I-95 Needham	A1	161,796	55	4+4	10	35	580	90.1	59.6	1.61	SW
8/21/2018	I-495 Bridgewater	A1	71,301	65	4+4	10	80	1250	72.0	74	2.71	E
8/24/2018	9W Natick	A2	55,001	50	2+2	11	130	875	80.0	52.4	1.1	WSW
8/29/2018	9E Framingham	A2	87,209	50	2+2	6	107	1010	93.2	54.9	2.58	NW
9/17/2018	Framingham 30	A3	16,986	40	1+1	2	150	720	77.9	70.8	0.13	W
9/24/2018	Rt 1 Saugus	A2	103,054	50	3+3	8	70	460	57.4	58.8	4.15	NE
10/10/2018	Jamaicaway	A3	38,599	30	2+2	16	51	1020	82.5	62.8	1.73	NE
10/15/2018	Walpole 27	A3	15,086	40	1+1	13	200	870	54.8	88.9	0	
10/17/2018	Medfield 109	A3	14,562	30	1+1	2	150	670	59.6	49.4	2.27	SE
10/19/2018	Marlborough 20	A3	18,304	40	2+2	12	110	550	55.6	46.3	2.38	WSW
10/24/2018	9E Framingham	A2	87,209	50	2+2	6	107	1010	45.0	81.1	4.85	NNW
10/26/2018	Hammond pond	A3	29,208	40	2+2	2	81	680	--	--	--	--
11/7/2018	I-90 Natick	A1	114,872	65	3+3	25	190	576	60.6	45.1	7.92	NNE
11/29/2018	Holliston 16	A3	17,396	40	1+1	8	195	576	42.9	58.5	1.85	NW
12/4/2018	I-90 Framingham	A1	105,510	65	3+3	23	145	950	37.9	44.5	2.9	NE
12/6/2018	Milford 85	A3	8,374	40	1+1	16	88	510	36.3	51.8	3.71	WNW
12/18/2018	I-495 Bridgewater	A1	71,301	65	4+4	10	80	1250	31.3	45	7.21	NNW
1/16/2019	I-95 Dedham	A1	168,126	55	4+4	10	87	1010	36.1	57.9	4.91	W

*Annual average daily vehicle count (in 2018 or 2019) from MA Department of Transportation Road Inventory (https://gis.massdot.state.ma.us/roadinventory/)

Figure S2a. Gross beta activity (Bq/mg) of ambient $PM_{2.5}$ at roadside and background sites. Average MDA shown as solid black line.

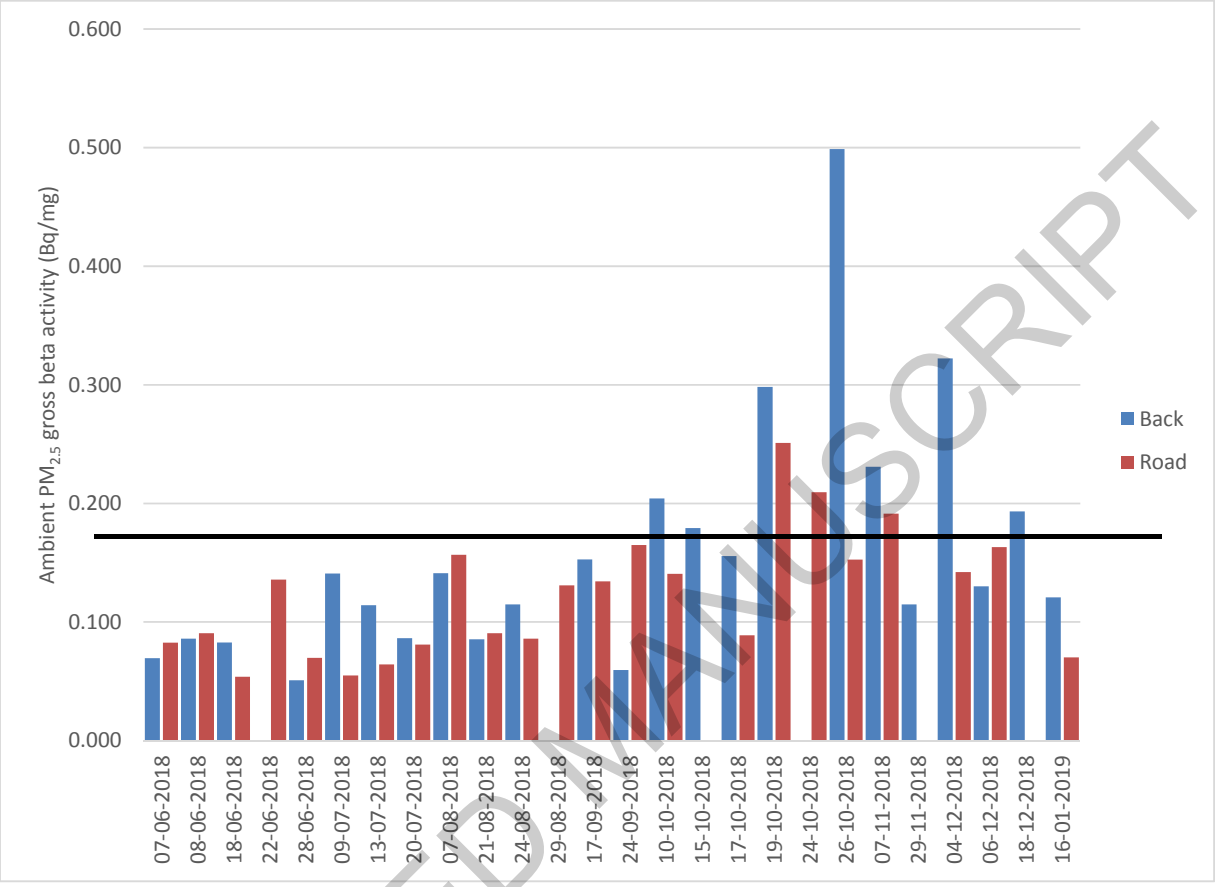

Figure S2b. Gross beta activity (Bq/m^3) of ambient $PM_{2.5}$ at roadside and background sites. Average MDA shown as solid black line.

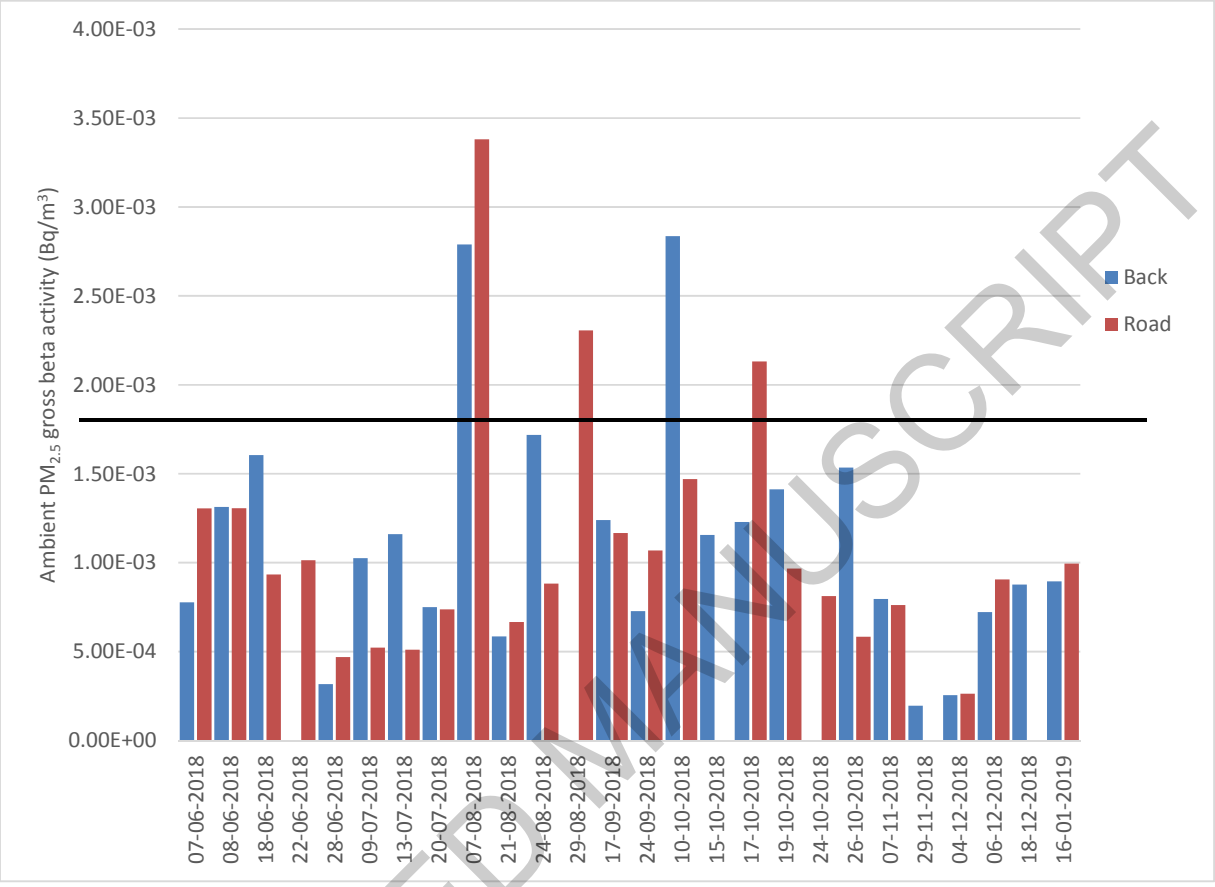

Figure S2c. Gross beta activity (Bq/mg) of fine road dust ($RD_{2.5}$) at roadside and background sites. Average MDA shown as solid black line.

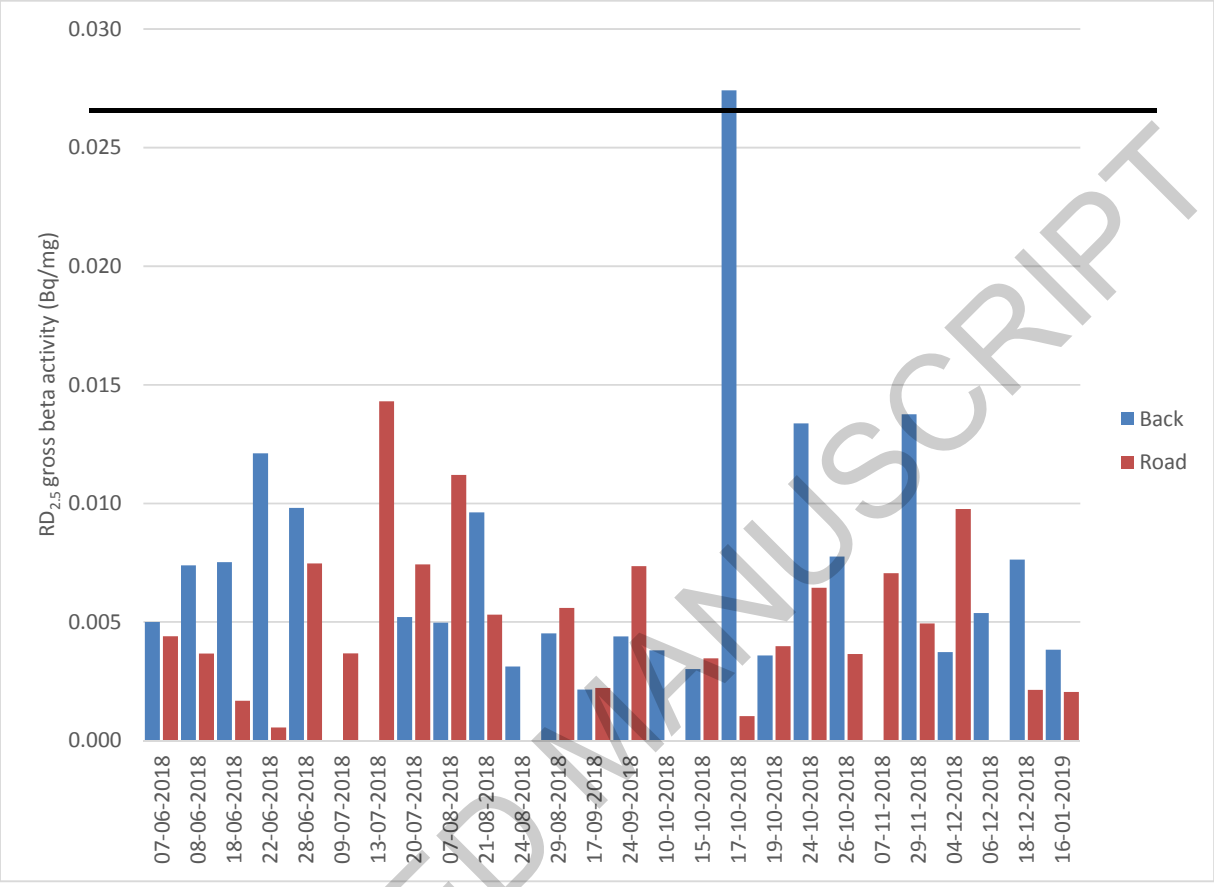

Table 1. Ratio of ambient $PM_{2.5}$ to $RD_{2.5}$ alpha -activity (Bq/mg).

Position	Air/Dust Alpha Activity Ratio		
	Average (SD)*	Minimum	Maximum
Roadside	23.5 (11.0)	1.7	40.8
Background	19.7 (8.7)	9.0	51.7
Overall	21.6 (10.0)	1.7	51.7

*Ratios are all statistically significant; paired sample t-test for $PM_{2.5}$ and $RD_{2.5}$ roadside, background and overall yield p-values $<<0.001$.

Table 2. Fixed effect estimates for ambient $PM_{2.5}$ and road dust $RD_{2.5}$ alpha activity models.

Model	Fixed Effect	Slope	SE	p-value
Ambient $PM_{2.5}$	Season Cold (Warm=0)*	0.07434	0.02344	0.0037
	Position Background (Roadside=0)	0.04469	0.02269	0.0589
Road Dust ($RD_{2.5}$)	Zinc*	0.00338	0.00100	0.0020
	EC1*	0.00019	0.00005	0.0004
	Position Background (Roadside=0)*	0.00431	0.00136	0.0040

*Statistically significant predictors of alpha activity level in Bq/mg at $p<0.05$.

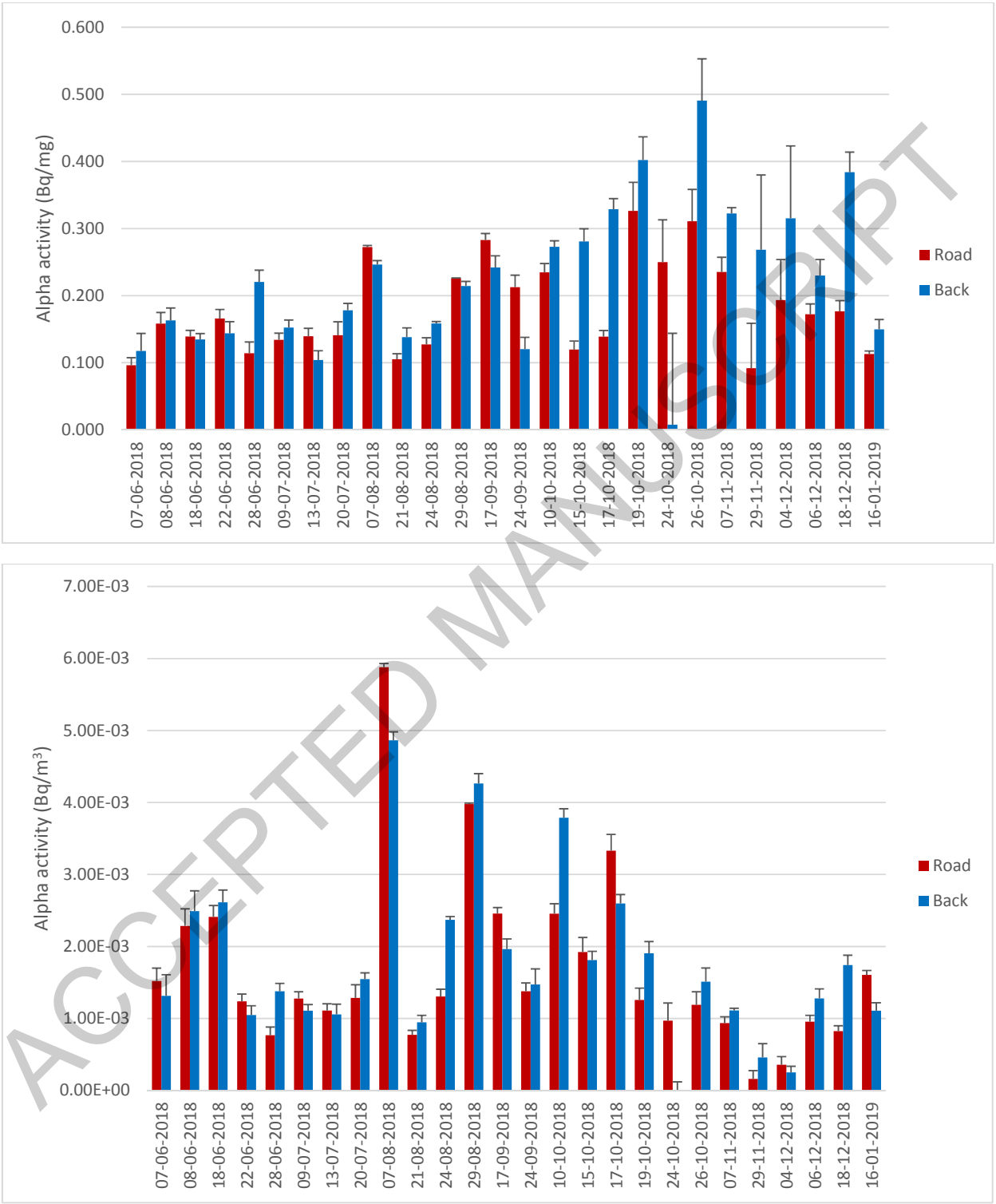

Figure 3. Ambient $PM_{2.5}$ alpha activity (Bq/mg PM and Bq/m^3) and uncertainty measured in samples collected at the roadside and local background positions at major roadways in the greater Boston area.

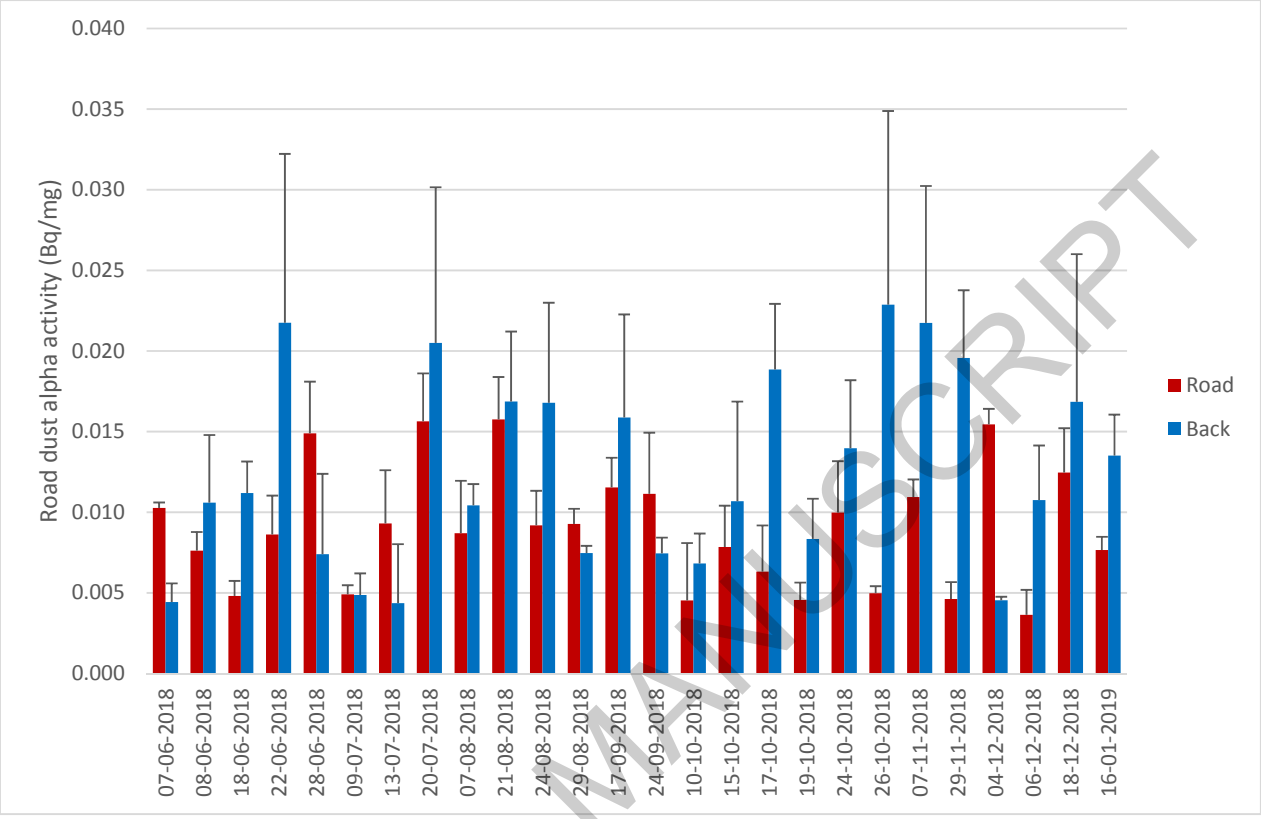

Figure 4. Road dust ($RD_{2.5}$) alpha activity (Bq/mg RD) and uncertainty measured in samples collected at the roadside and local background positions at major roadways in the greater Boston area.