Addendum to the final report for "Contribution of Particle Emissions from a Cement-Related Facility to Outdoor Dust in Surrounding Community"

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Comments of Dr. Gary Norris – USEPA-NERL

1. List CMB profiles that were used in a table.

Table S1. Potential source profiles constructed for CMB model running

Potential Sources used in CMB	Description	From
RCM	Raw cement material	This study
ROCK	Global-averaged reference rock	Mason (Rahm, 1976)
SOIL	Global-averaged reference soil	Bowen (Rahm, 1976)
MARIN	Marine aerosol	PACS (Watson, 1979)
CDUST	Continental dust	PACS (Watson, 1979)
UDUST	Urban dust	PACS (Watson, 1979)
AUTPB	Leaded auto exhaust	PACS (Watson, 1979)
RDOIL	Residual oil combustion	PACS (Watson, 1979)
VBRN1	Vegetative Burning Profile 1	PACS (Watson, 1979)
VBRN2	Vegetative Burning Profile 2	PACS (Watson, 1979)
KRAFT	Kraft Paper Mill	PACS (Watson, 1979)
SULFT	Sulfite Paper Mill	PACS (Watson, 1979)
HOGFU	Hogged Fuel Boiler	PACS (Watson, 1979)
ALPRO	Aluminum Production	PACS (Watson, 1979)
STEEL	Steel Blast Furnace	PACS (Watson, 1979)
PERMN	Ferromanganese Furnace	PACS (Watson, 1979)
CARBO	Carborundum Furnace	PACS (Watson, 1979)
GLASS	Glass Furnace	PACS (Watson, 1979)
CARBF	Carborundum Furnace	PACS (Watson, 1979)

2. Discuss inorganic analytical methodology used by PACs and how it compared to the method used in this study. Also, report SRM recovery results.
The Portland Aerosol Characterization Study employed X-ray fluorescence for elemental analysis, thermal/optical reflectance for EC/OC, and ion chromatography for sulfates/nitrates. This study employed IC/ICPMS to quantify the elemental concentrations in collected samples. Due to the use of a microwave oven-assisted digestion method with concentrated high purity HNO_3 (EPA method TO-3051), only HNO_3 soluble Si was detected in this study. Therefore, the concentration of Si obtained in this study did not represent the total Si concentration in the field samples. Due to the discrepancy in quantifying Si concentrations by the two analytical methods, Si was not used in the CMB estimation in our study, and thus, the estimate of contribution from the facility will not be affected by the measurements of Si.

Accuracy was measured against elemental standards (High Purity Standards, Inc., Charleston, SC) certified to 0.5%. Acceptable quality assurance checks were deemed to be 100±20 % of the certified values.

3. Discuss the applicability of PACS profiles (coarse PM) to the size fraction evaluated in this study.
The PACS reported elemental concentrations in fine particles (<2.5 μm) and coarse particle (<30 μm). The particle size distribution collected in this study showed most particles were below 10 μm in diameter and the largest particle size was around 30 μm determined by microscopical analyses. Therefore, the profile of coarse particles from PACS was appropriate for use in CMB estimation.

4. Discuss the MPIN matrix in more detail and provide the species for each source that were most influential in the CMB analysis.

Response: The MPIN (modified pseudo inverse normalized) matrix identifies which fitting species have the largest influences on the source contribution estimates from each profile. MPIN is normalized to values of -1 to 1. Species with absolute values of 0.5 to 1.0 are considered influential species. Noninfluential species have MPIN absolute values of 0.3 or less. Species with absolute values between 0.3 and 0.5 are ambiguous but generally be considered noninfluential.

Table S2. The most influential species of each source for the CMB estimation

Source	Species	Number of Samples	MPIN value
CEMENT	Ca	28	0.92 ~ 1.00
MARIN	Mg	28	1.00
RDOIL	Fe	28	0.91 ~ 1.00
FERMN	Mn	28	1.00
SOIL	Al	25	1.00
UDUST	Ti	9	1.00
ROCK	Al	1	1.00
AUTPB	Pb	1	1.00
ALPRO	Al	1	1.00

Comments of Dr. Rachelle Duvall – USEPA-NERL

1. *Are the methods used in this study appropriate to answer the question: To what extent does the St. Lawrence Cement facility contribute to the dust deposited in the Waterfront South area of Camden?*

The methods are appropriate for the study. Ambient and surface dust samples were collected in the area neighboring the cement facility. A sample from the cement pile was also collected to obtain a representative source profile for the cement facility dust. Source apportionment modeling was conducted using the EPA CMB 8.2 model and particle classification was determined with SEM.

2. *Do you agree with the overall conclusion of the report that the St.Lawrence Cement facility contributes about 10% of the dust deposited in the Waterfront South community? If not, can you identify a different conclusion from the data presented in the report?*

I do not agree with the conclusion. The model results indicated that the plant contributes on average 5-22% of outdoor dust. It seems like the value "10%" was arbitrarily selected. More justification is needed as to how the final value of 10% was deduced.

The estimated RCM contribution to the outdoor dust at the control site in Chester City Park was 8.9%. If we, however, consider this as an urban background level of cement-related contribution to total outdoor dust in Camden area, the background-subtracted contribution from the facility to the residential areas surrounding the cement facility will be ~12.9%. Additionally, the particle size distribution showed that the deposition samplers collected mostly particles sized below 10 μm (Table 5 in the report), thus, the total particle mass determined by deposition sampling approach may be lower than the true dust mass deposited in the neighborhood. Therefore, the contribution based on the ratio of the TSP concentrations predicted by the ISCST3 dispersion model and the total PM mass collected by the deposition sampler could be overestimated. The contribution from the facility to the neighborhood dust pollution was also estimated based on the PM_{10} concentration. Assuming that approximately 40% of the predicted TSP concentrations is PM_{10} (Mugica et al., 2002), the contributions from the facility to the neighborhood dust pollution ranged from 7.3 to 13.5%, lower than the previously estimated values. Based on these analyses, we concluded that the contribution from the cement facility to the neighborhood outdoor dust was approximately 10%.

Table S3. The estimated RCM contributions to total outdoor dust in WFS, Camden, NJ by three different approaches

Range	Ca Regression Estimation	CMB Estimation		ISCST3 Estimation	
		No Background Subtraction	Background-subtracted	Based on TSP	Based on PM_{10}[b]
Minimum	2.4%	5.6%	0.0%	18.3%	7.3%
Maximum	8.1%	21.8%	12.9%	33.7% (4.0%)	13.5%

[a]The contributions by the CMB model were subtracted from the contribution of a background site (i.e., 8.9%)
[b]The contributions were obtained through the estimated PM_{10} concentration (assuming PM_{10} mass concentration constitutes 40% of TSP mass concentration)

3. Additional Comments/suggestions/questions:
- Section 2.2: Was there a reason for selecting the summer/fall time frame for collecting dust samples?
No.

Are the wind patterns the same throughout the year?
From the nearby airport meteorological data, the main wind direction is from west (ranging from southwest to northwest) and consistent throughout the year. Please see Figure S1 below for general wind patterns in the study area.

- Section 2.4: Did you consider collected a road dust sample near the cement facility?
No

- Section 2.6: How many and which species were used in the CMB model for the study samples?
See Table S2 in the previous response.

- Section 3.3: Tables 5, 6, and 7 should be labeled as Tables 6, 7, and 8, respectively.
Yes, the reviewer is correct.

- Section 3.5: Consider adding an unknown slice in the CMB contribution pie charts (Figure 8a and 8b). Since the Ferromanganese furnace contributions are so small, consider including this with the residual oil contribution slice (and indicating that ferromanganese furnace contribution is less than <1%). If you would like to keep the ferromanganese contribution separate change the contributions to "< 0.5%" on the pie charts (rather than 0 or 0.2%) and also change the color since it looks the same as the color for soil.
We provide extended results in Tables S4 and S5 for EF and Ca/Fe ratios and Table S6 for CMB estimation. But, please note that the results were conducted for the data collected from the second deposition sampling.

What species were markers for each source?
Please, see Table S2.

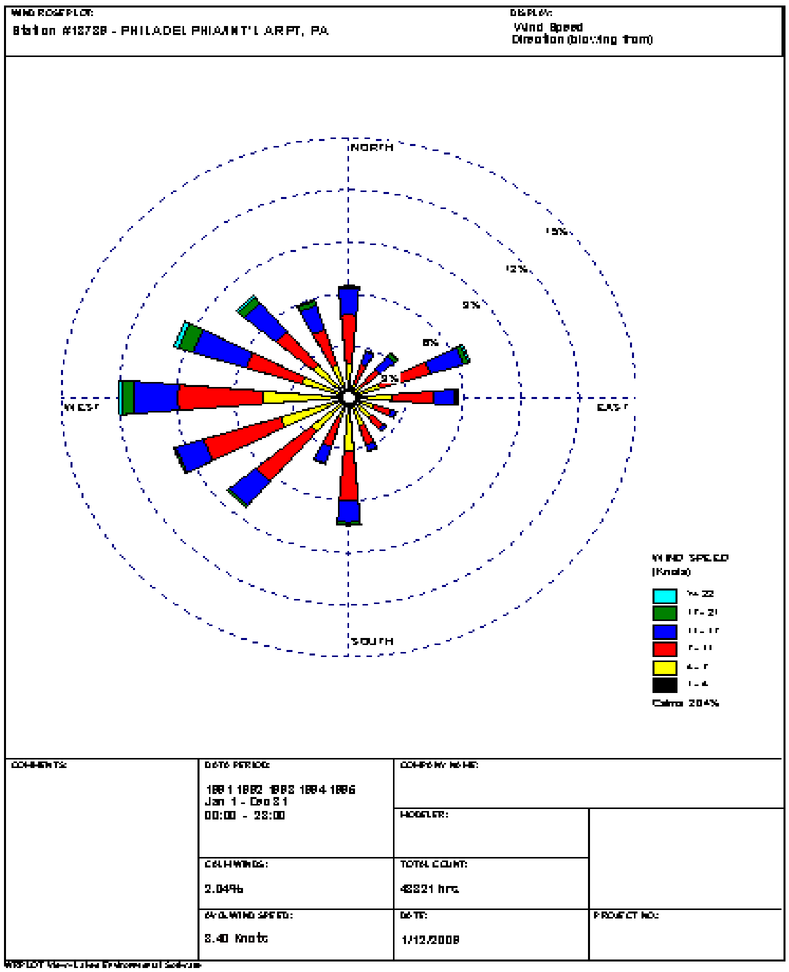

Figure S1. A wind rose plot acquired from the nearby Philadelphia International Airport for the duration of 5 years (source: NJDEP)

Table S4. Enrichment factors and Ca/Fe ratios for the deposited dust samples.

L.	Distance (km)	Al	Ca	Cd	Cr	Cu	Fe	Mg	Mn	Pb	V	Zn	Ca/Fe
RCM	0.00	1.02	36.40	NA	1.47	4.63	0.68	3.06	4.04	3.31	0.34	9.25	38.71
1	0.19	0.71	8.19	47.39	3.26	12.67	2.04	2.60	3.21	101.38	1.65	137.31	2.92
2	0.29	0.68	7.22	69.48	7.41	17.05	2.64	2.75	3.33	214.23	2.13	184.98	2.00
3	0.27	0.75	9.02	NA	4.28	21.32	2.67	3.02	3.51	138.00	2.19	192.12	2.45
4	0.35	0.78	9.85	99.91	5.54	33.84	4.10	3.77	3.70	252.18	2.58	227.60	1.74
5	0.38	0.83	11.02	106.57	4.44	28.72	4.22	4.12	3.81	197.96	2.76	256.14	1.90
6	0.61	0.96	8.25	228.88	7.43	39.43	3.97	3.52	4.76	316.29	3.08	338.99	1.51
7	0.55	1.37	11.16	493.30	62.85	40.42	9.09	6.01	7.57	374.68	4.29	732.80	0.89
8	0.45	1.12	12.19	235.71	16.62	41.44	5.17	5.24	5.55	334.52	4.14	475.97	1.71
9[a]	2.38	0.94	8.86	182.17	6.74	35.51	3.78	5.11	4.17	176.08	6.51	464.77	1.70
10[a]	2.20	0.75	7.18	136.64	7.63	23.59	3.07	4.44	2.99	111.13	4.97	377.30	1.70

[a]Locations are at background site, Gloucester City Park

Table S5. Enrichment factors and Ca/Fe ratios for the surface dust samples.

L.	Distance (km)	Al	Ca	Cd	Cr	Cu	Fe	Mg	Mn	Pb	V	Zn	Ca/Fe
RCM	0.00	1.02	36.40	NA	4.63	0.68	4.43	3.06	4.04	3.31	0.34	9.25	38.71
1	0.27	7.97	4.67	131.90	60.10	7.54	11.89	1.46	4.03	512.46	5.00	1394.79	0.58
2	0.27	1.66	3.70	474.01	22.72	4.18	6.47	1.67	2.89	153.17	2.36	137.63	0.10
3	0.27	0.95	4.43	29.68	13.63	5.85	115.49	6.27	2.57	167.88	2.00	141.42	0.52
4	0.35	0.43	1.78	484.36	11.09	3.57	4.55	3.02	1.73	142.08	2.04	43.46	0.38
5	0.38	1.26	9.26	53.44	24.41	6.74	5.74	1.80	2.57	67.76	2.11	80.43	1.00
6	0.61	1.02	21.16	88.14	19.03	8.11	7.24	4.11	3.93	202.68	9.43	337.89	1.89
7	0.55	1.44	4.22	50007.50	62.02	29.48	13.81	1.90	4.34	1338.11	2.97	2240.47	0.10
8	0.45	0.93	6.32	67.43	25.51	11.27	12.02	2.25	4.04	221.48	3.95	73.13	0.41
9	2.38	1.54	4.09	1714.75	30.56	8.89	7.34	2.18	2.86	2144.32	4.60	170.17	0.31
10	2.38	1.21	4.21	178.48	21.45	6.22	9.71	2.23	2.95	256.26	2.78	107.96	0.49
11	2.38	1.30	7.71	103.86	45.91	9.40	134.10	3.92	3.57	440.09	3.46	193.33	0.60
12	2.38	1.31	5.38	115.62	32.92	8.79	8.03	2.96	3.63	260.18	3.64	282.79	0.44
13	2.38	0.71	4.87	17.96	11.56	4.47	2.08	2.19	1.25	252.22	2.04	220.37	0.79
14[a]	2.38	0.69	2.26	26.27	14.74	2.88	3.82	1.37	1.51	113.62	3.85	126.18	0.57
15[a]	2.20	1.72	0.09	33.56	37.64	7.00	6.09	1.98	2.67	173.06	5.54	56.66	0.32

[a]Locations are at background site, Gloucester City Park

Table S6. The percent estimation of RCM contribution to outdoor dusts by using an EPA approved CMB model with elemental concentrations for RCM and other potential dust sources

L.	Sample	Distance (km)	Source									R^2	χ^2	%Mass
			RCM	ROCK	SOIL	MARIN	UDUST	AUTPB	RDOIL	ALPRO	FERMN			
1	D001-B	0.19	20.5%	E[a]	9.4%	20.1%	4.0%	E	45.9%	E	0.2%	0.98	2.00	95.7
	D001-C		21.8%	E	9.4%	21.4%	3.9%	E	43.3%	E	0.2%	0.97	2.33	92.1
2	D003-C	0.29	15.1%	E	8.3%	20.5%	2.5%	E	53.5%	E	0.1%	0.97	1.93	86.7
	D003-D		12.7%	E	7.5%	20.4%	1.1%	0.0%	58.2%	E	0.1%	0.97	2.49	88.8
3	D006-C	0.27	17.0%	E	8.2%	19.9%	0.7%	E	54.1%	E	0.1%	0.98	1.45	95.5
4	D007-D	0.35	12.4%	E	6.0%	22.0%	E	E	59.5%	E	0.0%	0.96	1.42	130.1
5	D008-B	0.38	14.0%	E	6.5%	23.0%	E	E	56.5%	E	0.0%	0.96	1.88	95.0
6	D011-B	0.61	9.0%	E	9.0%	20.2%	E	E	61.7%	E	0.1%	0.98	0.86	108.5
7	D012-D	0.55	5.6%	E	7.6%	24.5%	E	E	62.2%	E	0.1%	0.90	4.07	142.7
8	D014-C	0.45	10.9%	E	7.4%	24.5%	E	E	57.1%	E	0.1%	0.96	1.87	121.1
9[b]	D016-C	2.38	8.9%	E	7.7%	27.3%	E	E	56.0%	E	0.1%	0.96	1.70	122.5
10[b]	D020-C	2.20	8.9%	E	7.3%	27.6%	E	E	56.3%	E	0.0%	0.95	1.88	117.4

[a]E denotes an eliminated source from CMB model calculation; thus, the contribution percentage was not available.

[b]Locations are at background site, Gloucester City Park.

Note: Six potential dust sources were obtained through Portland Aerosol Characterization Study and each source is abbreviated as follows: MARIN (marine aerosol), UDUST (urban dust), AUTPB (leaded auto exhaust), RDOIL (residual oil combustion), ALPRO (aluminum production), and FERMN (ferromanganese furnace).

Comments of Dr. Patrick T. O'Shaughnessy, PhD, CIH, Associate Professor, The University of Iowa Department of Occupational and Environmental Health
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1. Are the methods used in this study appropriate to answer the question: To what extent does the St. Lawrence Cement facility contribute to the dust deposited in the Waterfront South area of Camden?

I believe that the methods used in this study were appropriate to answer the question stated above. From reading the report I observed that the primary methods included:

- the use of “deposition” samplers to obtain approximately a month-worth of settled dust in the area of concern,
- surface-wipe sampling of horizontal surfaces in the area, and
- the use of a Chemical Mass Balance source-receptor model to determine the percent of all dust deposited in the area that can be attributed to the cement facility.

The primary result that 10% of the deposited dust originates from the facility is based on the CMB model results.

As indicated by the authors of the report, results obtained from the deposition samplers were the most accurate in terms of answering the question because the surface wipe samples contained dust from local sources such as paint flakes and metal oxides from rusting surfaces. Although not stated, certainly those samples would also be contaminated by organically derived dust sources such as tree and plant pollen. Therefore, the method of obtaining surface samples was appropriate but the authors were correct in not basing their overall conclusion on results obtained from the surface samples.

The methods used to analyze the deposition samplers were appropriate. The samplers themselves are a novel instrument, but have been previously tested by the authors. However the placement of those samplers was less than ideal. The best-case scenario would be to place them in areas with an unhindered view of the cement facility to negate local turbulence effects (building wake) that may either under- or over-represent the actual amount of dust originating from the facility. The authors indicated the need to make the samplers as inconspicuous as possible, which was reasonable given some losses (apparently via vandalism). However, the local “micro-climate” within which they were placed certainly added to the variability of their results.

We agree with the comments with the reviewer. However, due to many practical limitations for the placement of deposition samplers in the field, there were no ideal sampling locations as described by the reviewers.

The CMB modeling method is one that has been used extensively by the USEPA and, therefore, was an appropriate method to apply for this situation.

The question above asked whether the methods were “appropriate” for which I can say they were. However, in my estimation the methods may not have been entirely “adequate” to answer the question, or at least to best indicate the confidence in the final assessment.

The problem of apportioning sources of a pollutant (dry dust in this case) is hugely complex and will necessarily involve some level of estimation. The most "hypothetical" aspect of the CMB analysis was the use of a data set from another study conducted in Portland, Oregon to apply additional potential sources of dust to the model. Certainly, it would be naïve to think that the only source of dust is the cement facility, however an attempt to determine the adequacy of the data obtained from the Portland study should have been conducted and described. This analysis would result in a range of possible "source profiles" which would then result in a range of possible percent-contributions by the cement facility. The only indication of an attempt to indicate the potential variability of these estimates was the author's decision to assume an uncertainty of 10% for each element. Apparently this uncertainty level resulted in the variability seen in the results presented in Table 11. However, a justification for such a narrow range of uncertainty could not be found.
We agree with the reviewer's comments. The ideal approach would be the use of the sources profiles generated in the study area for modeling. Unfortunately, they are not available. The Portland Aerosol Characterization Study (PACS, Watson 1979) included source profiles of comprehensive source types (e.g., geological, vegetative burning, industry, transportation…etc), which are very close to the source types in our study area. Thus, the PACS was used in our modeling. An uncertainty of ~10% for the mean of each element was used in the CMB estimation, as suggested by Vega et al. (2000).

In addition, we estimated the contribution by a simple Ca-regression analysis and ISCST3 modeling approach, as described earlier. The estimates from the other two approaches are close to the CMB estimates, suggesting that the estimates from the CMB modeling is reasonable.

Furthermore, I felt that some analysis of local meteorological events occurring during the two sampling episodes, and for the area in general, should have been performed. The underlying assumption of this analysis is that the cement dust will disperse homogenously in all radial directions from the facility, i.e. a concentration profile of deposited dust from the facility will have a peak at the facility and radiate out in all directions with the same rate of decay. An analysis of wind directions and speeds during the sampling period, and averaged over several years would help to convince the reader that this is a fair assumption by showing a relatively random pattern of wind directions and speeds via a "wind rose". Such an analysis could also help to demonstrate the overall potential for dust from the raw cement material (RCM) pile to become airborne and thus contribute to downwind dust deposition. Although a highly-variable phenomenon, studies on the resuspension of dust from roads, deserts, and agricultural fields could be used to estimate the potential for resuspension given the RCM particle size distribution and local wind velocities. For example, a basic assessment criteria is: if the wind velocities are never high enough to resuspend dust, and wind directions never are from facility to the community sampled, then it's impossible for the dust in that community to originate from the facility, and therefore 0% of the dust from the facility contributes to dust in the sampled area. An analysis of the potential for any condition other than that which would create this situation would have been helpful.

We agreed with the reviewer's comments. As shown in Figure 1, the study area is right by a river and has strong wind most time of the year (Figure S1). The median wind speed in the study area is 3.5 m/s and dominated by W/NW/SW wind. Thus, the wind speed and wind direction are sufficient to re-suspend RCM piled inside the facility and spread out the residential areas surrounding the facility. Please see Table S7 for the travelling distances of different particle sizes at the 25% and median wind speeds in the study area.

Table S7. The traveling distance for different size particles that may be emitted from the cement pile at the median wind speed (3.5 m/s)[a] in the WFS neighborhood (Hinds, 1999).

Particle diameter (μm)	Settling velocity (cm/sec)	Particle travelling distance (m) for a settling height of 4.5 m[b]		Particle travelling distance (m) for a settling height of 9.0 m[c]	
		25% wind speed[d]	Median wind speed[e]	25% wind speed[d]	Median wind speed[e]
10	0.3	4,500	5,297	9,000	10,595
20	1.2	1,125	1,324	2,250	2,649
38	4.3	312	367	623	734
75	16.9	80	94	160	188
100	24.8	54	64	109	128
150	47.2	29	34	57	67
250	94.3	14	17	29	34
1,000	386.0	3	4	7	8

[a]The median wind speed during June and September, 2006 was obtained from the Philadelphia International Airport Weather Station located 12 km west of the cement facility.

[b]Assuming each sized aerodynamic particle traveled from the middle height of the cement pile (4.5 m) to a deposition sampler 2.5 m above the ground level.

[c]Assuming each sized aerodynamic particle traveled from the top of the cement pile (9 m) to a deposition sampler which was placed 2.5 m above the ground level.

[d]The wind speed of 25 percentile (3.0 m/s) from the Philadelphia International Airport Weather Station during June, 2006 ~ September, 2006.

[e]The median wind speed (3.5 m/s) from the Philadelphia International Airport Weather Station during June, 2006 ~ September, 2006.

2. Do you agree with the overall conclusion of the report that the St. Lawrence Cement facility contributes about 10% of the dust deposited in the Waterfront South community? If not, can you identify a different conclusion from the data presented in the report?

The results from Table 11 demonstrate a range of RCM contribution to local dust loadings between 4.9% and 21.8%. Therefore, I agree that a general estimate of "about 10%" is possible from the results given.

As a caveat to that statement, it is unfortunate that the type of assessment performed does not allow for an indication of statistical confidence in that percentage, i.e. a p-value. It seems that this would have been possible if, for example, RCM dust had an easily-identifiable chemical "signature" that could distinguish those particles from all other particles analyzed via the SEM x-ray analysis procedure so that the percentage of RCM particles could be directly measured relative to all other particles. Given that calcium concentrations were discovered to show a relationship with downwind distance (Figure 6), it is interesting that an attempt of that sort was not made with calcium as the label for RCM dust given that the other sources would not typically consist of calcium (soil, marine aerosol, oil combustion, ferromanganese furnace). However, there must be more to this than suspected as I'm sure such an attempt would have been made if possible.

Lacking the data needed to obtain a confidence estimate, the next-best approach would be to obtain the result from a second, independent analysis. Admittedly, that may not have been possible. However, the use of the ISTS3 dispersion modeling technique was brought up (in the Results only, and, given their inclusion in the Conclusion statement, more information should have been provided in the body of the report) with estimated contributions ranging from 18.3% to 33.7%. These values were considered "reasonable" but were also considered very conservative whereas they appear to justify the upper level of the CMB results (~ 20%) rather than the CMB mid-range as suggested by the value of 10%.
Please find the appendix I for the estimation based on ISCST3 model results. Also, see the previous response for the final decision of 10% contribution for the facility.

In summary, the answer to the overall question of how much the cement plant contributes to local settled dust was determined with a combination of sampling (real values), modeling (estimates), and the expert judgment of the authors. Given the variability in the results, their final statement that the dust contribution is "probably on the order of 10%" reflects their judgment, which should be respected. However, given the combination of both CMB and dispersion modeling results, I would have erred toward the higher end, and ended the final sentence with the "outdoor dust is on average of <20%".

Additional information of the study

Prior to Camden field sampling, a series of field evaluation tests were conducted for hooded and un-hooded samplers that were placed side-by-side at various locations. The results of mass collected through the evaluation test are presented in Table S8. The difference between hooded and un-hooded sampler was not significant ($p=0.8054$; $N=22$). Therefore, the hooded design did not significantly affect the mass of dust deposition in the field, compared to the sampler without a severe weather protection cover. A picture of a deposition sampler placed in the field is also attached.

Table S8. The summarized results for the field evaluation of deposition samplers (hooded vs. un-hooded type) at various locations and sampling durations.

Round	Sampler Type	Collected Mass		Sampling Duration	Sampling Place
		Mean±SD	RPD[a]		
I	Hooded	0.580±0.074 mg	29.7%	10 days	House porch
	Un-hooded	0.782±0.028 mg			
II	Hooded	0.405±0.034 mg	2.0%	14 days	House porch
	Un-hooded	0.397±0.041 mg			
III	Hooded	0.661±0.012 mg	12.5%	15 days	Park shelter
	Un-hooded	0.583±0.028 mg			
IV	Hooded	0.396±0.033 mg	9.8%	21 days	House porch
	Un-hooded	0.359±0.041 mg			
V	Hooded	0.295±0.061 mg	14.2%	20 days	House porch
	Un-hooded	0.340±0.056 mg			
VI	Hooded	0.266±0.087 mg	10.7%	20 days	Bus stop
	Un-hooded	0.296±0.109 mg			

[a]Relative percent difference between means of two co-located samplers at the same location