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Weight Losses of Marble and Limestone Briquettes Exposed to Outdoor Environment in the Eastern United States

Results of Exposure 1988-1992

by K. J. Reimann

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EASTERN UNITED STATES
RESULTS OF EXPOSURE 1988-1992**

by

K. J. Reimann

Energy Technology Division
Instrumentation and NDE Section

MASTER

April 1995

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U.S. NATIONAL PARK SERVICE
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ABSTRACT

Monitoring continued on weight changes in marble and limestone briquettes exposed to the outdoor environment at sites in the eastern United States. This report presents data for the exposure period 1988-1992 and summarizes results for the entire period from 1984. Since 1989, only three exposure sites have remained active, but briquettes from preexposed material were added at those sites. A linear relationship was found between cumulative gravimetric losses and exposure period. These losses resulted in an average recession rate of 11 to 21 $\mu\text{m}/\text{yr}$ for marble and 21 to 45 $\mu\text{m}/\text{yr}$ for limestone. The recession rates are site-dependent and can be described with respect to rain depth and other atmospheric conditions, as evidenced by the very low rates at the Ohio site on the movable rack, dry regime. Weight monitoring is continuing in a planned 10-year program.

1 INTRODUCTION

Studies of the effects of acid precipitation and air pollution on the weathering of Shelburne marble and Salem limestone at test sites in the eastern United States are continuing under the sponsorship of the U.S. Department of Interior, National Park Service. Monitoring of weight changes of marble and limestone briquettes after exposure to the environment at the test sites is one of several tasks performed at Argonne National Laboratory (ANL).

The environmental exposure of stone briquettes began in May 1984 at test sites in Raleigh, NC; Chester, NJ; Newcomb, NY; Washington, DC; and in 1986 in Steubenville, OH¹. The exposure sites are instrumented to record a wide range of environmental parameters.

Sets of gravimetry briquettes are installed in racks at each site¹ for annual or multiyear exposure periods. Withdrawal, weighing, and reinstallation of gravimetric briquettes follows a strict protocol,¹ summarized in the description of experimental procedures in Refs. 2 and 3. Outdoor exposure of briquettes at the NJ site was discontinued in 1987, but the quinquennial briquettes were retrieved in 1989 and weighed. Similarly, exposure at the NC site was discontinued in 1989.

This report details the results obtained by weight measurements from 1988 to 1992. All data from 1984 to 1992 were used to generate the graphs in order to assess trends of long-time exposure. Weight changes of individual briquettes are listed in the Appendix.

2 PROCEDURES AND RESULTS

Three briquettes of each stone type exposed at each active site, together with the site simulation samples, were retrieved according to schedule, dried, weighed, and returned to the exposure site for reinstallation. Included in the measurement procedure² were three control samples that were kept at ANL. In 1989, three quinquennial briquettes of each

stone type were retrieved and measured, together with the annual samples from the respective exposure sites (except NJ and OH). At the NJ site, no annual samples were exposed after 1987. Quinquennial briquettes from OH were not due for retrieval in that year. In 1988, after installation of preexposed briquettes at NC, NY, DC, and OH, their retrieval and weight measurements were made to coincide with those of annual briquettes formed from fresh stone.

Some deviation in the retrieval schedule occurred in OH and DC, mainly due to changes in site personnel and weather conditions. Dates of installation and removal of gravimetric briquettes at their respective field exposure sites are shown in Table 1. Briquettes from discontinued sites (NJ, NC) were archived at ANL after weight measurements.

Quinquennial briquettes at NY and DC were exposed for a short period after the five-year exposure, to compare weight losses during that short interval with runoff measurements. These briquettes were subsequently reinstalled for additional exposure.

Table 1. Dates of gravimetric specimen installation and removal at field exposure sites.

STONE AND OPERATION	PERIOD ^a	NC	NJ	NY	DC	OH	
						OHS STATIC RACK	OHM MOVABLE RACK
Fresh Stone							
Install	5	8-02-88	-	8-12-88	10-22-88	10-17-88	10-17-88
Remove		5-23-89	-	6-23-89	6-23-89	9-01-90	9-01-90
Install	6	Disc. ^b	-	7-21-89	8-10-89	10-10-90	10-10-90
Remove		-	-	7-11-90	8-30-90	-	-
Install	7	-	-	9-26-90	12-12-91	-	-
Remove		-	-	6-29-92	9-02-92	-	-
Install		-	-	9-28-92	2-04-93	-	-
Install	Q1	5-25-84	6-5-84	6-19-84	8-11-84	-	-
Remove		5-23-89	6-26-89	6-23-89	6-23-89	-	-
Install	S1	Disc. ^b	Disc. ^b	7-21-89	8-10-89	-	-
Remove		-	-	9-15-89	12-01-89	-	-
Install		-	-	1-08-90	6-29-90	-	-
Preexposed Stone							
Install	1	6-09-88	-	6-10-88	6-08-88	6-01-88	6-01-88
Remove		5-23-89	-	6-23-89	6-23-89	9-01-90	9-01-90
Install	2	Disc. ^b	-	7-21-89	8-10-89	10-10-90	10-10-90
Remove		-	-	7-11-90	8-30-90	-	-
Install	3	-	-	9-26-90	12-12-91	-	-
Remove		-	-	6-29-92	9-02-92	-	-
Install		-	-	9-28-92	2-04-93	-	-

^aAnnual or multi-year exposure period designation.

^bDiscontinued

Averages and standard deviation of incremental weight changes by stone type, exposure site, and exposure period are summarized in Tables 2-5. Results from the quinquennial briquettes (5 year and runoff experiment exposure) are included in these tables. Incremental weight changes in the field control and in ANL control briquettes for the same period and site are included for comparison. These control briquettes are kept indoors in sealed plastic bags, except at the DC site, where they are not kept in bags. All briquettes, including simulations and ANL controls, were dried in an oven at 51°C for one week prior to weighing.

Table 2. Incremental weight changes and associated recession of fresh marble briquettes for the period 1988-1992

PERIOD AND SITE	EXPOSURE TIME (DAYS)	EXPOSED BRIQUETTES		AVG. LOSS (g)	
		AVG. LOSS and STD. DEVIATION (g)	RECESSION PRORATED ($\mu\text{m}/\text{yr}$)	FIELD CONTROL	LAB CONTROL
Period 5					
1988-1989					
NC	294	0.2154 $\pm$ 0.0107	15.7 $\pm$ 0.8	0.0026	0.0026
NY	315	0.1989 $\pm$ 0.0030	13.5 $\pm$ 0.2	0.0064	0.0043
DC	244	0.2284 $\pm$ 0.0030	20.1 $\pm$ 0.3	-0.0050	-0.0069
OHS ^a	449 ^b	0.4383 $\pm$ 0.0292	20.9 $\pm$ 1.4	0.0000	-0.0011
OHMD ^a	449 ^b	0.0766 $\pm$ -	3.7 -	0.0000	-0.0011
OHMS ^a	449 ^b	0.2397 $\pm$ -	11.5 -	0.0000	-0.0011
Period 6					
1989-1990					
NY	355	0.2574 $\pm$ 0.0011	15.6 $\pm$ 0.1	0.0009	0.0046
DC	385	0.2985 $\pm$ 0.0244	16.6 $\pm$ 1.4	0.0010	0.0031
Period 7					
1990 1992					
NY	642	0.3644 $\pm$ 0.0110	12.2 $\pm$ 0.4	-0.0016	-0.0018
DC	283	0.1961 $\pm$ 0.0102	14.9 $\pm$ 0.8	0.0032	0.0036
Period Q1 (Quin-					
quennial)					
1984-1989					
NC	1824	1.2580 $\pm$ 0.0581	14.8 $\pm$ 0.7	0.0026	0.0026
NJ	1847	1.9065 $\pm$ 0.0407	22.1 $\pm$ 0.5	0.0019	0.0041
NY	1830	1.1743 $\pm$ 0.0311	13.8 $\pm$ 0.4	0.0064	0.0043
DC	1777	1.4468 $\pm$ 0.0216	17.5 $\pm$ 0.3	-0.0050	-0.0069
Period Sh 1 (Quin-					
quennial) 1989					
NY ^c	56	0.0496 $\pm$ 0.0012	19.0 $\pm$ 0.5	0.0019	-0.0041
DC ^c	113	0.0956 $\pm$ 0.0041	18.2 $\pm$ 0.8	0.0042	0.0061

^aOHS = Ohio stationary rack; OHMD = Ohio movable rack, dry regime; OHMS = OHIO movable rack, sprayed regime.

^bRetrieved in 1990.

^cQuinquennial gravimetry briquettes were used for the runoff experiment.

Table 3. Incremental weight changes and associated recession of fresh limestone briquettes for the period 1988-1992

PERIOD AND SITE	EXPOSURE TIME (DAYS)	EXPOSED BRIQUETTES		AVG. LOSS (g)	
		AVG. LOSS and STD. DEVIATION (g)	RECESSION PRORATED ($\mu\text{m}/\text{yr}$)	FIELD CONTROL	LAB CONTROL
Period 5					
1988-1989					
NC	294	0.3072 $\pm$ 0.0209	26.3 $\pm$ 1.8	-0.0153	-0.0152
NY	315	0.2659 $\pm$ 0.0105	21.3 $\pm$ 0.8	0.0015	0.0008
DC	244	0.5404 $\pm$ 0.0056	55.8 $\pm$ 0.6	-0.0584	-0.0566
OHS ^a	449 ^b	1.1466 $\pm$ 0.0404	64.3 $\pm$ 2.3	-0.0738	-0.0574
OHMD ^a	449 ^b	0.1526 $\pm$ -	8.6 -	-0.0738	-0.0574
OHMS ^a	449 ^b	0.4320 $\pm$ -	24.2 -	-0.0738	-0.0574
Period 6					
1989-1990					
NY	355	0.3722 $\pm$ 0.0265	26.4 $\pm$ 1.9	0.0304	0.0192
DC	385	0.6694 $\pm$ 0.0259	43.8 $\pm$ 1.7	0.0855	0.0429
Period 7					
1990-1992					
NY	642	0.5609 $\pm$ 0.0207	22.0 $\pm$ 0.8	0.0180	0.0205
DC	283	0.3022 $\pm$ 0.0102	26.9 $\pm$ 0.9	-0.0200	-0.0264
Period Q1					
(Quin- quennial)					
1984-1989					
NC	1824	2.9619 $\pm$ 0.0070	40.9 $\pm$ 0.1	-0.0153	-0.0152
NJ	1847	4.3734 $\pm$ 0.0774	59.6 $\pm$ 1.1	0.0778	0.0816
NY	1830	2.9099 $\pm$ 0.0522	40.1 $\pm$ 0.7	0.0015	0.0008
DC	1777	4.0306 $\pm$ 0.0441	57.1 $\pm$ 0.6	-0.0584	-0.0566
Period Sh					
1 (Quin- quennial)					
1989					
NY ^c	56	0.0964 $\pm$ 0.0010	43.4 $\pm$ 0.4	0.0360	0.0251
DC ^c	113	0.3108 $\pm$ -	69.3 -	0.1017	0.1051

^aOHS = Ohio stationary rack; OHMD = Ohio movable rack, dry regime; OHMS = OHIO movable rack, sprayed regime.

^bRetrieved in 1990.

^cQuinquennial gravimetry briquettes were used for the runoff experiment.

Table 4. Incremental weight changes and associated recession of preexposed marble briquettes for the period 1988-1992

PERIOD AND SITE	EXPOSURE TIME (DAYS)	EXPOSED BRIQUETTES		AVG. LOSS (g)	
		AVG. LOSS and STD. DEVIATION (g)	RECESSION PRORATED (µm/yr)	FIELD CONTROL	LAB CONTROL
Period 5					
1988-1989					
NC	348	0.5133 ± 0.0008	31.7 ± 0.05	0.0026	0.0026
NY	378	0.5167 ± 0.0139	29.3 ± 0.8	0.0064	0.0043
DC	380	0.6349 ± 0.0345	35.8 ± 1.9	-0.0050	-0.0069
OHS	587	1.1004 ± 0.0504	40.2 ± 1.8	0.0003	-0.0011
OHMS	587	0.3902 ± -	14.3 -	0.0003	-0.0011
Period 6					
1989-1990					
NY	355	0.4084 ± 0.0032	24.7 ± 0.2	0.0009	0.0046
DC	385	0.4413 ± 0.0308	24.6 ± 1.7	0.0010	0.0031
Period 7					
1990-1992					
NY	642	0.5476 ± 0.0220	18.3 ± 0.7	-0.0016	-0.0018
DC	283	0.2271 ± 0.0412	17.2 ± 3.1	0.0032	0.0036

Table 5. Incremental weight changes and associated recession of preexposed limestone briquettes for the period 1988-1992

PERIOD AND SITE	EXPOSURE TIME (DAYS)	EXPOSED BRIQUETTES		AVG. LOSS (g)	
		AVG. LOSS and STD. DEVIATION (g)	RECESSION PRORATED (µm/yr)	FIELD CONTROL	LAB CONTROL
Period 5					
1988-1989					
NC	348	2.1909 ± 0.1143	158.6 ± 8.3	-0.0153	-0.0152
NY	378	2.6921 ± -	179.4 -	0.0015	0.0008
DC	380	2.4662 ± 0.3381	163.5 ± 22.4	-0.0584	-0.0566
OHS	587	3.2761 ± 0.0726	140.6 ± 3.1	-0.0738	-0.0574
OHMS	587	0.6965 ± -	29.9 -	-0.0738	-0.0574
Period 6					
1989-1990					
NY	355	1.5531 ± 0.6373	110.2 ± 45.2	0.0304	0.0192
DC	385	1.1650 ± 0.7343	76.2 ± 48.0	0.0449	0.0429
Period 7					
1990-1992					
NY	642	0.7413 ± 0.1053	29.2 ± 4.1	0.0180	0.0205
DC	283	0.4488 ± 0.0206	39.9 ± 1.8	-0.0200	-0.0264

The standard deviation was calculated by

$$\sigma = \left[\frac{\sum_{i=1}^N (x_i - \bar{x})^2}{N} \right]^{1/2},$$

where x = weight change of briquette,

$\bar{x}$ = average weight change,

N = number of briquettes (generally, $N = 3$), and

σ = standard deviation.

N was chosen as the weighting factor for the standard deviation because it results in a maximum likelihood estimation that is generally used to describe populations. $(N-1)$ is an unbiased estimator, customarily used for sampled data.

The quoted prorated recession for the exposed briquettes was calculated as follows:

$$\text{Prorated recession} = \frac{\text{average gravimetric weight loss (g)}}{(\text{exposure area}) \times (\text{stone density})} \times \frac{365}{(\text{days of exposure})}$$

The exposed area of the skyward surface of each briquette is $\approx 63 \text{ cm}^2$, and the densities used in the calculations were 2.7 g/cm^3 for marble and 2.3 g/cm^3 for limestone.

Cumulative weight changes over the period 1988–1992 are summarized in Tables 6–10. These average changes and standard deviations were calculated by summing the changes for each briquette over the various time periods, averaging the results for each site, and calculating the standard deviation as described earlier. Weight changes of exposed briquettes are tabulated with respect to cumulative exposure time, whereas changes in field and ANL control briquettes are tabulated by cumulative elapsed time between subsequent weight measurements.

Because the weight measurements of preexposed stones were performed simultaneously with measurements on fresh stone briquettes, the same site and ANL control briquettes were used. Similarly, the same control briquettes were used for the OH exposure site stationary and movable racks.

Preexposed stone briquettes were reexposed at the different sites in two ways, one with the preexposed surface facing skyward (marked "up" in the Appendix), the other with the preexposed surface facing groundward (marked "down" in the Appendix). Weight changes of both exposure-type briquettes were combined to calculate the averages in Tables 4, 5, 8, and 9.

Table 6. Cumulative weight changes (g) of fresh marble briquettes, 1988-1992

SITE	EXPOSED BRIQUETTES		FIELD CONTROLS	
	CUM. EXPOSURE TIME (DAYS)	AVG. LOSS AND STD. DEV.	CUM. ELAPSED TIME (DAYS)	AVG. LOSS AND STD. DEV.
NC	1407	1.0055 ± .0538	1818	.1172 ± .0135
	1824 ^a	1.2581 ± .0581	1818	.1172 ± .0135
NJ	1847 ^a	1.9065 ± .0407	1869	.0009 ± .0007
NY	1385	.9708 ± .0224	1842	.0068 ± .0023
	1740	1.1982 ± .0217	2245	.0061 ± .0002
	1830 ^a	1.1746 ± .0311	1842	.0068 ± .0023
	1886 ^b	1.2242 ± .0302	1934	.0073 ± .0006
	2382	1.5625 ± .0314	2946	.0035 ± .0001
DC	1468	1.1974 ± .0062	1855	.0056 ± .0021
	1777 ^a	1.4468 ± .0216	1855	.0056 ± .0021
	1853	1.4793 ± .0146	2442	.0066 ± .0023
	1890 ^a	1.5424 ± .0227	2007	.0097 ± .0023
	2136	1.6690 ± .0202	3016	.0098 ± .0025
OHS ^b	1205	1.1958 ± .0785	1473	.0138 ± .0000
OHMD ^b	859	.1921 ± -	1473	.0138 ± .0000
OHMS ^b	859	.4871 ± -	1473	.0138 ± .0000

^aQuinquennial briquettes.

^bOHS = Ohio stationary rack; OHMD = Ohio movable rack, dry regime;
OHMS = Ohio movable rack, sprayed regime.

Table 7. Cumulative weight changes (g) of fresh limestone briquettes, 1988-1992

SITE	EXPOSED BRIQUETTES		FIELD CONTROLS	
	CUM. EXPOSURE TIME (DAYS)	AVG. LOSS AND STD. DEV.	CUM. ELAPSED TIME (DAYS)	AVG. LOSS AND STD. DEV.
NC	1407	2.7441 ± .2184	1818	.1172 ± .0135
	1824 ^a	2.9619 ± .0070	1818	.1172 ± .0135
NJ	1847 ^a	4.3734 ± .0774	1869	.1410 ± .0073
NY	1385	2.5469 ± .1315	1842	.1225 ± -
	1740	2.9191 ± .1437	2245	.1436 ± -
	1830 ^a	2.9099 ± .0522	1842	.1225 ± -
	1886 ^b	3.0063 ± .0517	1934	.1496 ± -
	2382	3.4765 ± .1965	2946	.1700 ± -
DC	1468	3.4333 ± .0771	1855	.1970 ± .0124
	1777 ^a	4.0306 ± .0441	1855	.1970 ± .0124
	1853	4.0843 ± .0783	2442	.2395 ± .0138
	1890 ^a	4.2973 ± .0000	2007	.2987 ± .0121
	2136	4.3909 ± .0827	3016	.2187 ± .0139
OHS ^b	1205	3.4061 ± .1200	1473	.0138 ± .0000
OHMD ^b	859	.9052 ± -	1473	.0138 ± .0000
OHMS ^b	859	1.4129 ± -	1473	.0138 ± .0000

^aQuinquennial briquettes.

^bOHS = Ohio stationary rack; OHMD = Ohio movable rack, dry regime;
OHMS = Ohio movable rack, sprayed regime.

Table 8. Cumulative weight changes (g) of preexposed marble briquettes, 1988-1992

SITE	EXPOSED BRIQUETTES		FIELD CONTROLS	
	CUM. EXPOSURE TIME (DAYS)	AVG. LOSS AND STD. DEV.	CUM. ELAPSED TIME (DAYS)	AVG. LOSS AND STD. DEV.
NC	348	.5133 ± .0008	1818	.1172 ± .0135
NY	378	.5167 ± .0139	1842	.0068 ± .0023
	733	.9252 ± .0107	2245	.0061 ± .0002
	1375	1.4728 ± .0327	2946	.0035 ± .0001
DC	380	.6349 ± .0346	1855	.0056 ± .0021
	765	1.0773 ± .0640	2442	.0066 ± .0023
	1048	1.3044 ± .0228	3016	.0098 ± .0025
OHS ^a	587	1.1004 ± .0504	1473	.0053 ± .0008
OHMS ^a	587	.3902 ± -	1473	.0053 ± .0008

^aOHS = Ohio stationary rack; OHMD = Ohio movable rack, dry regime;
OHMS = Ohio movable rack, sprayed regime.

Table 9. Cumulative weight changes (g) of preexposed limestone briquettes, 1988-1992

SITE	EXPOSED BRIQUETTES		FIELD CONTROLS	
	CUM. EXPOSURE TIME (DAYS)	AVG. LOSS AND STD. DEV.	CUM. ELAPSED TIME (DAYS)	AVG. LOSS AND STD. DEV.
NC	348	2.1909 ± .1143	1818	.1172 ± .0135
NY	378	2.6921 -	1842	.1225 -
	733	3.6079 -	2245	.1436 -
	1375	4.2439 -	2946	.1700 -
DC	380	2.4662 ± .3381	1855	.1970 ± .0124
	765	3.6311 ± .3962	2442	.2395 ± .0138
	1048	4.0799 ± .4168	3016	.2187 ± .0139
OHS ^a	587	3.2761 ± .0726	1473	.0138 ± .0000
OHMS ^a	587	.6965 ± -	1473	.0138 ± .0000

^aOHS = Ohio stationary rack; OHMD = Ohio movable rack, dry regime;
OHMS = Ohio movable rack, sprayed regime.

Table 10. Cumulative weight changes (g) of fresh marble briquettes, 1988-1992

CUMULATIVE ELAPSED TIME (DAYS)	AVG. AND STANDARD DEVIATION	
	MARBLE	LIMESTONE
1818	.0066 ± .0014	.1409 ± .0095
1842	.0113 ± .0012	.1476 ± .0102
1855	.0033 ± .0027	.1854 ± .0090
1869	.0072 ± .0007	.1857 ± .0092
1934	.0072 ± .0012	.1727 ± .0090
2007	.0094 ± .0013	.2905 ± .0107
2226	.0071 ± .0009	.1474 ± .0098
2245	.0064 ± .0011	.2283 ± .0097
2442	.0067 ± .0009	.1668 ± .0095
2946	.0049 ± .0009	.1873 ± .0105
3016	.0100 ± .0010	.2019 ± .0096

3 DISCUSSION AND CONCLUSIONS

The trend in weight loss discussed in Ref. 3 continued during the exposure period 1988-1992. These were (a) limestone weight losses for any given site and period were greater than marble weight losses; and (b) weight losses for both stone types were site dependent. For completeness, and to assess the effect of long-term exposure on gravimetric behavior, the results presented here incorporate data from the beginning of the exposure period (1984) until 1992. Measurements of quinquennial briquettes are included.

The average cumulative weight loss values and their variances are plotted in Figs. 1a and b for fresh marble and limestone, respectively, as functions of cumulative exposure time for the various exposure sites. The shown regression lines, calculated with omission of the initial values at 0 years, can be expressed by the equations in Table 11.

A linear relationship between weight loss and exposure time for both stone types is accentuated by the high correlation coefficients. The slope of the regression lines is a measure of the annual weight loss and/or recession at a given exposure site. These values, however, include any weight losses incurred during handling, transportation, and oven drying. Losses from handling, transportation, and oven drying can be deduced from weight changes that occurred in unexposed control briquettes from each site and in the ANL control briquettes. Results of cumulative control briquette losses are plotted in Figs. 2a and b for marble and limestone, respectively, as function of cumulative elapsed time between successive weighings. The weight changes of the control briquettes are very small compared to those of their exposed counterparts, and the relationship between weight changes and cumulative elapsed times is less linear, especially in marble, resulting in low correlation coefficients. It can be seen from Figs. 1a and b, however, that these changes are larger for the limestone control briquettes than for the marble ones.

Error bars are one standard deviation of the mean of three determinations (three briquettes). In some cases, briquettes were damaged and the results deleted. Preexposed (or weathered) briquettes were installed in racks at all sites, except NJ in 1988. Half of these weathered briquettes were reexposed with the preexposed surface facing skyward (up), the other half, with the preexposed surface facing groundward (down).

Fig. 1. Cumulative weight loss as a function of cumulative exposure time: (a) fresh marble and (b) fresh limestone. OHMD refers to the dry regime of the movable rack at Ohio; OHMS indicates the sprayed regime of the movable rack.

Fig. 2. Cumulative weight loss of control briquettes as a function of cumulative elapsed time for (a) marble and (b) limestone.

For calculation of averages and standard deviations, no differentiation was made between these methods of reexposure. The results are represented graphically by regression lines in Figs. 3a and b for marble and limestone, respectively. The regression line equations for weight losses of preexposed briquettes are shown in Table 12.

Table 11. Equations for regression lines shown in Figures 1a and b

SAMPLE AND SITE	EQUATION ^a	CORRELATION COEFFICIENT
Fresh Marble		
NC	$y = .246x + .040$	.999
NJ	$y = .356x + .120$	.997
NY	$y = .227x + .072$	.997
DC	$y = .286x + .037$	.998
OHS	$y = .330x + .097$	.998
OHMD	$y = .063x + .046$	1.000
OHMS	$y = .195x + .028$	1.000
Fresh Limestone		
NC	$y = .365x + 1.253$	.973
NJ	$y = .615x + 1.295$	.993
NY	$y = .333x + 1.291$	.998
DC	$y = .671x + .663$	.989
OHS	$y = .835x + .612$	.994
OHMD	$y = .124x + .613$	1.000
OHMS	$y = .352x + .586$	1.000

^ay = cumulative weight loss in grams; x = cumulative exposure period in years

Table 12. Equations for regression lines shown in Figures 3a and b

SAMPLE AND SITE	EQUATION ^a	CORRELATION COEFFICIENT
Fresh Marble		
NC	single point	-
NY	$y = .3465x + .1861$	.994
DC	$y = .3690x + .2670$	.991
OHS	single point	-
OHMS	single point	-
Fresh Limestone		
NC	single point	-
NY	$y = .5452x + 2.2788$	.930
DC	$y = .8948x + 1.6014$	.974
OHS	single point	-
OHMS	single point	-

^ay = cumulative weight loss in grams; x = cumulative exposure time in years

Fig. 3. Cumulative weight loss of preexposed briquettes as a function of cumulative exposure time for (a) marble and (b) limestone.

At the two sites where more than one data point was obtained for preexposed briquettes, a linear relationship between weight loss as function of exposure time is evident from the high correlation coefficient. Determination of any trend in weight loss at the sites where only one measurement was available awaits additional data.

From the slopes of the regression lines, a prorated annual weight loss and/or recession can be deduced. If the prorated annual recession of control briquettes is subtracted from that of exposed briquettes, we obtain the actual recession due to exposure only; see Table 13. A comparison of results shown in Table 11 with those shown in Table 7 of Ref. 3 indicates, in most cases, a slight reduction of recession rate with prolonged exposure. Poor correlation in the weight changes of the simulation and control briquettes will significantly affect the results. The prorated recession rate of briquettes from preexposed materials is greater than that of briquettes from fresh materials. A reason for this finding could be the difference in the origin of the materials and a shorter current exposure time. A problem encountered with the gravimetry samples was chipping of the briquettes, especially the limestone briquettes, during retrieval or transport, thus restricting cumulative weight loss calculations to fewer briquettes.

Table 13. Average recession rates ($\mu\text{m}/\text{yr}$) of exposed briquettes, by exposure site

SAMPLE AND SITE	EXPOSED BRIQUETTES	CONTROL BRIQUETTES	EXPOSURE ALONE
Fresh Marble			
NC	14.5	.1	14.4
NJ	20.9	.0	20.9
NY	13.4	.0	13.4
DC	16.8	.1	16.7
OHS	19.4	.1	19.3
OHMD	3.7	.1	3.6
OHMS	11.5	.1	11.4
Weathered Marble			
NC	Single point	-	-
NY	20.4	.0	20.4
DC	21.7	.1	21.6
OHS	Single point	-	-
OHMS	Single point	-	-
Fresh Limestone			
NC	25.2	1.3	23.9
NJ	42.4	1.7	40.7
NY	23.0	1.2	21.8
DC	46.3	1.7	44.6
OHS	57.6	.6	57.0
OHMD	8.6	.6	8.0
OHMS	24.3	.6	23.7
Weathered Limestone			
NC	Single point	-	-
NY	37.6	1.2	36.4
DC	61.8	1.7	60.1
OHS	Single point	-	-
OHMS	Single point	-	-

Incomplete data, the result of instrument failure^{4,5} made it necessary to abandon efforts to correlate weight losses with amounts of precipitation during the exposure periods. The same is true for other atmospheric data, such as SO₂ and H⁺ concentrations. We anticipate that all of the data will be available after the 10-year exposure period.

Holographic recession and roughness measurements were postponed until the end of the 10-year exposure period because of budgetary restrictions. A comparison of the two recession measurement methods will also be postponed until then.

The results obtained by gravimetry can be summarized as follows:

1. The linear trend of weight loss or recession as a function of exposure time continued over the period 1984-1992, with a high correlation coefficient.

2. Prorated recession for marble, ranging from 11 to 21 $\mu\text{m}/\text{year}$, is half the value for limestone, which ranged from 21 to 45 $\mu\text{m}/\text{year}$, and is dependent on exposure site.

3. Weight changes in simulation and control briquettes are very small and show poor correlation, indicating effects of drying and ambient measurement conditions.

4. The smallest weight losses/recession occurred at OH in the movable rack, dry regime, indicating a strong influence of the amount of rain.

5. A statistical evaluation of rain effect could not be performed, because site data were incomplete.

6. Weight losses of quinquennial briquettes correspond well with the cumulative losses of annual briquettes.

Forthcoming gravimetry results will allow assessment of recession over even longer cumulative exposure times and comparison of the results with those from laser interferometry measurements.

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Appendix:
Weight Losses from Individual Briquettes
(1988-1992)

Table A.1. Weight changes (g) in exposed and control briquettes:
fresh stone, annuals, 1988-1989

SAMPLE AND SITE	EXPOS. TIME, DAYS	SLOT	EXPOSED BRIQUETTES		FIELD CONTROLS		LAB CONTROLS	
			SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE
Marble:								
NC	294	263	H14-17	0.2280	G36-05	0.0023	H14-18	0.0044
		207	G36-19	0.2019	H36-21	0.0026	G36-23	0.0018
		265	G26-02	0.2163	I14-01	0.0030	G26-10	0.0017
NY	315	263	H14-15	0.2028	I14-03	0.0047	H14-18	0.0039
		207	G36-17	0.1954	K36-20	-0.0053	G36-23	0.0046
		265	G26-22	0.1984	G36-07	0.0198	G26-10	0.0045
DC	244	263	H14-10	0.8559 ^a	H36-22	-0.0049	H14-18	-0.0042
		207	G36-22	0.2254	K36-22	-0.0049	G36-23	-0.0064
		265	G26-11	0.2313	G36-08	-0.0053	G26-10	-0.0101
OH ^a	449	263	G34-21	0.4091	K15-13	0.0003	H14-18	-0.0007
		207	K13-08	0.4675	G22-03	-0.0009	G36-23	-0.0015
		265	G22-17	missing	G34-12	0.0007	G26-10	-0.0011
OHMD ^{b,c}	449	353	G24-19	0.0766	-	-	-	-
OHMS ^{c,d}	449	335	K14-14	0.2397	-	-	-	-
Limestone:								
NC	294	163	HU5-05	0.3191	HU6-01	-0.0165	HU1-15	-0.0148
		107	IL4-22	0.2778	GU1-06	-0.0147	HU1-20	-0.0178
		165	GL7-09	0.3247	HU4-24	-0.0146	GL7-03	-0.0129
NY	315	163	HU5-22	0.2715	GU1-12	0.0015	HU1-15	0.0007
		107	IL4-03	0.2511	HU6-24	0.2005 ^a	HU1-20	-0.0020
		165	GL7-18	0.2750	HU5-18	0.2845 ^a	GL7-03	0.0036
DC	244	163	HU5-10	0.5348	GL7-04	-0.0602	HU1-15	-0.0587
		107	IL4-19	2.1678 ^a	GL7-11	-0.0575	HU1-20	-0.0573
		165	GL7-15	0.5459	IU4-15	-0.0575	GL7-03	-0.0537
OH ^a	449	163	IL8-22	1.0897	HL2-05	-0.0764	HU1-15	-0.0585
		107	HL2-12	1.1701	IL8-02	-0.0686	HU1-20	-0.0598
		165	GL6-15	1.1799	GL6-09	-0.0765	GL7-03	-0.0540
OHMD ^{b,c}	449	338	IL8-06	0.1526	-	-	-	-
OHMS ^{c,d}	449	320	HL2-19	0.4320	-	-	-	-

^aChipped stone.

^bOHMD - Ohio site movable rack - dry regime.

^cretrieved in 1990.

^dOHMS - Ohio site movable rack - sprayed regime.

Table A.2. Weight changes (g) in exposed and control briquettes:
fresh stone, annuals, 1989-1990 and 1990-1992

SAMPLE PERIOD, SITE	EXPOS. TIME, DAYS	SLOT	EXPOSED BRIQUETTES		FIELD CONTROLS		LAB CONTROLS	
			SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE
Marble: Period 1989 - 1990								
NY	355	263	H14-15	0.2559	I14-03	-0.0032	H14-18	-0.0042
		207	G36-17	0.2585	K36-20	0.0018	G36-23	-0.0050
		265	G26-22	0.2578	G36-07	0.0041	G26-10	-0.0047
DC	385	263	H14-10	0.3316	H36-22	0.0008	H14-18	0.0009
		207	G36-22	0.2735	K36-22	0.0021	G36-23	0.0017
		265	G26-11	0.2903	G36-08	0.0002	G26-10	0.0067
Limestone: Period 1989 - 1990								
NY	355	163	HU5-22	0.4035	GU1-12	0.0211	HU1-15	0.0209
		107	IL4-03	0.3397	HU6-24	0.0345	HU1-20	0.0203
		165	GL7-18	0.3724	HU5-18	0.0357	GL7-03	0.0163
DC	385	163	HU5-10	0.6498	GL7-04	0.1667 ^a	HU1-15	0.0456
		107	IL4-19	0.7060	GL7-11	0.0441	HU1-20	0.0429
		165	GL7-15	0.6523	IU4-15	0.0457	GL7-03	0.0403
Marble: Period 1990 - 1992								
NY	642	263	H14-15	0.3770	I14-03	-0.0025	H14-18	-0.0025
		207	G36-17	0.3501	K36-20	-0.0028	G36-23	-0.0011
		265	G26-22	0.3660	G36-07	0.0006	G26-10	-0.0018
DC	283	263	H14-10	0.2090	H36-22	0.0042	H14-18	0.0038
		207	G36-22	0.1841	K36-22	0.0013	G36-23	0.0036
		265	G26-11	0.1953	G36-08	0.0041	G26-10	0.0034
Limestone: Period 1990 - 1992								
NY	642	163	HU5-22	0.8371 ^a	GU1-12	0.0264	HU1-15	0.0228
		107	IL4-03	0.5402	HU6-24	-0.0017	HU1-20	0.0195
		165	GL7-18	0.5815	HU5-18	0.0293	GL7-03	0.0193
DC	283	163	HU5-10	0.3022	GL7-04	-0.0184	HU1-15	0.0279
		107	IL4-19	0.2880	GL7-11	-0.0207	HU1-20	0.0270
		165	GL7-15	0.3110	IU4-15	-0.0209	GL7-03	0.0243

^aChipped stone.

Table A.3. Weight changes (g) in exposed and control briquettes:
fresh stone, quinquennials, 1984-1989

SAMPLE AND SITE	EXPOS. TIME, DAYS	SLOT	EXPOSED BRIQUETTES		FIELD CONTROLS		LAB CONTROLS	
			SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE
Marble: Period 1984 - 1989								
NC	1824	219	E26-13	37.8916 ^a	G36-05	0.0078	H14-18	0.0047
		272	A33-08	1.1999	H36-21	0.0071	G36-23	0.0079
		245	I36-21	1.3161	I14-01	0.0070	G26-10	0.0082
NJ	1847	219	A33-14	1.8518	I14-02	0.0000	H14-18	0.0064
		272	A33-22	1.9182	K36-18	0.0017	G36-23	0.0081
		245	I36-01	1.9494	G36-06	0.0011	G26-10	0.0070
NY	1830	219	A33-15	1.1844	I14-03	0.0091	H14-18	0.0098
		272	A33-06	1.2069	K36-20	0.0045	G36-23	0.0127
		245	I36-06	1.1326	G36-07	0.7865 ^a	G26-10	0.0114
DC	1777	219	A33-19	1.4587	H36-22	0.0083	H14-18	0.0042
		272	A33-03	1.4652	K36-22	0.0051	G36-23	0.0061
		245	I36-10	1.4164	G36-08	0.0033	G26-10	-0.0003
Limestone: Period 1984 - 1989								
NC	1824	119	HU5-14	2.9589	HU6-01	0.1043	HU1-15	0.1474
		172	IL4-06	2.9716	GU1-06	0.1359	HU1-20	0.1274
		145	GL7-22	2.9552	HU4-24	0.1114	GL7-03	0.1478
NJ	1847	119	HU5-23	4.4736	GU1-07	1.4230 ^a	HU1-15	0.1941
		172	IL4-18	4.2852	HU6-04	0.1483	HU1-20	0.1731
		145	GL7-06	4.3614	HU5-04	0.1337	GL7-03	0.1890
NY	1830	119	HU5-09	2.8590	GU1-12	0.1225	HU1-15	0.1542
		172	IL4-11	2.8891	HU6-24	0.3210 ^a	HU1-20	0.1332
		145	GL7-19	2.9817	HU5-18	0.3956 ^a	GL7-03	0.1554
DC	1777	119	HU5-24	3.9865	GL7-04	0.2017	HU1-15	0.1939
		172	IL4-23	6.1143*	GL7-11	0.2092	HU1-20	0.1730
		145	GL7-14	4.0747	IU4-15	0.1800	GL7-03	0.1892

^aChipped stone.

Table A.4. Weight changes (g) in exposed and control briquettes:
fresh stone, quinquennials, 1989 runoff

SAMPLE AND SITE	EXPOS. TIME. DAYS	SLOT	EXPOSED BRIQUETTES		FIELD CONTROLS		LAB CONTROLS	
			SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE
Marble:								
NY	56	219	A33-15	0.0483	I14-03	-0.0024	H14-18	-0.0043
		272	A33-06	0.0494	K36-20	0.0033	G36-23	-0.0043
		245	I36-06	0.0511	G36-07	0.0047	G26-10	-0.0036
DC	113	219	A33-19	0.1013	H36-22	0.0044	H14-18	0.0037
		272	A33-03	0.0916	K36-22	0.0043	G36-23	0.0049
		245	I36-10	0.0939	G36-08	0.0038	G26-10	0.0096
Limestone:								
NY	56	119	HU5-09	0.0978	GU1-12	0.0271	HU1-15	0.0269
		172	IL4-11	0.0954	HU6-24	0.0393 ^a	HU1-20	0.0270
		145	GL7-19	0.0959	HU5-18	0.0415 ^a	GL7-03	0.0213
DC	113	119	HU5-24	0.3108	GL7-04	0.1038	HU1-15	0.1106
		172	IL4-23	0.9778 ^a	GL7-11	0.0997	HU1-20	0.1056
		145	GL7-14	0.8232 ^a	IU4-15	0.1017	GL7-03	0.0991

^aChipped stone.

Table A.5. Weight changes (g) in exposed and control briquettes:
preexposed stone, annuals, 1988-1989

SAMPLE AND SITE	EXPOS. TIME, DAYS	SLOT	EXPOSED BRIQUETTES		FIELD CONTROLS		LAB CONTROLS	
			SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE
Marble								
NC	348	259	W22-05 Up	0.5140	G36-05	0.0023	H14-18	0.0044
		251	W22-01 Dn	0.5125	H36-21	0.0026	G36-23	0.0018
					I14-01	0.0030	G26-10	0.0017
NY	378	259	W22-21 Up	0.5305	I14-03	0.0047	H14-18	0.0039
		251	W22-03 Dn	0.5028	K36-20	-0.0053	G36-23	0.0046
					G36-07	0.0198	G26-10	0.0045
DC	380	259	W22-23 Up	0.6004	H36-22	-0.0049	H14-18	-0.0042
		251	W22-12 Dn	0.6693	K36-22	-0.0049	G36-23	-0.0064
					G36-08	-0.0053	G26-10	-0.0101
OH	587	258	W22-24 Up	1.0500	K15-13	0.0003	H14-18	-0.0007
		251	W22-17 Dn	1.1507	G22-03	-0.0009	G36-23	-0.0015
					G34-12	0.0007	G26-10	-0.0011
OHMS ^a	587	312	W23-15 Up	0.3902				
Limestone								
NC	348	159	XW1-06 Up	2.3051	HU6-01	-0.0165	HU1-15	-0.0148
		151	XW1-10 Dn	2.0766	GU1-06	-0.0147	HU1-20	-0.0178
					HU4-24	-0.0146	GL7-03	-0.0129
NY	378	159	PW1-16 Up	2.6921	GU1-12	0.0015	HU1-15	0.0007
		151	XW1-04 Dn	3.1146	HU6-24	0.2005 ^b	HU1-20	-0.0020
					HU5-18	0.2845 ^b	GL7-03	0.0036
DC	380	159	PW1-21 Up	2.8042	GL7-04	-0.0602	HU1-15	-0.0587
		151	PW1-02 Dn	2.1281	GL7-11	-0.0575	HU1-20	-0.0573
					IU4-15	-0.0575	GL7-03	-0.0537
OH	587	158	PW1-23 Up	3.3487	HL2-05	-0.0764	HU1-15	-0.0585
		151	PW1-01 Dn	3.2035	IL8-02	-0.0686	HU1-20	-0.0598
					GL6-09	-0.0765	GL7-03	-0.054
OHMS ^a	587	307	PW1-22 Up	0.6965	-	-	-	-

^aOHMS - Ohio site movable rack - sprayed regime.

^bChipped stone.

Table A.6. Weight changes (g) in exposed and control briquettes:
preexposed stone, annuals, 1989-1990 and 1990-1992

SAMPLE PERIOD, SITE	EXPOS. TIME, DAYS	SLOT	EXPOSED BRIQUETTES		FIELD CONTROLS		LAB CONTROLS	
			SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE	SAMPLE NO.	WEIGHT CHANGE
Marble: Period 1989 - 1990								
NY	259	259	W22-21 Up	0.4054	I14-03	-0.0032	H14-18	-0.0042
		251	W22-03 Dn	0.4117 ^a	K36-20	0.0018	G36-23	-0.0050
					G36-07	0.0041	G26-10	-0.0047
DC	385	259	W22-23 Up	0.4129 ^a	H36-22	0.0008	H14-18	0.0009
		251	W22-12 Dn	0.4721	K36-22	0.0021	G36-23	0.0017
					G36-08	0.0002	G26-10	0.0067
Limestone: Period 1989 - 1990								
NY	259	159	PW1-16 Up	0.9158	GU1-12	0.0211	HU1-15	0.0209
		151	XW1-04 Dn	2.1903 ^a	HU6-24	0.0345	HU1-20	0.0203
					HU5-18	0.0357	GL7-03	0.0163
DC	385	159	PW1-21 Up	0.4307	GL7-04	0.1667 ^b	HU1-15	0.0456
		151	PW1-02 Dn	1.8992 ^a	GL7-11	0.0441	HU1-20	0.0429
					IU4-15	0.0457	GL7-03	0.0403
Marble: Period 1990 - 1992								
NY	642	259	W22-21 Up	0.5696	I14-03	-0.0025	H14-18	-0.0025
		251	W22-03 Dn	0.5256	K36-20	-0.0028	G36-23	-0.0011
					G36-07	-0.0006 ^b	G26-10	-0.0018
DC	283	259	W22-23 Up	0.2683	H36-22	0.0042	H14-18	0.0038
		251	W22-12 Dn	0.1859	K36-22	0.0013	G36-23	0.0036
					G36-08	0.0041	G26-10	0.0034
Limestone: Period 1990 - 1992								
NY	642	159	PW1-16 Up	0.6360	GU1-12	0.0264	HU1-15	0.0228
		151	XW1-04 Dn	0.8466	HU6-24	-0.0017	HU1-20	0.0195
					HU5-18	0.0293	GL7-03	0.0193
DC	283	159	PW1-21 Up	0.4282	GL7-04	-0.0184	HU1-15	-0.0279
		151	PW1-02 Dn	0.4694	GL7-11	-0.0207	HU1-20	-0.0270
					IU4-15	-0.0209	GL7-03	-0.0243

^aExposed upside down.

^bChipped stone.

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