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**COLOR MEASUREMENTS ON MARBLE AND LIMESTONE BRIQUETTES
EXPOSED TO OUTDOOR ENVIRONMENT
IN THE EASTERN UNITED STATES**

VOLUME I: RESULTS OF EXPOSURE 1984-1990

by

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Energy Technology Division
Instrumentation and NDE Section

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ABSTRACT

In a long-term program that began in 1984, limestone and marble briquettes have been exposed to both anthropogenic acid deposition and natural weathering at four field sites in the eastern United States. Similar tests began at an Ohio site in 1986. Effects of exposure on the briquettes and other materials at the sites are evaluated periodically by several federal agencies cooperating in the National Acid Precipitation Assessment Program (NAPAP). One of the primary contributions of Argonne National Laboratory to the NAPAP has been the measurement of tristimulus color change on samples exposed to the environment.

Results from the first six years indicate a yellowing of the marble and a darkening of limestone on both the skyward and groundward surfaces of fresh and preexposed briquettes. The relationship between discoloration and exposure period appears to be linear. Discoloration rates as a function of a cumulative exposure time are almost constant for marble and slightly decreasing for limestone.

Dark spots on groundward surfaces were measured with tristimulus color equipment prior to chemical analysis to determine if a correlation exists between darkening (change in reflectance) and SO_4 concentration. If we take exposure time into consideration, and assume that the airborne concentration of dark particles, which cause darkening, is proportional to airborne SO_2 concentration, we can establish a linear relationship between exposure time, darkening, and SO_2 concentration. The program is continuing so that additional data can be obtained.

1 INTRODUCTION

The effects of acid rain and air pollution on the weathering of Shelburne marble and Salem limestone samples exposed to the outdoor environments at four test sites in the eastern United States are being investigated under the National Acid Precipitation Program (NAPAP).¹ The studies of the effects on stone and other building materials are conducted by several federal agencies cooperating in the NAPAP. Argonne National Laboratory (ANL) participates in the NAPAP effort as a facility of the U.S. Department of Energy, under the sponsorship of the Department of Interior, National Park Service (NPS).

Various methods (e.g., gravimetry, color change, interferometry of roughness/recession change, ion chromatography of surface accumulations, petrographic analyses, and runoff chemistry) are applied to measure stone deterioration. Monitoring of color changes on briquettes after exposure to the environments at the test sites became one of the several research tasks performed by ANL. The method of color measurement was developed by the National Bureau of Standards (NBS) and the base line measurements were performed at the NBS prior to transferring the equipment and task to ANL. The same briquettes were used for gravimetry and measurement of color change.

Environmental exposure of the stone samples was begun in May 1984 at four established NAPAP materials test sites near Raleigh, NC; Chester, NJ; Newcomb, NY; and in Washington, DC. A site in Steubenville, OH was established in 1986 to replace the NJ site. The sites (designated NC, NJ, NY, DC, and OH, respectively) are instrumented to record a wide range of environmental parameters.

Sets of sample briquettes for measurement of color change were installed in exposure racks at each site. Every two years, the briquettes were retrieved, color changes were measured, and the briquettes were returned to their respective field sites for further exposure.

In 1987, additional briquettes were installed in a movable rack at the Ohio site. The movable rack slides into a shelter as soon as a rainfall begins. In the protective shelter, some of the briquettes are sprayed with an amount of deionized water equal to the rainfall per surface area (sprayed regime), while the rest of the briquettes stayed dry (dry regime). At the end of the rainfall, the movable rack leaves the shelter, to be exposed to the environment.

To study color changes of briquettes after prolonged exposure, preexposed slabs of limestone and marble were obtained from buildings destined for demolition or renovation. From these preexposed slabs, briquettes were fabricated and installed at the various sites in 1988. Limestone slabs were preexposed for 80 years and marble slabs were preexposed for 20 years. Two types of briquettes were fabricated: one type with the preexposed surface facing skyward, the other, with the preexposed surface facing groundward.

This report records the results of color measurements on briquettes that were exposed from 1984 to 1990.

The description of experimental procedures is summarized in Section 2 of this report; results are presented in Section 3; and they are discussed in Section 4. Color measurements of individual briquettes are listed in Appendix A.

2 EXPERIMENTAL PROCEDURES

2.1 PREPARATION OF BRIQUETTES FOR EXPOSURE

Briquettes, 51 mm thick, were cut from slabs of marble and limestone obtained by the NAPAP Stone Research Group. The briquette design specified rectangular bottom surfaces

75 X 89 mm (3 x 3.5 in.) and smaller top surfaces 75 X 83 mm (3 x 3.3 in.). Briquettes made to these nominal dimensions were wet-ground at the edges and corners to reduce the probability of any subsequent weight loss due to damage of sharp edges during handling. The top surfaces of marble slabs had been given an 80-grit finish by the supplier. This finish was retained on the tops of the briquettes; the other surfaces were wet-ground to 80 grit at ANL. Top surfaces of limestone slabs had been given a "smooth, planar finish," which was retained on the tops of limestone briquettes. The other surfaces were wet-ground to 180 grit at ANL. After fabrication, the samples were rinsed under flowing distilled water as surface powder was removed with a soft-bristle brush; they were then allowed to dry in a clean laboratory room and they were sealed in bags to await further disposition.

Final shaping of briquettes used for color measurements and gravimetry consisted of rounding all edges and corners by wet-grinding marble with 80-grit and limestone with 180-grit abrasive to minimize weight losses from handling. Three briquettes of each stone type were assigned to each field site for periodic exchange.

The sets of samples were shipped to and from field sites in hard-cased containers lined with soft padding. Each sample was sealed in its bag and wrapped with two layers of plastic bubble wrap. Ground transport was utilized when previously exposed briquettes were present in the shipment to eliminate potential effects of pressure changes, etc., associated with air transport.

At the field sites, the Lucite racks in which the briquettes were exposed held the samples inclined 30° from the horizontal, facing south, and allowed rainwater to drain away freely. The handling protocol specified that vinyl gloves be worn by site operators when briquettes were installed or removed from racks; samples were to be allowed to dry in the rack before removal on or near specified dates and were to be kept in sealed bags when not in the racks.

In 1984, before briquettes were installed in the four original exposure sites (NC, NJ, NY, and DC), base line color of the briquettes was measured by the NBS. After the first year of exposure in 1985, exposed briquettes were weighed, sent to NBS for repeated color measurements, and subsequently returned to the sites for redeployment. The color test schedule was changed to a biennial cycle after the second year of exposure. In 1987, the briquettes from all sites, including OH, were color tested in accordance with the NBS protocol at ANL before they were returned to the sites for redeployment. Base line color of briquettes cut from preexposed slabs of marble and limestone was measured in 1988 before the briquettes were deployed at the four active exposure sites (NC, NY, DC, and OH). Installation of the preexposed briquettes effected a rescheduling of the next color testing, which occurred in 1990.

2.2 TRISTIMULUS PHOTOELECTRIC COLOR MEASUREMENTS

2.2.1 Principle

Diffused reflection of a standard International Commission of Illumination (ICI) or Commission Internationale de l'Éclairage (CIE) illuminant is measured with a photocell at three wavelengths (amber, green, and blue). The angle of illumination is 0° from the

perpendicular with light striking the oblique sample at 90° relative to the sample surface. The viewing angle of the colorimeter is 45° from the perpendicular with light returning at 45° relative to the sample surface. Splitting of the spectrum of the illuminant into the desired wavelengths is accomplished with three interchangeable filters. Three measurements per location are necessary to obtain color information. With proper instrument calibration and color standard surfaces, various parameters, such as ICI color standard system coordinates, chromaticity coordinates, amount of color difference, hue angle, etc., can be computed.¹

2.2.2 Equipment

The colorimeter selected by NBS was a Reflection Meter 670, used in conjunction with a Search Unit 610T, both manufactured by Photovolt Seragen Diagnostics.² The reasons for selecting this colorimeter included its availability, portability, satisfactory accuracy, and repeatability when evaluated by the NBS. The instrument was calibrated by the manufacturer on standard reflectance enamel panels, traceable to the NBS, with Gardner 2005 and 805 colorimeters, prior to its usage on stone briquettes. A plastic template was procured to position the search unit accurately in the center of the "calibration" and "reference" reflectance enamel panels. A removable attachment was fabricated for the search unit to enable accurate and repeatable positioning of the search unit on the skyward surfaces of stone briquettes. Although color measurements in 1984 and 1985 were made at the NBS, the colorimeter, search unit with template and attachment, and the two sets of standard enamel reflectance panels were transferred to ANL for subsequent measurements.

2.2.3 Measurement Procedures

Although an American Society for Testing Materials (ASTM) method for instrumental evaluation of color differences of opaque materials is available,³ no specific procedure for measuring color differences on stone briquettes was available. The NBS developed a procedure⁴ to measure color and color differences on the surfaces of building-stone briquettes with the specific diffuse reflectance, tristimulus colorimeter described above.

The reflection meter is set up according to the manufacturer's operating instruction;² a color filter is selected (amber, green, or blue); the instrument is turned on, with the search unit vertical on a flat clear surface and the light pointing down on that surface, and allowed to warm up for at least 30 min. After a sufficient warmup time, the search unit is placed on one of the standard reflection panels (70-72 for marble and 40-72 for limestone) with the centering template. The sensitivity of the reflectometer is then adjusted to the value for a given color that is specified on the back of the panel and based on 100% of full scale. Multiple repeated meter readings (usually four) are taken at 1-min intervals to check for instrument drift. The last two readings must agree within 0.2 units. After completing this process, the search unit is placed on the "check" panel (70-6 for marble and 40-6 for limestone), with the centering template. At least two repeated readings are taken at 1-min intervals until the last two readings agree within 0.2 units. The average of these last two readings must agree within 1.5 units of the calibration values listed on the back of the "check" panel. Differences larger than 1.5 units necessitate a repetition of the calibration procedure. Use of the "check" panel ensures proper equipment operation and detects any changes in calibration panel reflectance.

If the results for the "check" panel are within limits, the search unit is again placed on the calibration panel with the positioning template and at least two repeated readings at 1-min intervals are taken until the last two readings agree within 0.2 units. The difference between the average of these two readings should not exceed 1.5 units of the calibration values for the particular color listed on the back of the panel. This concludes the instrument calibration on the standard reflectance panels.

The positioning attachment is subsequently attached to the search unit if the above calibration results are acceptable, and readings are taken on the standard briquette (No. C16-21 for marble and No. HU5-12 for limestone). The standard briquette is placed on a clean, flat surface with the beveled and numbered surface facing the operator and the number not inverted (upside down). The search unit is placed in Position 1 (see Fig. 1), with the adapter tightly against the nonbeveled stone surface. At least four repeated readings are recorded at 1-min intervals until the last two readings are within 0.2 units. The search unit is immediately moved and aligned at Position 2 and at least two repeated readings are recorded until the last two readings are within 0.2 units. The search unit is immediately moved to Position 3 and the same reading procedure is followed as at Position 2. The same procedure is followed for Position 4. After readings are obtained, at Position 4, the search unit is moved back to Position 1 and the same procedures are followed as for Positions 2, 3, and 4. The average of the last two readings at Position 1 must agree within 1.0 unit of the average of the original readings at Position 1; if it does not, the calibration procedure on the standard briquette must be repeated.

Fig. 1. Four positions where color of briquettes was measured

The positioning attachment is subsequently removed and the search unit placed on the calibration panel (70-72 or 40-72 for marble or limestone, respectively) with the centering template. At least two repeated readings are taken with the same requirements as during

the original calibration on the standard panel. This measure ensures tolerable instrument drift. If the drift exceeds 1.5 units, the whole calibration procedure must be repeated.

After successful calibration of the equipment, the color of the same stone-color combination can be measured on exposed briquettes. Color measurements of the first exposed briquette follow the same procedures as for the standard briquette, without the end step of recalibration on the standard reflectance panel. The color of the second and third exposed briquette is measured the same way as the first one, but only at least two initial measurements in Position 1 are required. Measurement tolerances remain the same for all exposed briquettes as for the standard briquette.

When the color of no more than three exposed briquettes has been measured, or after 45 min, the positioning attachment is removed from the search unit and the search unit is placed on the standard calibration panel with the centering template. At least four repetitive readings are recorded at 1-min intervals until the same criteria are fulfilled as during the initial calibration. This process ensures satisfactory instrument performance (i.e., no drift). Excessive drift requires retesting of the three exposed briquettes. Satisfactory completion of color measurements on the first set of three exposed briquettes allows color measurement of three additional sets of three briquettes for the same stone-color combination. The acceptance criteria and measurement procedures for these sets remain the same as for the first set. After each set of three briquettes has been measured, the instrument is recalibrated on the standard reflectance panel in the same manner as between the first and second set of three briquettes. After one stone-color combination has been measured or after the color of no more than 12 exposed briquettes has been measured, the instrument is checked for drift and acceptability by repeating the initial standard "check" panel and standard briquette calibration.

It was noticed that after two periods of exposure, 1984 to 1985 and 1985 to 1987, a much darker area was forming on the groundward surface of the exposed briquettes, especially on the limestone briquettes. In 1987 and thereafter, color was measured on the center of the darker area. At least two readings were taken immediately after color had been measured at the four positions on the skyward surface of each briquette. At least two of the readings, at 1-min intervals, had to be within 0.2 units. Because the positioning attachment had to be removed from the search unit, the drift of the instrument was checked on the standard reflectance panel, with the centering template, before measuring the color of the next exposed briquettes. Positioning of the search unit on the groundward surface was done visually because the size and place of the darker spot varied. A more detailed description of the procedure is contained in Ref. 4.

3 RESULTS AND DISCUSSION

The tristimulus colorimeter readings for each briquette are tabulated in Appendix A by site, exposure period, and measuring position on the briquette. These data, however, must be modified to yield useful information in any of the existing colorimetric representation systems. Color is, in general, represented by trilinear coordinates in a color solid. The ICI (or CIE) system is the international standard for color specification. It generally is represented by x and y coordinates specifying hue and saturation, and by Y classifying the

reflectance or perceived color lightness. The other widely accepted color designation system is the Munsell system. It can be visualized as a cylindrical coordinate system. The reflectance, called value, is along the cylinder axis; the hue is specified in numbers from 0-10, and by color name (Y-yellow, etc.) along the periphery; and saturation is expressed in numbers (0-10) radially. The drawback of the Munsell system is the need for color charts adjacent to both sides of the color to be specified. The coordinate accuracy is, therefore, limited to the availability of dye substances. The accuracy of the Munsell system was improved when the Inter-Society Color Council (ISCC) in cooperation with the NBS (currently, National Institute of Standards and Technology, NIST), introduced additional color names and definitions.⁵ A variation to the ICI system is the α - β -uniform chromaticness scale coordinate system, which has the white (MgO) point at the origin of the α - β coordinates. In the ICI coordinate system, the white (MgO) point has the coordinates $x = 0.3101$ and $y = 0.3163$. The lightness L in the α - β system is specified in a way similar to the reflectance Y in the ICI system; i.e., while the coordinates, x , y , and α , β are the same for black and for white, the lightness or reflectance varies from 0 for black to 1.0 for white, with various shades of gray for in-between values. The same is true for the value number in the Munsell system. The α - β system is easily transformed into polar coordinates with: $\phi = \arctg \beta/\alpha$ (hue angle); $S = \sqrt{(\alpha^2 + \beta^2)} Y^{1/2}$ (saturation index); and $L = Y^{1/2}$ (lightness index). Because all of the color coordinate systems are three dimensional, it becomes difficult to represent data graphically as a function of time (fourth dimension). The concept of "yellowness" was introduced to enable rating of white or near-white surfaces. The yellowness value is 0 for MgO (white); it increases for yellowish surfaces and decreases (negative values) for bluish surfaces. The various color coordinate systems are described in greater detail in Ref. 1.

The measured tristimulus readings (G for green, A for amber, and B for blue) can be used to calculate the various coordinates with the following formulas:¹

ICI coordinates:

$$x = \frac{.8A + .18B}{.8A + G + 1.36B}$$

$$y = \frac{G}{.8A + G + 1.36B}$$

$$Y = G/100 \quad (\text{Lightness})$$

α - β coordinates:

$$\alpha = \frac{A - G}{A + B + 2G}$$

$$\beta = \frac{.4(G - B)}{A + B + 2G}$$

$$L = Y^{1/2} = (G/100)^{1/2} \quad (\text{Lightness})$$

Polar coordinates for α - β :

$$\phi = \arctg \frac{\beta}{\alpha} \quad (\text{Hue angle})$$

$$L = Y^{1/2} = (G/100)^{1/2} \quad (\text{Lightness index})$$

$$S = \sqrt{(\alpha^2 + \beta^2)} L \quad (\text{Saturation index})$$

Yellowness:

$$\text{Yellowness} = \frac{A - B}{G}$$

These formulas are approximations because of spectral inaccuracies and deviations from the definition of "standard observer" and "standard illumination source," but the errors are negligible.

Munsell color notations can also be obtained from tristimulus readings, but the procedure is more complicated and requires extrapolation from curves.⁶

The tristimulus colorimeter readings for the calibration and the "check" panels are as follows:

<u>Panel #</u>	<u>Stone Type</u>	<u>Green (G)</u>	<u>Amber(A)</u>	<u>Blue (B)</u>
70-72	Calibration Marble	69.12	68.12	71.05
40-72	Calibration Limestone	39.96	39.40	41.75
70-6	"Check" Marble	69.59	68.52	71.63
40-6	"Check" Limestone	40.33	39.71	42.28

From these tristimulus readings, the following coordinates can be calculated:

<u>Coordinates</u>		<u>70-72</u>	<u>70-6</u>	<u>40-72</u>	<u>40-6</u>
ICI (CIE)	x	.3055	.3052	.3043	.3038
	y	.3138	.3137	.3116	.3112
	Y	.6912	.6959	.3996	.4033
α - β	α	-.0036	-.0038	-.0035	-.0038
	β	-.0028	-.0029	-.0044	-.0048
	L	.8314	.8342	.6321	.6351
Polar α - β	$\phi(^{\circ})$	217.678	217.33	231.97	231.52
	L	.8314	.8342	.6321	.6351
	S	.0042	.0044	.0045	.0049

Yellowness	-0.0424	-0.0447	-0.0588	-0.0637
Munsell system	N 8.54/	N 8.56/	N 6.78/	N 6.81/

Although the yellowness parameter in the Munsell system indicates a bluish tinting, the x, y ICI coordinates are very close to the neutral shades of white to black; therefore, N (neutral) was chosen for color designation.

The data tabulated in Appendix A were averaged by stone type, site, exposure period, and exposure surface and are shown in Appendix B. The various color coordinates were calculated from these averages and are exhibited in Tables 1-22.

Table 1. Average color coordinates
fresh marble - NC

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	
Std.		0.3120	0.3198	0.7080	0.0000	0.0024	0.8414	90.00	0.8414	0.0022	0.0240
Top	0	0.3113	0.3202	0.6880	-0.0011	0.0025	0.8295	113.81	0.8295	0.0025	0.0203
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3126	0.3202	0.7090	0.0004	0.0028	0.8420	82.87	0.8420	0.0026	0.0296
Top	356	0.3137	0.3201	0.7280	0.0017	0.0030	0.8532	60.40	0.8532	0.0032	0.0371
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3130	0.3198	0.7060	0.0011	0.0027	0.8402	68.46	0.8402	0.0027	0.0312
Top	792	0.3161	0.3216	0.7310	0.0034	0.0046	0.8550	52.85	0.8550	0.0053	0.0588
Bottom		0.3158	0.3184	0.7240	0.0052	0.0026	0.8509	26.76	0.8509	0.0054	0.0470

^aStd. = Standard

Table 2. Average color coordinates
fresh marble - NJ

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	
Std.		0.3121	0.3195	0.7070	0.0004	0.0023	0.8408	81.12	0.8408	0.0021	0.0240
Top	0	0.3132	0.3197	0.7120	0.0014	0.0027	0.8438	62.24	0.8438	0.0028	0.0323
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3126	0.3202	0.7090	0.0004	0.0028	0.8420	82.87	0.8420	0.0026	0.0296
Top	367	0.3139	0.3204	0.7280	0.0017	0.0033	0.8532	62.49	0.8532	0.0035	0.0398
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3126	0.3208	0.7040	0.0000	0.0031	0.8390	90.00	0.8390	0.0029	0.0313
Top	818	0.3166	0.3235	0.7190	0.0028	0.0058	0.8479	64.00	0.8479	0.0059	0.0682
Bottom		0.3146	0.3228	0.7450	0.0010	0.0049	0.8631	78.23	0.8631	0.0046	0.0523

^aStd. = Standard

Table 3. Average color coordinates
fresh marble - NY

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	Yellowness
Std.		0.3116	0.3207	0.7080	-0.0011	0.0028	0.8414	110.56	0.8414	0.0028	0.0240
Top	0	0.3141	0.3197	0.7190	0.0024	0.0029	0.8479	50.19	0.8479	0.0035	0.0389
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3119	0.3213	0.7080	-0.0011	0.0033	0.8414	108.06	0.8414	0.0032	0.0282
Top	367	0.3146	0.3212	0.7490	0.0020	0.0039	0.8654	62.65	0.8654	0.0041	0.0467
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3133	0.3199	0.7060	0.0014	0.0028	0.8402	63.43	0.8402	0.0029	0.0340
Top	755	0.3205	0.3205	0.7510	0.0040	0.0039	0.8666	44.03	0.8666	0.0052	0.0546
Bottom		0.3129	0.3129	0.6350	0.0039-0.0017		0.7969	-23.75	0.7969	0.0038	-0.0016
Std.		0.3124	0.3211	0.6990	-0.0004	0.0033	0.8361	96.20	0.8361	0.0031	0.0315
Top	1740	0.3178	0.3243	0.7530	0.0037	0.0066	0.8678	60.70	0.8678	0.0070	0.0797
Bottom		0.3140	0.3193	0.6720	0.0026-0.0027		0.8198	45.81	0.8198	0.0034	0.0372

^aStd. = Standard

Table 4. Average color coordinates
fresh marble - DC

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	Yellowness
Std.		0.3119	0.3208	0.7090	-0.0007	0.0030	0.8420	103.39	0.8420	0.0028	0.0268
Top	0	0.3126	0.3204	0.7150	0.0004	0.0030	0.8456	83.21	0.8456	0.0027	0.0308
Bottom		-	-	-	-	-	-	-	-	-	-
Std.		0.3119	0.3213	0.7080	-0.0011	0.0033	0.8414	108.06	0.8414	0.0032	0.0282
Top	366	0.3148	0.3221	0.7330	0.0017	0.0045	0.8562	69.25	0.8562	0.0045	0.0518
Bottom		0.3191	0.3246	0.7160	0.0050	0.0071	0.8462	55.01	0.8462	0.0079	0.0894
Std.		0.3127	0.3194	0.7050	0.0011	0.0024	0.8396	66.19	0.8396	0.0024	0.0284
Top	858	0.3184	0.3223	0.7200	0.0056	0.0056	0.8485	45.00	0.8485	0.0073	0.0778
Bottom		0.3215	0.3233	0.6840	0.0085	0.0069	0.8270	39.26	0.8270	0.0100	0.1023
Std.		0.3130	0.3204	0.6980	0.0007	0.0030	0.8355	76.61	0.8355	0.0028	0.0330
Top	1853	0.3196	0.3271	0.7220	0.0039	0.0086	0.8497	65.73	0.8497	0.0087	0.0997
Bottom		0.3227	0.3305	0.6420	0.0052	0.0113	0.8012	65.40	0.8012	0.0111	0.1308

^aStd. = Standard

Table 5. Average color coordinates
fresh marble - OH

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	
Std.		0.3125	0.3195	0.7040	0.0007	0.0024	0.8390	73.61	0.8390	0.0023	0.0270
Top	376	0.3186	0.3240	0.6890	0.0048	0.0066	0.8301	54.16	0.8301	0.0074	0.0842
Bottom		0.3153	0.3166	0.6100	0.0057	0.0015	0.7810	14.42	0.7810	0.0052	0.0377
Std.		0.3136	0.3203	0.6970	0.0014	0.0032	0.8349	65.56	0.8349	0.0032	0.0373
Top	1205	0.3236	0.3309	0.6430	0.0060	0.0118	0.8019	63.13	0.8019	0.0118	0.1384
Bottom		0.3257	0.3250	0.5680	0.0009	0.0064	0.7537	82.09	0.7537	0.0056	0.0669

^aStd. = Standard

Table 6. Average color coordinates
fresh marble - OH movable

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3121	0.3205	0.7070	-0.0004	0.0029	0.8408	97.13	0.8408	0.0026	0.0269	-
Top	0	0.3123	0.3197	0.6930	0.0004	0.0025	0.8325	81.63	0.8325	0.0023	0.0260	Dry
Bottom		0.3150	0.3213	0.7380	0.0024	0.0041	0.8591	59.74	0.8591	0.0044	0.0501	Dry
Std.		0.3138	0.3207	0.6970	0.0014	0.0035	0.8349	67.38	0.8349	0.0034	0.0402	-
Top	859	0.3177	0.3247	0.6010	0.0034	0.0067	0.7752	63.43	0.7752	0.0066	0.0799	Dry
Bottom		0.3203	0.3274	0.7380	0.0045	0.0090	0.8591	63.43	0.8591	0.0093	0.1057	Dry
Std.		0.3121	0.3205	0.7070	-0.0004	0.0029	0.8408	97.13	0.8408	0.0026	0.0269	-
Top	0	0.3147	0.3209	0.7440	0.0024	0.0038	0.8626	57.99	0.8626	0.0042	0.0470	Sprayed
Bottom		0.3147	0.3208	0.7330	0.0024	0.0037	0.8562	57.05	0.8562	0.0041	0.0464	Sprayed
Std.		0.3138	0.3207	0.6970	0.0014	0.0035	0.8349	67.38	0.8349	0.0034	0.0402	-
Top	859	0.3168	0.3284	0.7100	0.0000	0.0086	0.8426	90.00	0.8426	0.0079	0.0845	Sprayed
Bottom		0.3149	0.3234	0.7700	0.0010	0.0053	0.8775	79.38	0.8775	0.0050	0.0558	Sprayed

^aStd. = Standard

Table 7. Average color coordinates
weathered marble - NC

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3127	0.3210	0.7060	0.0000	0.0033	0.8402	90.00	0.8402	0.0030	0.0326	-
Top	0	0.3200	0.3278	0.7930	0.0039	0.0091	0.8905	67.09	0.8905	0.0093	0.1047	Up
Bottom		0.3175	0.3212	0.8510	0.0053	0.0047	0.9225	41.63	0.9225	0.0068	0.0682	Up
Top		0.3138	0.3220	0.7730	0.0007	0.0042	0.8792	81.12	0.8792	0.0040	0.0440	Down
Bottom		0.3194	0.3247	0.7860	0.0052	0.0072	0.8866	54.46	0.8866	0.0084	0.0916	Down
Std.		0.3122	0.3212	0.7050	-0.0007	0.0033	0.8396	102.26	0.8396	0.0031	0.0298	-
Top	348	0.3193	0.3248	0.8110	0.0050	0.0072	0.9006	55.41	0.9006	0.0084	0.0912	Up
Bottom		0.3183	0.3249	0.8490	0.0039	0.0070	0.9214	61.15	0.9214	0.0077	0.0848	Up
Top		0.3166	0.3234	0.7810	0.0029	0.0057	0.8837	62.92	0.8837	0.0060	0.0679	Down
Bottom		0.3173	0.3293	0.7500	0.0000	0.0093	0.8660	90.00	0.8660	0.0086	0.0907	Down

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = preexposed surface facing groundward

Table 8. Average color coordinates
weathered marble - NY

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3122	0.3207	0.7070	-0.0004	0.0030	0.8408	96.79	0.8408	0.0028	0.0283	-
Top	0	0.3155	0.3259	0.7390	0.0000	0.0069	0.8597	90.00	0.8597	0.0064	0.0677	Up
Bottom		0.3174	0.3205	0.7960	0.0057	0.0043	0.8922	37.07	0.8922	0.0067	0.0653	Up
Top		0.3174	0.3231	0.8080	0.0041	0.0058	0.8989	54.76	0.8989	0.0067	0.0730	Down
Bottom		0.3131	0.3188	0.7960	0.0019	0.0021	0.8922	48.58	0.8922	0.0027	0.0289	Down
Std.		0.3124	0.3211	0.6990	-0.0004	0.0033	0.8361	96.20	0.8361	0.0031	0.0315	-
Top	733	0.3183	0.3260	0.7270	0.0031	0.0077	0.8526	67.75	0.8526	0.0077	0.0880	Up
Bottom		0.3157	0.3205	0.8110	0.0037	0.0038	0.9006	45.94	0.9006	0.0051	0.0530	Up
Top		0.3214	0.3243	0.7850	0.0077	0.0075	0.8860	44.03	0.8860	0.0101	0.1045	Down ^c
Bottom		0.2892	0.3392	0.7950	-0.0379	0.0076	0.8916	168.69	0.8916	0.0365	-0.0717	Down ^c

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = Preexposed surface facing groundward

^cExposed upside down

Table 9. Average color coordinates
weathered marble - DC

Sample ^a Side	Exp. Days	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3122	0.3207	0.7070	-0.0004	0.0030	0.8408	96.79	0.8408	0.0028	0.0283	-
Top	0	0.3175	0.3255	0.7810	0.0026	0.0071	0.8837	70.02	0.8837	0.0072	0.0807	Up
Bottom		0.3170	0.3125	0.6490	0.0103	-0.0005	0.8056	-2.54	0.8056	0.0092	0.0370	Up
Top		0.3146	0.3224	0.7870	0.0013	0.0046	0.8871	74.48	0.8871	0.0045	0.0508	Down
Bottom		0.3183	0.3233	0.8300	0.0049	0.0061	0.9110	51.34	0.9110	0.0074	0.0795	Down
Std.		0.3130	0.3204	0.6980	0.0007	0.0030	0.8355	76.61	0.8355	0.0028	0.0330	-
Top	765	0.3184	0.3265	0.7800	0.0029	0.0080	0.8832	69.75	0.8832	0.0080	0.0897	Up
Bottom		0.3290	0.3294	0.6520	0.0129	0.0123	0.8075	43.76	0.8075	0.0160	0.1718	Up
Top		0.3203	0.3281	0.7620	0.0040	0.0094	0.8729	66.80	0.8729	0.0095	0.1076	Down ^c
Bottom		0.3195	0.3257	0.8070	0.0047	0.0078	0.8983	58.83	0.8983	0.0086	0.0954	Down ^c

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = preexposed surface facing downward

^cExposed upside down

Table 10. Average color coordinates
weathered marble - OH

Sample ^a Side	Exp. Days	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3127	0.3210	0.7060	0.0000	0.0033	0.8402	90.00	0.8402	0.0030	0.0326	-
Top	0	0.3184	0.3249	0.7650	0.0040	0.0070	0.8746	60.49	0.8746	0.0075	0.0850	Up
Bottom		0.3202	0.3268	0.7450	0.0048	0.0086	0.8631	60.95	0.8631	0.0091	0.1034	Up
Top		0.3164	0.3226	0.7880	0.0032	0.0053	0.8877	58.63	0.8877	0.0058	0.0647	Down
Bottom		0.3170	0.3242	0.8038	0.0028	0.0063	0.8961	65.77	0.8961	0.0065	0.0735	Down
Std.		0.3123	0.3215	0.6970	-0.0007	0.0032	0.8349	101.77	0.8349	0.0032	0.0316	-
Top	587	0.3219	0.3297	0.6810	0.0049	0.0107	0.8252	65.40	0.8252	0.0106	0.1233	Up
Bottom		0.3228	0.3334	0.7460	0.0034	0.0130	0.8637	75.11	0.8637	0.0125	0.1394	Up
Top		0.3226	0.3301	0.7100	0.0054	0.0111	0.8426	64.03	0.8426	0.0113	0.1296	Down
Bottom		0.3244	0.3313	0.7780	0.0066	0.0122	0.8820	61.74	0.8820	0.0131	0.1452	Down

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = preexposed surface facing groundward

Table 11. Average color coordinates
weathered marble - OH movable

Sample ^a Side	Exp. Days	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3127	0.3210	0.7060	0.0000	0.0033	0.8402	90.00	0.8402	0.0030	0.0326	-
Top	0	0.3188	0.3274	0.7250	0.0028	0.0086	0.8515	71.85	0.8515	0.0083	0.0952	Up Sprayed
Bottom		0.3036	0.3234	0.6260	-0.0118	0.0023	0.7912	169.07	0.7912	0.0107	-0.0240	Up Sprayed
Std.		0.3123	0.3215	0.6970	-0.0007	0.0032	0.8349	101.77	0.8349	0.0032	0.0316	-
Top	587	0.3220	0.3321	0.6810	0.0034	0.0121	0.8252	74.29	0.8252	0.0114	0.1307	Up Sprayed
Bottom		0.3178	0.3241	0.5980	0.0038	0.0064	0.7733	59.37	0.7733	0.0066	0.0786	Up Sprayed

^aStd. = Standard

^bUp = preexposed surface facing skyward

Table 12. Average color coordinates
fresh limestone - NC

Sample ^a Side	Exp. Days	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	
Std.	0	0.3387	0.3355	0.4570	0.0196	0.0182	0.6760	42.79	0.6760	0.0220	0.2538
Top		0.3400	0.3364	0.4490	0.0206	0.0190	0.6701	42.68	0.6701	0.0229	0.2650
Bottom		-	-	-	-	-	-	-	-	-	-
Std.	356	0.3388	0.3348	0.4550	0.0203	0.0178	0.6745	41.27	0.6745	0.0221	0.2527
Top		0.3400	0.3372	0.4610	0.0201	0.0194	0.6790	44.03	0.6790	0.0230	0.2668
Bottom		0.3574	0.3383	0.3960	0.0381	0.0243	0.6293	32.51	0.6293	0.0358	0.3864
Std.	792	0.3378	0.3350	0.4580	0.0190	0.0177	0.6768	42.90	0.6768	0.0214	0.2467
Top		0.3418	0.3388	0.4430	0.0209	0.0207	0.6656	44.68	0.6656	0.0240	0.2882
Bottom		0.3432	0.3507	0.4230	0.0149	0.0274	0.6504	61.39	0.6504	0.0252	0.3168

^aStd. = Standard

Table 13. Average color coordinates
fresh limestone - NJ

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	Yellowness
Std.	0	0.3391	0.3353	0.4570	0.0202	0.0182	0.6760	41.99	0.6760	0.0223	0.2560
Top		0.3472	0.3482	0.4640	0.0209	0.0271	0.6812	52.37	0.6812	0.0282	0.3384
Bottom		-	-	-	-	-	-	-	-	-	-
Std.	367	0.3387	0.3355	0.4570	0.0196	0.0182	0.6760	42.79	0.6760	0.0220	0.2538
Top		0.3396	0.3364	0.4460	0.0201	0.0189	0.6678	43.14	0.6678	0.0226	0.2623
Bottom		0.3553	0.3475	0.3740	0.0299	0.0287	0.6116	43.78	0.6116	0.0324	0.3904
Std.	818	0.3388	0.3358	0.4590	0.0196	0.0183	0.6775	43.14	0.6775	0.0221	0.2549
Top		0.3438	0.3399	0.4260	0.0224	0.0218	0.6527	44.22	0.6527	0.0253	0.2981
Bottom		0.3272	0.3014	0.3610	0.0293	0.0043	0.6008	-8.28	0.6008	0.0239	0.0776

^aStd. = Standard

Table 14. Average color coordinates
fresh limestone - NY

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	Yellowness
Std.	0	0.3404	0.3340	0.4540	0.0225	0.0178	0.6738	38.31	0.6738	0.0235	0.2621
Top		0.3423	0.3380	0.4330	0.0220	0.0204	0.6580	42.92	0.6580	0.0244	0.2841
Bottom		-	-	-	-	-	-	-	-	-	-
Std.	367	0.3395	0.3352	0.4570	0.0207	0.0182	0.6760	41.21	0.6760	0.0227	0.2582
Top		0.3412	0.3371	0.4450	0.0214	0.0196	0.6671	42.58	0.6671	0.0237	0.2742
Bottom		-	-	-	-	-	-	-	-	-	-
Std.	755	0.3345	0.3390	0.4630	0.0128	0.0190	0.6804	55.92	0.6804	0.0189	0.2333
Top		0.3367	0.3420	0.4460	0.0134	0.0212	0.6678	57.71	0.6678	0.0205	0.2556
Bottom		0.3325	0.3327	0.4550	0.0146	0.0151	0.6745	45.87	0.6745	0.0172	0.2044
Std.	1740	0.3385	0.3354	0.4600	0.0195	0.0181	0.6782	42.79	0.6782	0.0219	0.2522
Top		0.3468	0.3449	0.3920	0.0226	0.0252	0.6261	48.18	0.6261	0.0268	0.3291
Bottom		0.3401	0.3396	0.4590	0.0186	0.0207	0.6775	48.12	0.6775	0.0229	0.2723

^aStd. = Standard

Table 15. Average color coordinates
fresh limestone - DC

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	
Std.	0	0.3396	0.3344	0.4550	0.0214	0.0178	0.6745	39.75	0.6745	0.0228	0.2571
Top		0.3421	0.3377	0.4330	0.0220	0.0202	0.6580	42.58	0.6580	0.0242	0.2818
Bottom		-	-	-	-	-	-	-	-	-	-
Std.	366	0.3346	0.3448	0.4770	0.0093	0.0221	0.6907	67.18	0.6907	0.0200	0.2474
Top		0.3394	0.3371	0.4500	0.0194	0.0192	0.6708	44.66	0.6708	0.0224	0.2622
Bottom		0.3592	0.3506	0.3420	0.0321	0.0312	0.5848	44.17	0.5848	0.0342	0.4211
Std.	858	0.3374	0.3371	0.4620	0.0173	0.0187	0.6797	47.31	0.6797	0.0210	0.2489
Top		0.3398	0.3415	0.4260	0.0170	0.0217	0.6527	51.82	0.6527	0.0223	0.2746
Bottom		0.3628	0.3587	0.3010	0.0308	0.0363	0.5486	49.65	0.5486	0.0352	0.4585
Std.	1853	0.3384	0.3362	0.4610	0.0189	0.0185	0.6790	44.32	0.6790	0.0218	0.2538
Top		0.3379	0.3420	0.3190	0.0147	0.0215	0.5648	55.71	0.5648	0.0196	0.2633
Bottom		0.3587	0.3573	0.3000	0.0274	0.0346	0.5477	51.66	0.5477	0.0326	0.4300

^aStd. = Standard

Table 16. Average color coordinates
fresh limestone - OH

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S	
Std.	376	0.3386	0.3357	0.4610	0.0195	0.0183	0.6790	43.14	0.6790	0.0220	0.2538
Top		0.3410	0.3375	0.4050	0.0210	0.0198	0.6364	43.39	0.6364	0.0230	0.2741
Bottom		0.3542	0.3475	0.3890	0.0288	0.0284	0.6237	44.60	0.6237	0.0319	0.3830
Std.	1205	0.3387	0.3357	0.4600	0.0195	0.0183	0.6782	43.14	0.6782	0.0220	0.2543
Top		0.3406	0.3422	0.3260	0.0175	0.0223	0.5710	51.84	0.5710	0.0214	0.2822
Bottom		0.3524	0.3489	0.3830	0.0259	0.0287	0.6189	47.86	0.6189	0.0304	0.3734

^aStd. = Standard

Table 17. Average color coordinates
fresh limestone - OH movable

Sample ^a Side	Exp. Days	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.	0	0.3378	0.3350	0.4580	0.0190	0.0177	0.6768	42.90	0.6768	0.0214	0.2467	-
Top		0.3357	0.3324	0.5010	0.0183	0.0157	0.7078	40.55	0.7078	0.0203	0.2255	Dry
Bottom		0.3423	0.3368	0.4520	0.0227	0.0198	0.6723	41.02	0.6723	0.0247	0.2810	Dry
Std.	859	0.3387	0.3357	0.4600	0.0195	0.0183	0.6782	43.14	0.6782	0.0220	0.2543	-
Top		0.3377	0.3385	0.3860	0.0167	0.0195	0.6213	49.43	0.6213	0.0203	0.2539	Dry
Bottom		0.3497	0.3446	0.4030	0.0258	0.0258	0.6348	45.00	0.6348	0.0290	0.3474	Dry
Std.	0	0.3378	0.3350	0.4580	0.0190	0.0177	0.6768	42.90	0.6768	0.0214	0.2467	-
Top		0.3379	0.3352	0.4720	0.0190	0.0178	0.6870	43.14	0.6870	0.0216	0.2479	Sprayed
Bottom		0.3399	0.3357	0.4530	0.0209	0.0186	0.6731	41.56	0.6731	0.0230	0.2627	Sprayed
Std.	859	0.3387	0.3357	0.4600	0.0195	0.0183	0.6782	43.14	0.6782	0.0220	0.2543	-
Top		0.3386	0.3394	0.4240	0.0171	0.0202	0.6512	49.86	0.6512	0.0214	0.2618	Sprayed
Bottom		0.3428	0.3334	0.4300	0.0255	0.0180	0.6557	35.30	0.6557	0.0253	0.2767	Sprayed

^aStd. = Standard

Table 18. Average color coordinates
weathered limestone - NC

Sample ^a Side	Exp. Days	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
		x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.	0	0.3382	0.3367	0.4620	0.0184	0.0187	0.6797	45.52	0.6797	0.0216	0.2532	-
Top		0.3325	0.3328	0.4880	0.0147	0.0151	0.6986	45.81	0.6986	0.0176	0.2049	Up
Bottom		0.3342	0.3297	0.5500	0.0185	0.0139	0.7416	36.87	0.7416	0.0199	0.2091	Up
Top		0.3329	0.3288	0.5310	0.0177	0.0130	0.7287	36.32	0.7287	0.0187	0.1977	Down
Bottom		0.3075	0.3754	0.5910	-0.0383	0.0318	0.7688	140.32	0.7688	0.0436	0.1472	Down
Std.	348	0.3380	0.3353	0.4590	0.0190	0.0179	0.6775	43.27	0.6775	0.0215	0.2484	-
Top		0.3382	0.3408	0.4110	0.0158	0.0209	0.6411	53.02	0.6411	0.0210	0.2628	Up
Bottom		0.3492	0.3480	0.3730	0.0231	0.0275	0.6107	49.91	0.6107	0.0281	0.3512	Up
Top		0.3371	0.3368	0.4350	0.0171	0.0185	0.6595	47.09	0.6595	0.0205	0.2460	Down
Bottom		0.3363	0.3436	0.4370	0.0119	0.0220	0.6611	61.48	0.6611	0.0203	0.2563	Down

^aStd. = Standard

^bUp = Preexposed surface facing skyward; down = preexposed surface facing groundward

Table 19. Average color coordinates
weathered limestone - NY

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3387	0.3357	0.4600	0.0195	0.0183	0.6782	43.14	0.6782	0.0220	0.2543	-
Top	0	0.3314	0.3318	0.5080	0.0141	0.0143	0.7127	45.41	0.7127	0.0169	0.1949	Up
Bottom		0.3493	0.3276	0.4630	0.0364	0.0165	0.6804	24.41	0.6804	0.0329	0.3089	Up
Top		0.3330	0.3326	0.5690	0.0153	0.0151	0.7543	44.66	0.7543	0.0187	0.2074	Down
Bottom		0.3342	0.3278	0.5000	0.0197	0.0128	0.7071	32.87	0.7071	0.0198	0.2040	Down
Std.		0.3385	0.3354	0.4600	0.0195	0.0181	0.6782	42.79	0.6782	0.0219	0.2522	-
Top	733	0.3429	0.3421	0.3870	0.0201	0.0228	0.6221	48.58	0.6221	0.0240	0.2972	Up
Bottom		0.3502	0.3477	0.3110	0.0243	0.0275	0.5577	48.52	0.5577	0.0275	0.3569	Up
Top		0.3538	0.3517	0.3170	0.0257	0.0305	0.5630	49.88	0.5630	0.0299	0.3880	Down ^c
Bottom		0.3419	0.3452	0.3970	0.0171	0.0242	0.6301	54.76	0.6301	0.0235	0.2972	Down ^c

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = preexposed surface facing groundward

^cExposed upside down

Table 20. Average color coordinates
weathered limestone - DC

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3382	0.3367	0.4620	0.0184	0.0187	0.6797	45.52	0.6797	0.0216	0.2532	-
Top	0	0.3328	0.3336	0.4780	0.0145	0.0156	0.6914	47.24	0.6914	0.0177	0.2092	Up
Bottom		0.3341	0.3318	0.5390	0.0170	0.0150	0.7342	41.28	0.7342	0.0194	0.2134	Up
Top		0.3337	0.3327	0.5260	0.0160	0.0154	0.7253	43.76	0.7253	0.0189	0.2129	Down
Bottom		0.3355	0.3359	0.4510	0.0160	0.0175	0.6716	47.73	0.6716	0.0194	0.2328	Down
Std.		0.3384	0.3362	0.4610	0.0189	0.0185	0.6790	44.32	0.6790	0.0218	0.2538	-
Top	765	0.3417	0.3431	0.3300	0.0181	0.0230	0.5745	51.77	0.5745	0.0222	0.2909	Up
Bottom		0.3439	0.3449	0.3230	0.0194	0.0245	0.5683	51.71	0.5683	0.0235	0.3096	Up
Top		0.3508	0.3504	0.2920	0.0234	0.0291	0.5404	51.25	0.5404	0.0274	0.3664	Down ^c
Bottom		0.3410	0.3424	0.3790	0.0178	0.0225	0.6156	51.60	0.6156	0.0225	0.2850	Down ^c

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = preexposed surface facing groundward

Table 21. Average color coordinates
weathered limestone - OH

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3382	0.3367	0.4620	0.0184	0.0187	0.6797	45.52	0.6797	0.0216	0.2532	-
Top	0	0.3331	0.3340	0.4770	0.0145	0.0159	0.6907	47.63	0.6907	0.0179	0.2117	Up
Bottom		0.3313	0.3343	0.5190	0.0124	0.0156	0.7204	51.65	0.7204	0.0169	0.2004	Up
Top		0.3319	0.3320	0.5480	0.0145	0.0145	0.7403	45.19	0.7403	0.0176	0.1989	Down
Bottom		0.3366	0.3376	0.4500	0.0160	0.0188	0.6708	49.51	0.6708	0.0202	0.2444	Down
Std.		0.3387	0.3357	0.4600	0.0195	0.0183	0.6782	43.14	0.6782	0.0220	0.2543	-
Top	587	0.3396	0.3404	0.3250	0.0175	0.0210	0.5701	50.20	0.5701	0.0207	0.2708	Up
Bottom		0.3415	0.3413	0.3530	0.0191	0.0220	0.5941	49.09	0.5941	0.0225	0.2861	Up
Top		0.3363	0.3379	0.3660	0.0155	0.0189	0.6050	50.62	0.6050	0.0190	0.2932	Down
Bottom		0.3412	0.3399	0.3810	0.0197	0.0212	0.6173	47.09	0.6173	0.0227	0.2808	Down

^aStd. = Standard

^bUp = preexposed surface facing skyward; down = preexposed surface facing groundward

Table 22. Average color coordinates
weathered limestone - OH movable

Sample ^a	Exp.	ICI (CIE) Coordinates			α - β Coordinates			Polar α - β Coordinates			Yellowness	Remarks ^b
Side	Days	x	y	Y	α	β	L	$\phi(^{\circ})$	L	S		
Std.		0.3382	0.3367	0.4620	0.0184	0.0187	0.6797	45.52	0.6797	0.0216	0.2532	-
Top	0	0.3352	0.3322	0.4690	0.0180	0.0155	0.6848	40.72	0.6848	0.0196	0.2217	Up Sprayed
Bottom		0.3343	0.3314	0.5380	0.0175	0.0148	0.7335	40.14	0.7335	0.0196	0.2138	Up Sprayed
Std.		0.3387	0.3357	0.4600	0.0195	0.0183	0.6782	43.14	0.6782	0.0220	0.2543	-
Top	587	0.3390	0.3379	0.3900	0.0185	0.0195	0.6245	46.59	0.6245	0.0213	0.2615	Up Sprayed
Bottom		0.3396	0.3421	0.4730	0.0165	0.0220	0.6877	53.13	0.6877	0.0228	0.2748	Up Sprayed

^aStd. = Standard

^bUp = preexposed surface facing skyward

Color changes of briquettes during exposure would attain greater significance if they could be correlated to changes in the concentration of certain chemical entities on the briquette surfaces, especially on the groundward surfaces. To explore such a correlation between the size of dark spots on groundward surfaces and tristimulus color data in the center of the spot, size and color were measured on surface-chemistry briquettes prior to layer removal for chemical analysis. The averaged data, including subsequently measured SO_4 concentrations, are shown in Tables 23 and 24. The tristimulus color readings, calculated color coordinates, measured correction factors, and SO_4 concentrations in Z layers of individual briquettes are tabulated in Appendix C.

Introduction of a correction factor to the measured SO_4 concentrations assumes that the concentration in the dark spot is higher than in the surrounding area. Therefore, the measured SO_4 concentration values are lower, because the powders used for chemical analysis contain SO_4 from both areas. The correction factor is, thus, proportional to the ratio of total removed area to the area removed from the dark spot, assuming uniform concentration throughout the thickness of the removed layer.

A correlation between the corrected chemical concentration of, for example, sulfate in the briquette surface and the darkness of the spot on the groundward surface can be achieved if some assumptions are made. The dark spot is caused by dark particulates in the air, deposited on the surface. Darkening of the spot on the groundward surface was assumed to be proportional to the average SO_2 concentration in air and to the exposure period. Thus, the relationship can be expressed in formula form as

Corrected SO_4 concentration (Z layers in ppm)

$$= \frac{[\text{Exposure period (days)}] \times [\text{Average } \text{SO}_2 \text{ concentration (ppb)}]}{\text{Reflectance (Y)}}$$

The purpose of the color measurements was to assess if outdoor exposure lightens or darkens the briquettes rather than to determine a change in the color itself. The difficulty of plotting three-dimensional color coordinates as a function of exposure time dictated the choice of which parameter to plot as a function of exposure time. Because of the difference in the color of marble and limestone briquettes, we decided to plot "yellowness" and reflectance Y as a function of exposure time for marble and limestone, respectively.

The average values of "yellowness" for fresh marble briquettes are plotted in Figs. 2a and b. Figure 2a shows the yellowing of the skyward surface, while Fig. 2b shows the discoloration on the groundward surface in the center of the briquette (dark spot). On the horizontal axis, the standard is plotted as a function of the time of measurements while the site samples are shown as a function of the actual exposure times. The graphs in Fig. 2 indicate at a glance that, while the standard shows very slight yellowing (even though it is kept in the dark and sealed in a plastic bag), all exposed stones show much more discoloration with time. A linear relationship can be established between yellowing of marble and exposure period if linear regression is considered. The regression equations for fresh marble are shown in Table 25, and for preexposed marble, in Table 26.

Table 23. Average values of dark-spot color coordinates, yellowness, and SO₄ concentrations on marble surface-chemistry briquettes

Site	Exposure Days	Color Coordinates			Yellowness	SO ₄ Measured ppm	SO ₄ Corrected ppm	Remarks
		x	y	Y				
NY	355	.311 ± .000	.318 ± .0005	.743 ± .0054	.0147 ± .0017	610 ± 378	610 ± 378	-
	1112	.310 ± .0016	.321 ± .0012	.722 ± .0136	.0130 ± .0114	1138 ± 444	1138 ± 444	-
	2213	.310 ± .0047	.314 ± .0059	.687 ± .1438	-.010 ± .0559	2390 ± 0 ^a	2390 ± 0 ^a	-
DC	330	.313 ± .004	.323 ± .0013	.682 ± .0272	.0363 ± .0363	2533 ± 536	3426 ± 670	-
	1115	.316 ± .0012	.325 ± .0017	.690 ± .0264	.0690 ± .0067	3080 ± 319	4729 ± 747	-
	2204	.324 ± .0005	.330 ± .0005	.626 ± .0112	.1387 ± .0031	7910 ± 943	13210 ± 787	-
OH	517	.323 ± .0057	.322 ± .0014	.695 ± .0424	.1113 ± .0372	5597 ± 261	7720 ± 688	-
	1272	.318 ± .0009	.325 ± .0005	.707 ± .0155	.082 ± .0064	6323 ± 1187	9295 ± 1350	-
OHM	517	.316 ± .0005	.323 ± .001	.722 ± .0015	.0575 ± .0005	3055 ± 595	3361 ± 655	Dry Dry
	896	.318 ± .0000	.323 ± .0005	.720 ± .0225	.075 ± .003	3865 ± 885	4856 ± 892	
	517	.317 ± .0005	.323 ± .0005	.722 ± .0085	.077 ± .005	3735 ± 755	4729 ± 1063	Sprayed Sprayed
	896	.316 ± .001	.323 ± .0015	.758 ± .0285	.060 ± .0135	3840 ± 0	5491 ± 0	
Std.	NA	.312 ± .001	.321 ± .0008	.702 ± .0022	.028 ± .0041	15	15	-

^aOne briquette

Table 24. Average values of dark-spot color coordinates, yellowness, and SO₄ concentrations on limestone surface-chemistry briquettes

Site	Exposure Days	Color Coordinates			Yellowness	SO ₄ Measured ppm	SO ₄ Corrected ppm	Remarks
		x	y	Y				
NY	355	.336 ± .0014	.336 ± .0013	.504 ± .0125	.235 ± .0134	601 ± 179	709 ± 203	-
	1112	.344 ± .0059	.342 ± .0054	.450 ± .0387	.303 ± .050	4357 ± 1689	6787 ± 2843	-
	2213	.346 ± .0021	.344 ± .0025	.426 ± .0082	.320 ± .0161	3510 ± 143	24149 ± 4119	-
DC	330	.354 ± .0008	.347 ± .0021	.356 ± .0042	.383 ± .0058	13100 ± 3432	20711 ± 4288	-
	1115	.359 ± .0005	.357 ± .0012	.339 ± .0028	.434 ± .0031	24500 ± 3182	32050 ± 4523	-
	2204	.361 ± .0014	.359 ± .0037	.304 ± .0105	.448 ± .0151	32067 ± 5000	47462 ± 4088	-
OH	517	.353 ± .0014	.352 ± .0024	.388 ± .0156	.384 ± .0133	20133 ± 1584	22358 ± 1890	-
	1272	.352 ± .0009	.348 ± .0019	.403 ± .0067	.367 ± .0097	25433 ± 2557	29697 ± 3239	-
OHM	517	.342 ± .0025	.342 ± .0025	.449 ± .029	.287 ± .025	10935 ± 3965	12217 ± 4620	Dry Dry
	896	.349 ± .001	.347 ± .0005	.396 ± .006	.347 ± .0065	21950 ± 3550	22170 ± 3586	
	517	.347 ± .006	.346 ± .0045	.378 ± .047	.331 ± .0505	11570 ± 3630	21966 ± 12994	Sprayed Sprayed
	896	.346 ± .0025	.348 ± .0025	.415 ± .0055	.3265 ± .0125	22100 ± 2000	27908 ± 434	
Std.	NA	.337 ± .000	.337 ± .0005	.461 ± .000	.246 ± .0019	75	75	-

Fig. 2. Yellowing of fresh marble briquettes as a function of exposure time: (a) skyward surface, (b) groundward surface. Vertical bars at each measurement point show the variance (σ). STD = standard; NC = North Carolina; NJ = New Jersey; NY = New York; DC = Washington, DC; OH = Ohio; OHD = Ohio movable rack, dry; and OHS = Ohio, movable rack, sprayed.

Table 25. Regression line equations for yellowing of fresh marble

Surface, Site	Regression line equation ^a	Correlation coefficient R ²
Skyward		
Standard	yellowing = .0015t + .0245	.931
NC ^b	yellowing = .0184t + .0006	1.000
NJ ^b	yellowing = .0163t + .0127	.938
NY	yellowing = .0088t + .0288	.995
DC	yellowing = .0131t + .0244	.938
OH	yellowing = .0229t - .0073	1.000
OHD	yellowing = .0229t - .0668	1.000
OHS	yellowing = .0160t - .0175	1.000
Groundward		
NY	yellowing = .0148t - .0535	1.000
DC	yellowing = .0101t + .0690	.998
OH	yellowing = .0137t - .0220	1.000
OHD ^c	yellowing = .0236t - .0443	1.000
OHS ^c	yellowing = .0041t + .0300	1.000

^at = exposure time in years.

^bOnly a single measurement was available on the groundward surface.

^cOHD = Ohio, movable rack, dry; OHS = Ohio, movable rack, dry.

Table 26. Regression line equations for yellowing of preexposed marble

Surface, Site	Ori-entation ^a	Regression line equation ^b
Skyward		
Standard		yellowing = .00044t + .0302
NC	Up	yellowing = -.0132t + .1171
	Dn	yellowing = .0251t + .0191
NY	Up	yellowing = .0107t + .0542
	Dn	yellowing = .0155t + .0585
DC	Up	yellowing = .0048t + .0752
	Dn	yellowing = .0276t + .0222
OH	Up	yellowing = .0234t + .0622
	Dn	yellowing = .0408t + .0236
OHS	Up	yellowing = .0244t + .0674
Groundward		
NC	Up	yellowing = .0175t + .0507
	Dn	yellowing = -.0009t + .0925
NY	Up	yellowing = -.0061t + .0714
	Dn	yellowing = -.0532t + .0884
DC	Up	yellowing = .0643t - .0273
	Dn	yellowing = .0076t + .0719
OH	Up	yellowing = .0224t + .0810
	Dn	yellowing = .0445t + .0290
OHS	Up	yellowing = .0638t - .0878

^aUp = preexposed surface faced skyward, Dn = preexposed surface faced groundward during exposure.

^bt = time in years of exposure.

The slope of the regression line indicates the rate of discoloration at the various sites. The highest rate at the skyward surface occurs in Ohio and the lowest in New York. At the groundward surface, the rate sequence is different, with the lowest rate occurring at the sprayed movable rack in Ohio and the highest on the dry regime of the movable rack in Ohio. If the discoloration of exposed marble briquettes is caused by airborne particles deposited on the surface, the color of the marble will depend on the color of the particles. Because dark particles will darken and not yellow the surface, "yellowness" values could distort the results somewhat. This is especially true for marble with gray-blue steaks.

Discoloration or yellowing of weathered (preexposed) marble briquettes as a function of exposure time is shown in Figs. 3a and b. Because these briquettes were acquired and exposed later (1987), only two points of measurement per exposure site are available. A superficial inspection of the graphs indicates a yellowing of the surface in some cases and a bleaching or changing towards blue-gray in others (negative slope). The weathered (preexposed) briquettes showed, in general, a much more streaky composition than fresh marble briquettes. The relationship of "yellowness" as a function of exposure time can be expressed as shown in Table 26.

The discoloration rates of preexposed briquettes are similar to the rates of fresh briquettes, with the highest rate occurring in Ohio. Because preexposed briquettes had previously acquired some yellowness, exposing them again at some sites to airborne particulates of a different color, changed the yellowness toward a blue-gray color. This would account for the negative slope of some graphs.

The color of limestone briquettes is light reddish-brown; therefore, the reflectance Y was selected for plotting discoloration (darkening) as a function of exposure time. Figures 4a and b show the graphs of color changes of fresh limestone briquettes. Because the reflection is 1 for white and 0 for black, darkening of a surface will show up as a downward-sloping line. It can be seen from the plot that, while the color of the standard changes only very slightly, the top surface of exposed briquettes darkens with time. Initial rinsing of exposed samples by rain can effect changes in the texture of the surface (and thus in its reflectivity), and result in a slight rise in reflectance of some samples, as evident from the graph of the skyward surfaces. The slopes of the regression lines indicate the rate of darkening, which, for the skyward surfaces, is the largest at the Ohio site and smallest at the New York site. The texture of the measured dark spots on the groundward surfaces is much smoother and shinier than the texture of the skyward surfaces, a circumstance that influences the measurements. The plots of the groundward surfaces show a higher reflectance rate at some sites (NC and NY) and a lower rate at the others. The rate of darkening on the groundward surfaces is, in general, smaller than on the skyward surfaces. Again, the largest change occurs at the movable rack, dry regime in Ohio and the smallest on the movable rack, sprayed regime, also in Ohio. Even though the dark spots on the groundward surfaces appear darker to the observer than the skyward surfaces, the phenomenon is not apparent in the measurements because of a difference in surface texture. A linear relationship between reflectance and exposure time of limestone briquettes can be expressed by the regression line equations presented in Table 27.

Fig. 3. Yellowing of weathered marble briquettes as a function of exposure time: (a) skyward surface, (b) groundward surface. STD = standard; NC = North Carolina, NJ = New Jersey, NY = New York, DC = Washington, DC, OH = Ohio, OHS = Ohio movable rack, sprayed. Up, preexposed surface of briquette faced skyward during exposure; Dn, preexposed surface of briquette faced groundward during exposure. A line through two corresponding measurement points is the regression line with a correlation coefficient $R^2 = 1$. Vertical bars at measurements to show variance (σ).

Fig. 4. Reflectance of fresh limestone briquettes as a function of exposure time: (a) skyward surface, (b) groundward surface. STD = standard; NC = North Carolina, NJ = New Jersey, NY = New York, DC = Washington, DC, OH = Ohio, OHD = Ohio movable rack, dry, OHS = Ohio movable rock, sprayed. Up, preexposed surface of briquette faced skyward during exposure; Dn, preexposed surface of briquette faced groundward during exposure. A line through two corresponding measurement points is the regression line with a correlation coefficient $R^2 = 1$. Vertical bars at measurements show variance (σ).

Table 27. Regression line equations for darkening of fresh limestone

Surface, Site	Regression line equation ^a	Correlation coefficient R ²
Skyward		
Standard	$Y = .00057t + .6745$	.762
NC	$Y = -.0016t + .6752$	.079
NJ	$Y = -.0127t + .6937$	.995
NY	$Y = -.0077t + .6775$	.646
DC	$Y = -.0202t + .6992$	.834
OH	$Y = -.0289t + .7532$	1.000
OHD	$Y = -.0368t + .8550$	1.000
OHS	$Y = -.0153t + .7482$	1.000
Groundward		
NC	$Y = .0165t + .5967$	1.000
NJ	$Y = -.0083t + .6279$	1.000
NY	$Y = .00137t + .6698$	1.000
DC	$Y = -.0079t + .5902$	.604
OH	$Y = -.00216t + .6321$	1.000
OHD	$Y = -.0159t + .7361$	1.000
OHS	$Y = -.0074t + .7027$	1.000

^a t = exposure time in years.

The linear relationship between reflectance and exposure time of fresh limestone briquettes for which more than two measurements were obtained is not as good as the yellowness of marble briquettes under similar conditions, as evident from the correlation coefficients.

Briquettes made from preexposed limestone slabs show a similar relationship between darkening and exposure time. The results are shown in Figs. 5a and b. Later initiation of exposure experiments resulted in only two measurement periods per site. As a consequence, the correlation coefficients for linear regression lines are $R^2 = 1.000$. The linear regression lines equations are presented in Table 28.

All the plots exhibit a negative slope, indicating a darkening of the briquettes. The rate of color change on the skyward surfaces is higher for previously exposed surfaces facing groundward than for previously exposed surfaces re-exposed skywards. For the skyward surface the highest discoloration rate occurs at Ohio and the lowest at New York. On the groundward surface the highest rate is at North Carolina and the lowest at Ohio. Both types of briquettes, marble and limestone exhibit similar discoloration behavior with respect to exposure sites. Briquettes, especially limestone, exposed at NC and NY showed a greenish color on the skyward surface, particularly with longer exposure times.

Fig. 5. Reflectance of weathered limestone briquettes as a function of exposure time: (a) skyward surface, (b) groundward surface. STD = standard; NC = North Carolina, NJ = New Jersey, NY = New York, DC = Washington, DC, OH = Ohio, OHS = Ohio movable rack, sprayed. Up, preexposed surface of briquette faced skyward during exposure; Dn, preexposed surface of briquette faced groundward during exposure. A line through two corresponding measurement points is the regression line with a correlation coefficient $R^2 = 1$. Vertical bars at measurements show variance (σ).

Table 28. Regression line equations for darkening of preexposed limestone

Surface, Site, Orientation ^a	Regression line equation ^b
Skyward	
Standard	$Y = -.0005t + .6795$
NC Up	$Y = -.0613t + .7603$
Dn	$Y = -.0725t + .0801$
NY Up	$Y = -.0453t + .7579$
Dn	$Y = -.0956t + .8500$
DC Up	$Y = -.0556t + .7468$
Dn	$Y = -.0885t + .8139$
OH Up	$Y = -.0752t + .7661$
Dn	$Y = -.0843t + .8244$
OHS Up	$Y = -.0374t + .7222$
Groundward	
NC Up	$Y = -.1374t + .8790$
Dn	$Y = -.1130t + .8818$
NY Up	$Y = -.0612t + .7416$
Dn	$Y = -.0384t + .7455$
DC Up	$Y = -.0792t + .8134$
Dn	$Y = -.0267t + .6983$
OH Up	$Y = -.0785t + .7989$
Dn	$Y = -.0333t + .7041$
OHS Up	$Y = -.0284t + .7619$

^aUp = preexposed surface faced skyward, Dn = preexposed surface faced groundward during exposure.

^bt = exposure time in years.

Even though a good linear relationship exists between darkening or yellowing and exposure time, a better insight can be obtained by plotting the rate of change as a function of cumulative exposure time. The rates of yellowness change for marble and the reflectance for limestone on the skyward surface are shown in Figs. 6a and b, respectively.

Yellowing rates of marble remain constant or diminish slightly with exposure time, except at the New Jersey site, where it increases considerably. Reflectance rate changes, on the other hand, decrease with increasing exposure time, as evident in Fig. 6b. Here again, the New Jersey site shows a slight increase. The change in yellowness and reflectance rate for the Ohio site is not plotted, because only one point is available and the rate can be obtained from the regression line equation. The same is true for the rate changes on the groundward surfaces. More than two data points are available only for the Washington, DC site. The results for that site are plotted in Figs. 7a and b. It is evident from these graphs that the rate of discoloration change on the skyward surface decreases with cumulative exposure time: the opposite is true for the groundward surface (dark spot). Additional measurements will determine if these trends continue.

Fig. 6. Rate of skyward surface discoloration as a function of cumulative exposure time: (a) marble, (b) limestone.

Fig. 7. Rate of discoloration on skyward and groundward surface at the DC site: (a) marble, and (b) limestone.

Although discoloration of the skyward surface was measurable, the changes in SO_4 concentrations, for instance, at these surfaces were minimal. Therefore, non-predried surface-chemistry briquettes were used to assess the existence of a correlation between dark-particle concentration, exposure time and SO_4 concentrations, as previously described. The results are plotted in Figs. 8a and b. Initial SO_4 concentrations of the bulk values of 15 ppm for marble and 75 ppm for limestone were used. A linear trend for corrected SO_4 concentration as function of dark-particle accumulation can be established, as evidenced by regression lines and correlation factors listed in Table 29.

Fig. 8. Corrected SO_4 concentration in Z layers as a function of dark-particle accumulation: (a) fresh marble, (b) fresh limestone.

Table 29. Regression line equations for corrected SO₄ concentrations as a function of SO₂ concentration, exposure time, and reflectance.

Material, Site	Regression line equation ^a	Correlation coefficient R ²
Fresh marble		
NY	SO ₄ conc. = .2459x + 157.0	.977
DC	SO ₄ conc. = .3170x + 278.6	.946
OH	SO ₄ conc. = .2231x + 1556.4	.793
OHD	SO ₄ conc. = .1753x + 215.0	.976
OHS	SO ₄ conc. = .2130x + 438.2	.912
Fresh limestone		
NY	SO ₄ conc. = 1.6713x - 1657	.960
DC	SO ₄ conc. = .5303x + 7794	.894
OH	SO ₄ conc. = .4122x + 3943	.861
OHD	SO ₄ conc. = .4355x + 535.2	.995
OHS	SO ₄ conc. = .5901x + 1353	.962

^ax is the dark-particle accumulation, proportional to the SO₂ concentration, exposure time, and inverse of reflectance.

The average SO₂ concentrations were calculated from EPA atmospheric data. In situations where the measured SO₄ concentrations were outside the average of previous measurements for the same exposure period, only values falling within the range of these averages were considered in the graphs. Low SO₂ concentrations, e.g., at the New York site, result in low dark-particle accumulations over the exposure period, as can be seen in Figs. 8a and b, horizontal axes. The high correlation coefficients of the regression lines indicate that this procedure could be utilized to calculate dark-spot SO₄ concentrations in the Z layers of exposed briquettes. Additional data on briquettes exposed even longer are being accumulated to assess if the linear correlation continues.

The results can be summarized as follows:

1. Yellowness of exposed fresh marble increases linearly with exposure time, on both the skyward and the groundward surface.
2. For preexposed marble, the yellowness of the skyward surfaces increases linearly, while on the groundward surface, the color changes in both directions.
3. Reflectance of fresh limestone decreases linearly on the skyward surface (darkening): on the groundward surface, it generally also decreases (darkens) and changes surface texture at the dark spot.

4. Preexposed limestone shows a linear decrease (darkening) of the reflectance on both the skyward and groundward surface.
5. The linear relationship between discoloration/darkening and exposure time is very good, as confirmed by the high correlation coefficients.
6. The rate of yellowness change on the skyward surface of fresh marble is almost constant, whereas the rate of reflectance on the skyward surface of fresh limestone decreases with cumulative exposure time.
7. Discoloration rates on the groundward surface of fresh stone, available only for the DC site, show a slight increase with cumulative exposure time.
8. A linear correlation can be established between reflectance (darkness), exposure time, and corrected SO_4 concentration in the outermost layer (Z) of the groundward surface, if some assumptions are made. No such correlation could be established for the outermost (A) layer of the skyward surface, because the SO_4 concentration stays unchanged.

Forthcoming additional tristimulus colorimeter readings will allow us to assess discoloration over even longer cumulative exposure periods.

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APPENDIX A

Tristimulus colorimeter readings at each position (described in Fig. 1) for every briquette as a function of exposure site and time.

Table A1. Tristimulus colorimeter readings on fresh marble: NC

Briquette	Post- tion	Year measured/Total days of exposure								
		1984/0			1985/367			1987/792		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	67.5	67.1	64.7	67.9	68.0	65.0	68.0	68.0	65.4
	2	73.0	73.2	71.8	72.9	73.1	71.0	73.0	73.3	71.1
	3	72.5	73.0	71.6	72.8	72.8	71.3	72.2	72.7	71.0
	4	70.0	69.8	68.2	69.9	69.9	68.2	69.0	69.4	67.1
H14-17	1	77.0	77.1	75.0	78.8	79.9	76.3	78.5	80.1	75.2
	2	76.0	76.1	74.0	78.2	79.0	76.0	78.0	79.2	75.0
	3	75.2	75.4	73.5	77.9	78.7	76.3	77.7	78.9	74.6
	4	75.8	76.0	74.0	78.0	79.0	75.9	77.9	79.1	74.9
	Back	-	-	-	-	-	-	74.0	75.9	72.8
G26-02	1	61.5	61.0	60.0	67.0	67.0	64.7	67.6	68.1	64.2
	2	62.4	62.1	61.5	67.4	67.2	65.0	67.3	68.2	64.0
	3	68.3	68.4	67.0	73.2	74.0	70.4	73.9	74.8	70.0
	4	63.2	63.0	62.0	68.2	68.1	65.9	69.4	70.2	65.9
	Back	-	-	-	-	-	-	70.3	70.9	66.7
G36-19	1	58.1	56.3	57.0	63.1	63.0	61.8	64.2	64.1	61.7
	2	68.8	68.0	66.9	73.8	74.0	71.5	73.9	74.8	70.7
	3	70.7	70.0	68.2	75.4	76.0	73.1	76.0	76.9	72.1
	4	68.0	68.0	65.9	73.0	74.0	69.9	73.3	74.7	69.0
	Back	-	-	-	-	-	-	72.9	74.9	72.0

Table A2. Tristimulus colorimeter readings on fresh marble: NJ

Briquette	Post- tion	Year measured/Total days of exposure								
		1984/0			1985/367			1987/818		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	67.8	67.1	64.7	67.9	68.0	65.0	68.1	68.1	65.2
	2	72.9	73.2	70.8	72.9	73.1	71.0	72.7	72.8	70.6
	3	72.0	73.0	72.0	72.8	72.8	71.3	71.9	71.8	70.3
	4	70.0	70.0	68.8	69.9	69.9	68.2	68.8	68.7	66.7
H14-24	1	74.1	75.0	72.0	74.9	75.8	72.1	74.3	75.2	69.8
	2	69.0	69.0	68.1	69.1	69.5	67.1	69.5	69.8	66.2
	3	67.4	67.3	66.0	68.0	68.0	65.7	68.0	68.1	64.2
	4	67.6	67.3	66.0	68.0	68.1	66.0	67.4	67.7	63.8
	Back	-	-	-	-	-	-	74.3	74.2	70.9
G36-10	1	71.0	71.1	69.0	73.1	74.0	70.7	72.4	73.7	68.5
	2	72.0	72.1	70.1	75.0	75.9	73.0	74.0	75.2	69.8
	3	71.9	72.0	69.1	74.1	75.0	71.6	72.8	74.2	68.7
	4	72.0	72.2	70.0	74.2	75.0	72.0	73.0	74.0	69.0
	Back	-	-	-	-	-	-	75.4	75.8	71.7
G36-12	1	71.0	71.7	69.0	73.3	73.4	70.9	72.6	73.3	68.0
	2	71.0	72.0	68.9	72.5	73.0	69.0	71.9	72.5	66.8
	3	73.2	74.1	71.3	75.0	75.2	72.8	73.0	74.0	68.8
	4	74.1	75.0	72.2	76.0	76.8	73.8	74.1	75.0	69.8
	Back	-	-	-	-	-	-	73.7	74.5	70.0

Table A3. Tristimulus colorimeter readings on fresh marble: NY

Briquette	Posi- tion	Year measured/Total days of exposure											
		1984/0			1985/367			1987/755			1990/1740		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	67.5	67.1	64.7	67.8	67.0	64.7	68.2	68.5	65.5	67.6	67.5	64.7
	2	73.0	72.9	70.2	73.0	73.0	70.2	73.1	73.7	71.1	72.5	72.4	70.5
	3	72.7	72.5	71.9	72.4	72.2	71.0	72.2	72.6	70.8	71.7	71.5	69.9
	4	69.9	69.6	68.4	69.8	69.8	68.1	69.0	69.3	66.9	67.7	67.8	65.3
G26-22	1	72.1	73.0	70.6	75.0	75.9	72.9	76.0	76.9	73.4	75.6	76.8	71.5
	2	71.5	72.0	69.8	74.8	75.1	72.8	75.2	76.0	72.8	75.2	76.2	71.6
	3	70.5	71.0	68.5	73.0	74.0	70.9	74.0	75.0	71.8	74.1	75.2	70.2
	4	70.6	71.0	69.1	73.0	73.2	71.0	73.8	75.0	71.6	74.0	75.0	70.3
	Back	-	-	-	-	-	-	73.0	74.3	75.6	74.9	75.8	71.7
H14-15	1	74.0	75.0	71.0	76.0	76.9	71.8	76.3	77.6	72.7	76.1	77.4	69.5
	2	76.0	76.9	73.0	77.8	78.3	74.1	78.2	79.3	75.0	78.0	79.2	72.7
	3	75.1	76.0	73.0	77.2	78.0	74.0	77.2	78.8	74.0	77.0	78.3	71.0
	4	75.4	76.1	73.0	77.2	78.0	74.0	77.5	79.0	74.0	77.0	78.3	70.7
	Back	-	-	-	-	-	-	76.6	78.2	75.8	77.0	78.0	74.0
G36-17	1	72.0	72.2	69.9	75.9	76.1	72.8	74.8	76.0	71.8	75.8	76.9	70.8
	2	72.0	72.5	70.0	76.0	76.9	73.5	76.2	77.2	73.7	76.7	77.7	72.0
	3	65.0	65.0	62.9	70.0	70.0	66.2	68.9	69.7	65.5	70.7	71.2	65.9
	4	69.1	70.0	67.1	73.2	74.0	70.0	73.4	74.6	73.9	73.9	74.9	68.6
	Back	-	-	-	-	-	-	41.0	41.0	42.5	49.8	49.9	50.5

Table A4. Tristimulus colorimeter readings on fresh marble: DC

Briquette	Posi- tion	Year measured/Total days of exposure											
		1984/0			1985/366			1987/858			1990/1853		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	67.5	67.3	65.0	67.8	67.0	64.7	68.1	68.3	65.8	67.6	67.6	65.0
	2	73.0	73.0	71.0	73.0	73.0	70.2	73.0	73.4	71.3	72.3	72.7	70.6
	3	73.0	72.4	71.1	72.4	72.2	71.0	72.0	72.3	70.9	71.4	71.7	69.8
	4	70.0	70.0	68.1	69.8	69.8	68.1	68.9	69.0	67.0	67.7	67.8	65.4
H14-10	1	75.8	76.0	73.4	77.0	78.0	74.0	75.3	77.4	71.3	74.7	76.2	67.8
	2	75.0	75.0	72.5	76.0	77.0	72.9	74.4	76.5	70.0	74.7	76.0	68.0
	3	75.1	75.5	73.0	76.2	77.1	73.5	74.5	76.6	70.5	74.3	75.9	68.2
	4	74.0	74.1	71.1	75.1	76.0	71.3	74.5	75.8	69.0	73.3	74.8	66.8
	Back	-	-	-	75.0	76.1	70.2	71.3	74.0	66.9	66.7	68.0	59.4
G36-22	1	65.0	64.9	62.0	68.0	68.0	64.0	67.1	68.3	63.0	68.9	70.0	62.8
	2	67.0	67.0	65.1	70.0	70.0	67.0	69.0	70.2	65.5	70.3	71.3	65.1
	3	65.9	65.5	63.8	68.0	68.1	64.3	67.0	68.1	62.8	68.0	68.8	62.2
	4	67.7	67.4	66.0	70.0	70.0	66.2	69.0	70.2	65.2	69.0	69.8	63.6
	Back	-	-	-	70.9	71.9	66.5	68.0	69.8	63.3	62.9	64.6	55.8
G26-11	1	73.5	74.0	71.9	74.8	75.0	71.7	73.3	75.0	69.9	73.0	74.0	67.3
	2	74.5	75.0	72.8	76.0	76.6	73.1	74.1	75.9	70.4	74.0	74.9	68.6
	3	72.9	73.0	70.8	74.8	75.0	71.2	73.1	74.9	69.1	73.2	74.2	67.0
	4	71.9	72.0	70.0	74.0	74.8	71.0	73.0	74.6	69.2	72.7	73.7	66.3
	Back	-	-	-	69.0	71.0	63.1	65.9	68.2	61.0	63.0	63.9	56.1

Table A5. Tristimulus colorimeter readings on fresh marble: OH

Briquette	Post- tion	Year measured/Total days of exposure					
		1987/376			1990/1205		
		Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	68.0	68.1	65.6	67.2	67.3	64.0
	2	72.9	73.1	71.4	72.2	72.4	70.6
	3	72.0	72.1	70.9	71.8	71.8	69.9
	4	68.7	69.0	67.0	67.7	68.8	65.4
G34-21	1	68.9	70.0	65.1	62.5	63.6	55.0
	2	64.9	66.6	61.8	61.0	62.0	54.7
	3	67.3	68.5	63.0	64.0	65.0	56.3
	4	65.0	66.0	60.4	62.0	63.0	55.3
	Back	41.8	43.4	43.1	43.3	44.3	42.1
K13-08	1	71.2	73.0	66.2	65.0	67.1	57.3
	2	72.8	74.8	67.6	66.8	68.5	58.9
	3	72.2	73.8	67.6	67.4	69.3	60.1
	4	71.1	72.9	66.0	66.0	67.6	57.9
	Back	70.1	71.4	67.3	70.3	69.7	64.3
G22-17	1	70.2	71.3	65.3	missing		
	2	70.3	71.7	65.4	missing		
	3	66.2	67.0	62.0	missing		
	4	66.1	66.8	62.0	missing		
	Back	71.1	72.3	70.0	missing		

Table A6. Tristimulus colorimeter readings on fresh marble: OH movable

Briquette and Conditions	Post- tion	Year measured/Total days of exposure					
		1987/0			1990/859		
		Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	68.1	68.0	65.5	67.2	67.3	64.2
	2	73.1	73.2	71.1	72.2	72.4	70.2
	3	72.5	72.3	71.0	71.8	71.8	69.7
	4	69.2	69.0	67.0	67.7	68.8	65.1
G24-19 Dry	1	71.0	71.2	69.2	62.4	63.3	57.7
	2	69.9	69.8	68.2	60.0	61.3	56.9
	3	65.0	65.0	64.1	56.8	57.1	53.0
	4	71.2	71.5	69.0	61.1	61.7	56.7
	Back	73.8	74.5	70.8	73.8	75.1	67.3
K14-14 Sprayed	1	73.8	74.4	70.9	71.8	72.1	65.7
	2	73.8	74.5	71.0	70.9	71.3	64.8
	3	75.9	76.5	73.2	70.9	70.1	65.0
	4	74.1	74.8	71.3	70.4	70.4	64.6
	Back	73.3	74.0	70.6	77.0	77.3	73.0

Table A7. Tristimulus colorimeter readings on weathered marble: NC

Briquette and Orientation ^a	Position	Year measured/Total days of exposure					
		1988/0			1990/348		
		Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	68.2	68.2	65.3	68.4	68.2	65.7
	2	73.0	72.9	70.9	73.2	72.8	71.0
	3	72.1	72.3	70.4	72.0	71.9	70.3
	4	69.0	69.0	66.7	68.3	68.3	65.9
W22-05 Up	1	77.4	79.3	69.9	81.9	83.5	75.3
	2	78.8	80.1	72.0	81.1	82.7	75.4
	3	80.9	82.8	74.4	81.2	82.7	76.0
	4	79.9	79.6	72.6	80.0	81.7	74.3
	Back	85.1	86.9	81.1	84.9	86.2	79.0
W22-01 Down	1	79.6	80.2	76.0	80.2	81.5	75.4
	2	72.3	72.0	69.3	72.5	72.7	68.5
	3	76.3	75.7	73.9	77.4	78.0	73.7
	4	81.0	82.2	77.1	82.1	83.9	77.0
	Back	78.6	80.2	73.0	75.0	75.0	68.2

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

Table A8. Tristimulus colorimeter readings on weathered marble: NY

Briquette and Orientation ^a	Position	Year measured/Total days of exposure					
		1988/0			1990/733		
		Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	68.5	68.2	65.4	67.6	67.5	64.7
	2	72.7	72.9	70.9	72.5	72.4	70.5
	3	72.3	72.3	71.0	71.7	71.5	69.9
	4	69.2	69.0	67.0	67.7	67.8	65.3
W22-21 Up	1	70.1	69.8	66.2	73.1	73.9	67.3
	2	73.2	72.8	68.3	78.3	79.2	72.2
	3	74.0	74.7	69.0	68.1	69.5	63.8
	4	78.4	78.1	72.0	71.3	71.7	65.3
	Back	79.6	81.4	76.2	81.1	82.3	78.0
W22-03 Down ^b	1	82.4	83.8	78.0	80.0	83.8	77.0
	2	79.3	80.3	74.0	78.7	80.0	73.0
	3	79.8	81.3	75.6	77.2	79.7	70.3
	4	81.6	83.1	77.0	77.9	80.0	70.3
	Back	79.6	80.7	77.9	79.5	68.1	73.8

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

^bExposed upside down

Table A9. Tristimulus colorimeter readings on weathered marble: DC

Briquette and Orientation ^a	Position	Year measured/Total days of exposure					
		1988/0			1990/765		
		Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	68.5	68.2	65.4	67.7	67.6	65.0
	2	72.7	72.9	70.9	72.3	72.7	70.6
	3	72.3	72.3	71.0	71.4	71.7	69.8
	4	69.2	69.0	67.0	67.7	67.8	65.4
W22-23 Up	1	77.7	78.9	71.8	77.7	78.7	71.5
	2	77.7	79.9	72.6	80.0	81.2	74.5
	3	77.8	76.8	71.8	76.2	77.0	69.2
	4	79.3	79.9	74.3	78.0	78.7	72.3
	Back	64.9	67.6	65.2	65.2	68.5	57.3
W22-12 Down ^b	1	79.8	79.9	76.1	77.0	78.3	69.8
	2	78.2	78.3	73.8	76.4	77.5	69.6
	3	80.0	80.8	76.3	76.9	78.5	69.8
	4	76.8	77.2	74.3	74.3	75.4	67.7
	Back	83.0	84.6	78.0	80.7	82.2	74.5

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

^bExposed upside down

Table A10. Tristimulus colorimeter readings on weathered marble: OH (stationary) and OHM (movable rack)

Briquette Orientation ^a Conditions	Position	Year measured/Total days of exposure					
		1988/0			1990/587		
		Green	Amber	Blue	Green	Amber	Blue
CI6-21 (Standard)	1	68.2	68.2	65.3	67.2	67.3	64.3
	2	73.0	72.9	70.9	72.2	72.0	70.2
	3	72.1	72.3	70.4	71.8	71.4	69.7
	4	69.0	69.0	66.7	67.7	67.4	65.0
W22-24 Up	1	77.3	78.2	72.1	69.2	70.4	62.2
	2	73.4	74.1	68.0	66.0	67.0	59.5
	3	77.5	80.1	72.7	67.7	69.2	60.5
	4	77.6	78.5	71.9	69.3	70.8	61.6
	Back	74.5	75.9	68.2	74.6	75.6	65.2
W22-17 Down	1	80.4	82.0	76.2	71.4	72.9	64.5
	2	78.3	79.2	74.6	69.6	71.1	61.2
	3	78.0	78.6	74.0	73.0	74.9	64.7
	4	78.3	79.3	74.0	70.0	71.2	62.7
	Back	80.3	81.2	75.3	77.8	79.8	68.5
W23-15 Up Sprayed	1	75.0	76.3	68.7	69.3	70.8	60.6
	2	73.9	75.8	67.9	70.8	72.2	62.9
	3	72.8	73.9	67.1	69.0	70.2	60.9
	4	68.2	67.1	61.9	63.2	62.8	55.8
	Back	62.6	59.7	61.2	59.8	60.7	56.0

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

Table A11. Tristimulus colorimeter readings on fresh limestone: NC

Briquette	Position	Year measured/Total days of exposure								
		1984/0			1985/356			1987/792		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.3	48.9	37.1	45.1	48.8	37.1	45.6	48.9	37.6
	2	46.0	49.3	37.9	45.9	49.3	38.0	46.1	49.5	38.1
	3	45.4	49.0	37.2	45.3	48.9	37.3	45.3	49.0	37.8
	4	46.0	49.5	38.0	45.8	49.2	38.0	46.0	49.5	38.0
HU5-05	1	45.0	49.0	36.9	45.8	49.5	36.8	44.0	47.5	34.3
	2	45.2	49.0	37.1	45.8	49.4	36.9	43.9	47.3	34.1
	3	46.0	49.9	38.0	46.7	50.2	37.9	44.8	48.1	35.2
	4	44.6	48.3	36.1	45.0	49.0	36.1	43.2	46.5	33.5
	Back	-	-	-	39.0	41.5	30.1	49.5	49.0	36.1
IL4-22	1	43.0	46.8	34.9	45.4	49.0	36.8	44.6	48.0	35.5
	2	43.0	46.8	34.6	46.8	50.0	38.0	44.5	48.0	35.1
	3	43.0	46.7	35.0	46.2	49.7	38.0	45.6	48.2	37.2
	4	44.9	48.0	37.0	47.2	50.2	40.0	47.1	50.1	39.6
	Back	-	-	-	42.9	47.0	32.9	38.1	41.8	27.5
GL7-09	1	46.0	49.5	37.1	46.0	50.0	37.0	44.4	47.8	35.0
	2	46.3	50.0	37.9	47.0	50.9	38.0	45.7	49.0	36.0
	3	44.5	48.0	36.0	45.0	48.8	36.2	43.3	46.5	34.0
	4	47.0	50.2	38.4	46.0	50.0	37.0	44.2	47.6	34.8
	Back	-	-	-	37.0	48.0	27.6	39.2	43.4	30.3

Table A12. Tristimulus colorimeter readings on fresh limestone: NJ

Briquette	Position	Year measured/Total days of exposure								
		1984/0			1985/367			1987/818		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.3	49.0	37.1	45.2	48.9	37.1	45.7	49.0	37.4
	2	46.0	49.6	37.9	46.0	49.5	38.0	46.2	49.7	37.9
	3	45.6	49.0	37.2	45.5	49.0	37.3	45.7	49.2	37.4
	4	46.0	49.5	38.0	46.0	49.3	38.0	46.0	49.7	38.0
HU5-13	1	48.7	52.5	41.1	45.6	49.1	37.4	43.8	47.8	35.0
	2	48.0	52.0	40.1	45.2	49.0	37.2	43.1	46.8	34.3
	3	47.0	51.0	39.0	45.0	48.8	37.0	42.7	46.3	33.8
	4	47.4	51.1	40.0	44.0	47.8	36.0	42.2	45.8	33.3
	Back	-	-	-	37.0	42.0	27.1	36.4	39.9	25.6
IL4-07	1	47.0	50.9	38.9	44.4	47.9	36.1	42.7	46.2	33.4
	2	46.3	50.0	38.1	45.1	48.5	37.0	43.1	47.7	34.1
	3	46.7	50.4	38.3	45.5	49.0	37.0	43.7	47.3	34.2
	4	47.1	51.0	39.0	45.0	48.1	36.4	43.0	46.7	33.7
	Back	-	-	-	38.1	42.1	27.1	36.7	41.3	25.3
GL7-01	1	46.0	49.0	37.9	44.7	48.0	36.9	42.8	46.2	33.7
	2	44.1	47.8	36.0	43.0	46.5	35.0	41.2	44.8	32.2
	3	44.0	47.2	35.9	43.3	47.0	35.1	41.5	45.0	32.6
	4	45.0	48.2	36.9	43.9	47.1	35.6	41.8	45.3	32.7
	Back	-	-	-	37.0	41.0	27.0	35.3	40.2	24.0

Table A13. Tristimulus colorimeter readings on fresh limestone: NY

Briquette	Position	Year measured/Total days of exposure											
		1984/0			1985/367			1987/755			1990/1740		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.0	49.0	37.1	45.1	48.9	37.1	46.0	48.1	37.5	45.7	49.1	37.7
	2	45.9	49.8	37.9	46.0	49.9	38.0	46.7	48.9	38.1	46.3	49.7	38.0
	3	45.0	49.0	37.1	45.5	49.0	37.2	46.1	48.6	37.6	45.8	49.3	37.7
	4	45.5	49.8	37.9	46.0	49.6	38.0	46.5	48.7	38.0	46.3	49.8	38.0
IL4-03	1	43.0	46.9	34.0	44.0	48.0	35.5	44.1	46.5	35.0	39.6	43.0	30.3
	2	43.6	47.4	35.0	44.9	48.6	36.2	45.0	47.3	35.9	39.3	42.6	30.0
	3	44.0	48.0	35.6	45.0	48.5	36.4	44.9	47.1	36.0	38.5	42.0	29.0
	4	43.1	47.0	35.0	44.1	47.9	35.8	44.4	46.8	35.9	39.6	43.2	29.9
	Back	-	-	-	-	-	-	42.0	45.5	39.8	47.3	50.3	38.4
GL7-18	1	43.2	47.0	35.0	44.2	48.0	36.0	44.3	46.8	35.2	38.4	41.6	29.1
	2	43.0	46.5	34.3	44.2	48.0	35.9	44.4	46.8	35.2	39.3	42.7	30.0
	3	43.2	46.9	35.0	44.0	47.2	35.5	43.8	46.0	34.8	38.8	42.2	29.4
	4	44.0	47.7	35.9	44.9	48.2	36.2	45.0	47.1	35.8	39.0	42.2	29.5
	Back	-	-	-	-	-	-	46.3	48.8	37.8	44.0	47.3	34.5
HU5-22	1	43.3	47.0	34.7	45.0	48.5	36.1	44.8	47.1	35.2	38.8	42.3	29.3
	2	43.7	47.2	35.0	44.3	48.0	36.0	44.7	46.9	35.6	40.0	43.5	30.3
	3	42.4	46.1	33.3	44.0	47.9	35.5	44.2	46.6	35.0	39.3	43.0	29.8
	4	42.9	46.8	33.8	45.1	49.0	36.5	45.6	48.0	36.0	39.3	42.7	30.0
	Back	-	-	-	-	-	-	48.1	50.1	38.8	46.5	50.0	37.3

Table A14. Tristimulus colorimeter readings on fresh limestone: DC

Briquette	Position	Year measured/Total days of exposure											
		1984/0			1985/366			1987/858			1990/1853		
		Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.1	49.0	37.1	45.1	48.9	37.1	45.9	49.0	37.5	45.8	49.2	37.6
	2	46.0	49.5	38.0	46.0	49.9	38.0	46.5	49.7	38.0	46.3	49.7	37.8
	3	45.1	49.0	37.1	45.5	49.0	37.2	46.0	49.1	37.5	45.9	49.3	37.6
	4	45.9	49.5	38.0	46.0	49.6	38.0	46.4	49.5	38.0	46.3	49.8	38.0
HU5-10	1	43.9	47.2	35.1	45.0	48.9	37.0	42.7	45.3	33.9	29.0	30.5	23.0
	2	43.1	47.0	35.0	45.6	49.1	37.1	43.0	45.8	34.0	29.5	31.1	23.3
	3	43.0	47.0	34.5	45.3	49.0	37.0	43.0	45.8	34.0	31.9	33.7	25.4
	4	43.8	47.1	35.4	44.8	48.0	36.0	41.4	44.1	32.8	31.3	33.0	25.0
	Back	-	-	-	35.0	38.9	25.0	30.2	33.7	20.2	29.9	33.0	19.7
IL4-19	1	42.8	46.1	34.1	44.2	47.8	36.0	42.0	44.8	33.3	31.0	32.8	24.3
	2	45.1	49.1	36.9	45.5	49.0	37.1	43.6	46.5	34.4	34.3	36.5	26.9
	3	43.7	47.4	35.1	46.0	49.1	37.9	43.8	46.5	35.1	35.4	37.6	28.7
	4	43.0	47.0	34.1	45.6	49.0	37.0	44.0	46.8	35.0	34.3	36.4	27.0
	Back	-	-	-	34.5	38.3	24.0	30.5	33.8	20.2	30.0	32.8	21.3
GL7-15	1	41.4	45.0	33.0	43.5	46.9	35.0	40.9	43.6	32.0	29.5	31.2	23.0
	2	44.0	47.8	35.1	45.0	48.2	36.9	42.9	45.9	33.8	33.0	34.7	27.0
	3	43.0	47.0	34.8	44.1	47.9	35.9	41.9	44.8	32.6	30.1	31.7	23.8
	4	42.8	46.0	34.0	44.9	48.0	36.7	42.1	45.0	33.2	32.9	34.8	25.7
	Back	-	-	-	33.0	37.9	23.0	29.7	33.2	19.0	30.0	33.6	19.5

Table A15. Tristimulus colorimeter readings
on fresh limestone: OH

Briquette	Posi- tion	Year measured/Total days of exposure					
		1987/376			1990/1205		
		Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.8	49.1	37.5	45.7	49.1	37.6
	2	46.3	50.0	38.1	46.2	49.7	38.0
	3	46.0	49.4	37.8	45.8	49.3	37.7
	4	46.2	49.8	38.0	46.3	49.8	38.0
IL8-22	1	40.4	43.8	32.8	31.7	33.7	25.0
	2	41.2	44.8	33.3	34.0	36.2	26.9
	3	41.0	44.4	33.8	34.6	36.8	27.8
	4	41.3	45.0	33.8	33.3	35.5	26.4
	Back	38.4	42.9	28.1	36.5	40.2	26.3
HL2-12	1	39.8	42.9	32.0	32.3	34.4	25.5
	2	41.0	44.0	32.8	33.5	35.9	26.2
	3	41.5	44.8	33.5	33.4	35.7	26.0
	4	40.0	43.2	32.0	32.8	35.1	25.4
	Back	39.2	43.8	28.9	39.3	43.0	28.7
GL6-15	1	40.8	44.0	32.8	32.0	34.3	24.8
	2	39.9	42.8	31.8	32.0	34.0	24.8
	3	39.9	43.0	32.2	31.3	33.5	24.5
	4	39.5	42.7	31.4	30.4	32.6	23.9
	Back	39.0	43.0	28.0	39.0	43.0	28.3

Table A16. Tristimulus colorimeter readings on
fresh limestone: OH movable

Briquette and Conditions	Posi- tion	Year measured/Total days of exposure					
		1987/0			1990/859		
		Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.6	48.9	37.6	45.7	49.1	37.6
	2	46.1	49.5	38.1	46.2	49.7	38.0
	3	45.3	49.0	37.8	45.8	49.3	37.7
	4	46.0	49.5	38.0	46.3	49.8	38.0
IL8-06 Dry	1	50.9	54.4	43.2	39.3	42.0	32.0
	2	50.2	54.0	42.5	38.2	40.8	30.8
	3	49.3	52.9	41.5	38.4	40.9	31.2
	4	49.9	53.5	42.2	38.3	40.8	31.1
	Back	45.2	49.2	36.5	40.3	44.3	30.3
HL2-19 Sprayed	1	47.1	50.8	38.8	42.4	45.2	34.1
	2	46.8	50.2	38.3	41.8	44.8	33.5
	3	46.8	50.1	38.6	42.7	45.3	34.6
	4	48.1	51.8	40.1	42.6	45.3	34.3
	Back	45.3	49.0	37.1	43.0	47.7	35.4

Table A17. Tristimulus colorimeter readings on weathered limestone: NC

Briquette and Orientations ^a	Position	Year measured/Total days of exposure					
		1988/0			1990/348		
		Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.9	49.1	37.5	45.8	49.0	37.7
	2	46.4	49.8	38.0	46.0	49.5	38.0
	3	46.0	49.3	37.6	45.7	49.2	37.8
	4	46.6	49.7	38.2	46.1	49.6	38.2
XW1-06 Up	1	49.7	52.6	42.6	40.7	43.3	32.3
	2	47.7	50.3	40.3	41.0	43.7	32.9
	3	49.2	52.1	42.0	41.5	43.3	33.3
	4	48.6	51.2	41.6	41.0	43.9	32.7
	Back	55.0	59.0	47.5	37.3	40.6	27.5
XW1-10 Down	1	55.1	59.0	46.5	43.0	45.7	35.0
	2	51.8	55.6	47.8	43.3	46.3	35.4
	3	53.2	56.7	46.6	44.2	46.8	36.6
	4	52.4	56.0	44.2	43.6	46.6	35.7
	Back	59.1	51.0	42.3	43.7	45.7	34.5

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

Table A18. Tristimulus colorimeter readings on weathered limestone: NY

Briquette and Orientation ^a	Position	Year measured/Total days of exposure					
		1988/0			1990/733		
		Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.7	49.1	37.5	45.7	49.1	37.7
	2	46.3	49.8	38.0	46.3	49.7	38.0
	3	45.8	49.3	37.6	45.8	49.3	37.7
	4	46.3	49.7	38.2	46.3	49.8	38.0
PW1-16 Up	1	53.2	56.3	46.1	39.2	42.2	30.8
	2	48.2	50.9	41.3	38.8	41.7	30.3
	3	48.2	50.0	41.0	37.9	41.0	29.6
	4	53.7	57.1	46.5	38.7	41.7	29.9
	Back	46.3	53.0	38.7	31.1	34.0	22.9
XW1-04 Down ^b	1	56.2	60.0	48.6	30.2	33.5	20.7
	2	57.9	61.7	50.1	32.8	36.0	23.7
	3	56.9	59.7	48.0	32.5	35.5	23.8
	4	56.7	59.6	47.1	31.1	34.3	21.6
	Back	50.0	53.9	43.7	39.7	42.3	30.5

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

^bExposed upside down

Table A19. Tristimulus colorimeter readings on weathered limestone: DC

Briquette and Orientation ^a	Position	Year of measured/Total days of exposure					
		1988/0			1990/765		
		Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.9	49.2	37.5	45.8	49.2	37.6
	2	46.4	49.8	38.0	46.3	49.7	37.8
	3	46.0	49.3	37.6	45.9	49.3	37.6
	4	46.6	49.8	38.2	46.3	49.8	38.0
PWI-21 Up	1	48.1	50.9	41.0	32.4	34.7	25.1
	2	48.2	51.0	40.8	33.7	36.0	26.0
	3	47.8	50.5	40.4	34.0	36.4	26.7
	4	47.0	49.7	39.9	32.0	34.0	24.9
	Back	53.9	57.5	46.0	32.3	34.7	24.7
PW1-02 Down ^b	1	52.4	55.8	44.2	28.2	30.7	20.2*
	2	51.5	55.0	43.4	27.8	30.3	19.6*
	3	53.2	56.3	45.2	28.5	31.0	20.1*
	4	53.4	56.5	45.8	32.4	35.0	24.3*
	Back	45.1	47.9	37.4	37.9	40.5	29.7*

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

^bExposed upside down

Table A20. Tristimulus colorimeter readings on weathered limestone: OH (stationary) and OHM (movable rack)

Briquette and Orientation ^a	Position	Year of measured/Total days of exposure					
		1988/0			1990/587		
		Green	Amber	Blue	Green	Amber	Blue
HU5-12 (Standard)	1	45.9	49.2	37.5	45.7	49.1	37.6
	2	46.4	49.8	38.0	46.2	49.7	38.0
	3	46.0	49.3	37.6	45.8	49.3	37.7
	4	46.6	49.8	38.2	46.3	49.8	38.0
PW1-23 Up	1	47.2	49.8	40.0	31.6	33.8	25.1
	2	47.0	50.0	39.4	33.1	35.3	26.3
	3	49.6	52.1	41.8	33.5	35.6	26.8
	4	47.1	49.5	39.9	31.7	34.0	25.2
	Back	51.9	54.4	44.0	35.3	37.9	27.8
PW1-01 Down	1	53.9	56.9	46.3	36.3	38.6	29.8
	2	55.5	58.8	47.8	36.8	39.0	30.0
	3	54.9	58.0	47.0	36.6	38.9	29.9
	4	54.8	58.0	46.7	36.5	38.7	29.8
	Back	45.0	47.8	36.8	38.1	41.0	30.3
PW1-22 Up Sprayed	1	46.2	49.3	39.1	39.0	41.8	31.4
	2	48.0	51.2	40.4	39.0	41.8	31.4
	3	47.3	50.7	40.4	39.2	41.9	32.2
	4	46.1	49.6	39.1	38.9	41.7	31.3
	Back	53.8	57.5	46.0	47.3	50.3	37.3

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

APPENDIX B

Averaged tristimulus colorimeter readings, calculated from readings obtained at all positions (described in Fig. 1) on the top or bottom surface of all briquettes for a given exposure site and exposure period.

The standard deviation was calculated as follows:

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

Table B1. Averaged tristimulus readings, measured and standard deviations, on fresh marble

Year	Position Site	Conditions ^a	Days of Exposure	Exposed Briquettes			Standard		
				Green	Amber	Blue	Green	Amber	Blue
Base Line-1984									
NC	Top		0	68.8±6.1	68.5±6.5	67.1±5.9	70.8±2.2	70.8±2.5	69.1±2.9
	Bottom			-	-	-	-	-	-
NJ	Top		0	71.2±2.1	71.6±2.5	69.3±1.9	70.7±2.0	70.8±2.5	69.1±2.8
	Bottom			-	-	-	-	-	-
NY	Top		0	71.9±2.9	72.6±3.1	69.8±2.7	70.8±2.2	70.5±2.4	68.8±2.7
	Bottom			-	-	-	-	-	-
DC	Top		0	71.5±3.8	71.6±4.0	69.4±3.8	70.9±2.3	70.7±2.2	68.8±2.5
	Bottom			-	-	-	-	-	-
1985									
NC	Top		356	72.8±5.0	73.3±5.5	70.6±5.0	70.9±2.1	71.0±2.1	68.9±2.5
	Bottom			-	-	-	-	-	-
NJ	Top		367	72.8±2.7	73.3±3.0	70.4±2.7	70.9±2.1	71.0±2.1	68.9±2.5
	Bottom			-	-	-	-	-	-
NY	Top		367	74.9±2.2	75.5±2.3	72.0±2.2	70.8±2.1	70.5±2.3	68.5±2.4
	Bottom			-	-	-	-	-	-
DC	Top		366	73.3±3.2	73.8±3.5	70.0±3.5	70.8±2.1	70.5±2.3	68.5±2.4
	Bottom			71.6±2.5	73.0±2.2	66.6±2.9	-	-	-

Table B1. (Contd.)

Year	Position Site	Days of Exposure	Exposed Briquettes			Standard		
			Green	Amber	Blue	Green	Amber	Blue
1987								
NC	Top	792	73.1±4.7	74.1±5.0	69.8±4.6	70.6±2.1	70.9±2.2	68.7±2.5
	Bottom		72.4±1.6	73.9±2.2	70.5±2.7	-	-	-
NJ	Top	818	71.9±2.2	72.7±2.6	67.8±2.0	70.4±2.0	70.4±2.0	68.2±2.3
	Bottom		74.5±0.7	74.8±0.7	70.9±0.7	-	-	-
NY	Top	755	75.1±2.4	76.3±2.5	72.2±2.4	70.6±2.1	71.0±2.2	68.6±2.4
	Bottom		63.5±16.0	64.5±16.7	64.6±15.7	-	-	-
DC	Top	858	72.0±3.0	73.6±3.3	68.0±2.9	70.5±2.1	70.8±2.2	68.8±2.4
	Bottom		68.4±2.2	70.7±2.4	63.7±2.4	-	-	-
OH	Top	376	68.9±2.7	70.2±3.0	64.4±2.3	70.4±2.1	70.6±2.1	68.7±2.5
	Bottom		61.0±13.6	62.4±13.4	60.1±12.1	-	-	-
OHM	Top D	0	69.3±2.5	69.4±2.6	67.6±2.1	70.7±2.1	70.6±2.2	68.7±2.5
	Bottom D		73.8	74.5	70.8	-	-	-
OHM	Top S	0	74.4±0.9	75.1±0.9	71.6±0.9	70.7±2.1	70.6±2.2	68.7±2.5
	Bottom S		73.3	74.0	70.6	-	-	-

Table B1. (Contd.)

Year	Position Site	Conditions ^a	Days of Exposure	Exposed Briquettes			Standard		
				Green	Amber	Blue	Green	Amber	Blue
1990									
NY	Top	1740	75.3±1.9	76.4±2.1	70.4±1.7	69.9±2.2	69.8±2.2	67.6±2.6	
	Bottom		67.2±12.4	67.9±12.8	65.4±10.6	-	-	-	
DC	Top	1853	72.2±2.3	73.3±2.5	66.1±2.1	69.8±2.1	70.0±2.3	67.7±2.5	
	Bottom		64.2±1.8	65.5±1.8	57.1±1.6	-	-	-	
OH ^b	Top	1205	64.3±2.2	65.8±2.6	56.9±1.8	69.7±2.3	70.1±2.1	67.5±2.8	
	Bottom		56.8±13.5	57.0±12.7	53.2±11.1	-	-	-	
OHM	Top D	859	60.1±2.1	60.9±2.3	56.1±1.8	69.7±2.3	70.1±2.1	67.3±2.7	
	Bottom D		73.8	75.1	67.3	-	-	-	
OHM	Top S	859	71.0±0.5	71.0±0.8	65.0±0.4	69.7±2.3	70.1±2.1	67.3±2.7	
	Bottom S		77.0	77.3	73.0	-	-	-	

^aD = dry; S = sprayed

^bAverage of two briquettes

Table B2. Averaged tristimulus readings, measured and standard deviations, on preexposed marble

Year	Site							
Orienta- tion ^a	Position Conditions ^b	Days of Exposure	Exposed Briquettes			Standard		
			Green	Amber	Blue	Green	Amber	Blue
Base Line-1988								
NC Up	Top	0	79.3±1.3	80.5±1.4	72.2±1.6	70.6±2.0	70.6±2.0	68.3±2.4
	Bottom		85.1	86.9	81.1	-	-	-
NC Down	Top	0	77.3±3.4	77.5±4.0	74.1±3.0	70.6±2.0	70.6±2.0	68.3±2.4
	Bottom		78.6	80.2	73.0	-	-	-
NY Up	Top	0	73.9±3.0	73.9±3.0	68.9±2.1	70.7±1.8	70.6±2.0	68.6±2.4
	Bottom		79.6	81.4	76.2	-	-	-
NY Down	Top	0	80.8±1.3	82.1±1.4	76.2±1.5	70.7±1.8	70.6±2.0	68.6±2.4
	Bottom		79.6	80.2	77.9	-	-	-
DC Up	Top	0	78.1±0.7	78.9±1.3	72.6±1.0	70.7±1.8	70.6±2.0	68.6±2.4
	Bottom		64.9	67.6	65.2	-	-	-
DC Down	Top	0	78.7±1.3	79.1±1.4	75.1±1.1	70.7±1.8	70.6±2.0	68.6±2.4
	Bottom		83.0	84.6	78.0	-	-	-
OH Up	Top	0	76.5±1.8	77.7±2.2	71.2±1.9	70.6±2.0	70.6±2.0	68.3±2.4
	Bottom		74.5	75.9	68.2	-	-	-
OH Down	Top	0	78.8±1.0	79.8±1.3	74.7±0.9	70.6±2.0	70.6±2.0	68.3±2.4
	Bottom		80.3	81.2	75.3	-	-	-
OHM Up	Top S	0	72.5±2.6	73.3±3.7	66.4±2.7	70.6±2.0	70.6±2.0	68.3±2.4
	Bottom S		62.6	59.7	61.2	-	-	-

Table B2. (Contd.)

Year	Site	Position Conditions ^b	Days of Exposure	Exposed Briquettes			Standard		
Orienta- tion ^a	Green			Amber	Blue	Green	Amber	Blue	
1990									
NC Up	Top	348	81.1±0.7	82.7±0.6	75.3±0.6	70.5±2.2	70.3±2.1	68.2±2.4	
	Bottom		84.9	86.2	79.0	-	-	-	
NC Down	Top	348	78.1±3.6	79.0±4.2	73.7±3.2	70.5±2.2	70.3±2.1	68.2±2.4	
	Bottom		75.0	75.0	68.2	-	-	-	
NY Up	Top	733	72.7±3.7	73.6±3.6	67.2±3.2	69.9±2.2	69.8±2.2	67.6±2.6	
	Bottom		81.1	82.3	78.0	-	-	-	
NY Down ^c	Top	733	78.5±1.0	80.9±1.7	72.7±2.7	69.9±2.2	69.8±2.2	67.6±2.6	
	Bottom		79.5	68.1	73.8	-	-	-	
DC Up	Top	765	78.0±1.4	78.9±1.5	71.9±1.9	69.8±2.1	70.0±2.3	67.7±2.5	
	Bottom		65.2	68.5	57.3	-	-	-	
DC Down ^c	Top	765	76.2±1.1	77.4±1.2	69.2±0.9	69.8±2.1	70.0±2.3	67.7±2.5	
	Bottom		80.7	82.2	74.5	-	-	-	
OH Up	Top	587	68.1±1.3	69.4±1.5	61.0±1.0	69.7±2.3	69.5±2.2	67.3±2.7	
	Bottom		74.6	75.6	65.2	-	-	-	
OH Down	Top	587	71.0±1.3	72.5±1.5	63.3±1.4	69.7±2.3	69.5±2.2	67.3±2.7	
	Bottom		77.8	79.8	68.5	-	-	-	
OHM Up	Top S	587	68.1±2.9	69.0±3.7	60.1±2.6	69.7±2.3	69.5±2.2	67.3±2.7	
	Bottom S		59.8	60.7	56.0	-	-	-	

^aExplanation Up and Down^bS = sprayed^cExposed upside down

Table B3. Averaged tristimulus readings, measured and standard deviations, on fresh limestone

Year Site	Position, Conditions ^a	Days of Exposure	Exposed Briquettes			Standard		
			Green	Amber	Blue	Green	Amber	Blue
Base Line-1984								
NC	Top	0	44.9±1.3	48.5±1.2	36.6±1.2	45.7±0.3	49.2±0.2	37.6±0.4
	Bottom		-	-	-	-	-	-
NJ	Top	0	46.4±1.9	50.1±1.6	34.4±1.5	45.7±0.3	49.3±0.3	37.6±0.4
	Bottom		-	-	-	-	-	-
NY	Top	0	43.3±0.5	47.0±0.5	34.7±0.7	45.4±0.4	49.4±0.4	37.5±0.4
	Bottom		-	-	-	-	-	-
DC	Top	0	43.3±0.9	47.0±1.0	34.8±0.9	45.5±0.4	49.3±0.3	37.6±0.5
	Bottom		-	-	-	-	-	-
1985								
NC	Top	356	46.1±0.7	49.7±0.6	37.4±1.0	45.5±0.3	49.1±0.2	37.6±0.4
	Bottom		39.6±2.4	45.5±2.9	30.2±2.2	-	-	-
NJ	Top	367	44.6±0.8	48.1±0.8	36.4±0.8	45.7±0.3	49.2±0.2	37.6±0.4
	Bottom		37.4±0.5	41.7±0.5	27.1±0.1	-	-	-
NY	Top	367	44.5±0.4	48.2±0.4	36.0±0.3	45.7±0.4	49.4±0.4	37.6±0.4
	Bottom		-	-	-	-	-	-
DC	Top	366	45.0±0.7	48.4±0.7	36.6±0.7	47.7±0.4	49.4±0.4	37.6±0.4
	Bottom		34.2±0.8	38.4±0.4	24.0±0.8	-	-	-

Table B3. (Contd.)

Year	Position, Site	Days of Exposure	Exposed Briquettes			Standard		
			Green	Amber	Blue	Green	Amber	Blue
1987								
NC	Top	792	44.3±1.0	47.9±0.9	35.4±1.6	45.8±0.3	49.2±0.3	37.9±0.2
	Bottom		42.3±5.1	44.7±3.1	31.3±3.6	-	-	-
NJ	Top	818	42.6±0.8	46.3±0.9	33.6±0.8	45.9±0.2	49.4±0.3	37.7±0.3
	Bottom		36.1±0.6	40.5±0.6	37.7±0.3	-	-	-
NY	Top	755	44.6±0.5	46.9±0.5	35.5±0.4	46.3±0.3	48.6±0.3	37.8±0.3
	Bottom		45.5±2.6	48.1±1.9	38.8±0.8	-	-	-
DC	Top	858	42.6±0.9	45.4±0.9	33.7±0.9	46.2±0.3	49.3±0.3	37.8±0.3
	Bottom		30.1±0.3	33.6±0.3	19.8±0.6	-	-	-
OH	Top	376	40.5±0.7	43.8±0.8	32.7±0.8	46.1±0.2	49.6±0.3	37.9±0.2
	Bottom		38.9±0.3	43.2±0.4	28.3±0.4	-	-	-
OHM	Top D	0	50.1±0.6	53.7±0.6	42.4±0.6	45.8±0.3	49.2±0.3	37.9±0.2
	Bottom D		45.2	49.2	36.5	-	-	-
OHM	Top S	0	47.2±0.5	50.7±0.7	39.0±0.7	45.8±0.3	49.2±0.3	37.9±0.2
	Bottom S		45.3	49.0	37.1	-	-	-

Table B3. (Contd.)

Year	Position, Site	Conditions ^a	Days of Exposure	Exposed Briquettes			Standard		
				Green	Amber	Blue	Green	Amber	Blue
1990									
NY	Top	Bottom	1740	39.2±0.5	42.6±0.5	29.7±0.4	46.0±0.3	49.5±0.3	37.9±0.2
				45.9±1.4	49.2±1.3	36.7±1.6	-	-	-
DC	Top	Bottom	1853	31.9±2.0	33.7±2.3	25.3±1.8	46.1±0.2	49.5±0.3	37.8±0.2
				30.0±0.05	33.1±0.3	20.2±0.8	-	-	-
OH	Top	Bottom	1205	32.6±1.2	34.8±1.2	25.6±1.1	46.0±0.3	49.5±0.3	37.8±0.2
				38.3±1.3	42.1±1.3	27.8±1.0	-	-	-
OHM	Top D	Bottom D	859	38.6±0.4	41.1±0.5	31.3±0.4	46.0±0.3	49.5±0.3	37.8±0.2
				40.3	44.3	30.3	-	-	-
OHM	Top S	Bottom S	859	42.4±0.3	45.2±0.2	34.1±0.4	46.0±0.3	49.5±0.3	37.8±0.2
				43.0	47.3	35.4	-	-	-

^aD = dry, S = sprayed

Table B4. Averaged tristimulus readings, measured and standard deviations, on preexposed limestone

Year	Site							
Orienta- tion ^a	Position, Conditions ^b	Days of Exposure	Exposed Briquettes			Standard		
			Green	Amber	Blue	Green	Amber	Blue
Base Line 1988								
NC Up	Top	0	48.8±0.7	51.6±0.9	41.6±0.8	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom		55.0	59.0	47.5	-	-	-
NC Down	Top	0	53.1±1.2	56.8±1.3	46.3±1.3	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom		59.1	51.0	42.3	-	-	-
NY Up	Top	0	50.8±2.6	53.6±3.2	43.7±2.6	46.0±0.3	49.5±0.3	37.8±0.3
	Bottom		46.3	53.0	38.7	-	-	-
NY Down	Top	0	56.9±0.6	60.3±0.9	48.5±1.1	46.0±0.3	49.5±0.3	37.8±0.3
	Bottom		50.0	53.9	43.7	-	-	-
DC Up	Top	0	47.8±0.5	50.5±0.5	40.5±0.4	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom		53.9	57.5	46.0	-	-	-
DC Down	Top	0	52.6±0.7	55.9±0.6	44.7±0.9	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom		45.1	47.9	37.4	-	-	-
OH Up	Top	0	47.7±1.1	50.4±1.0	40.3±0.9	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom		51.9	54.4	44.0	-	-	-
OH Down	Top	0	54.8±0.6	57.9±0.7	47.0±0.6	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom		45.0	47.8	36.8	-	-	-
OHM Up	Top S	0	46.9±0.8	50.2±0.8	39.8±0.7	46.2±0.3	49.5±0.3	37.8±0.3
	Bottom S		53.8	57.5	46.0	-	-	-

Table B4. (Contd.)

Year Site Orienta- tion ^a	Position, Conditions ^b	Days of Exposure	Exposed Briquettes			Standard		
			Green	Amber	Blue	Green	Amber	Blue
1990								
NC Up	Top	348	41.1±0.3	43.6±0.3	32.8±0.4	45.9±0.2	49.3±0.2	37.9±0.2
	Bottom		37.3	40.6	27.5	-	-	-
NC Down	Top	348	43.5±0.4	46.4±0.4	35.7±0.6	45.9±0.2	49.3±0.2	37.9±0.2
	Bottom		43.7	45.7	34.5	-	-	-
NY Up	Top	733	38.7±0.5	41.7±0.4	30.2±0.5	46.0±0.3	49.5±0.3	37.9±0.2
	Bottom		31.1	34.0	22.9	-	-	-
NY Down ^c	Top	733	31.7±1.1	34.8±1.0	22.5±1.3	46.0±0.3	49.5±0.3	37.9±0.2
	Bottom		39.7	42.3	30.5	-	-	-
DC Up	Top	765	33.0±0.8	35.3±1.0	25.7±0.7	46.1±0.2	49.5±0.3	37.8±0.2
	Bottom		32.3	34.7	24.7	-	-	-
DC Down ^c	Top	765	29.2±1.8	31.8±1.9	21.1±1.9	46.1±0.2	49.5±0.3	37.8±0.2
	Bottom		37.9	40.5	29.7	-	-	-
OH Up	Top	587	32.5±0.8	34.7±0.8	25.9±0.7	46.0±0.3	49.5±0.3	37.8±0.2
	Bottom		35.3	37.9	27.8	-	-	-
OH Down	Top	587	36.6±0.2	38.8±0.2	29.9±0.1	46.0±0.3	49.5±0.3	37.8±0.2
	Bottom		38.1	41.0	30.3	-	-	-
OHM Up Spray	Top S	587	39.0±0.1	41.8±0.1	31.6±0.4	46.0±0.3	49.5±0.3	37.8±0.2
	Bottom S		47.3	50.3	37.3	-	-	-

^aUp - preexposed surface facing skyward

Down - preexposed surface facing groundward

APPENDIX C

Tristimulus readings obtained at the center dark spot of the groundward side of surface-chemistry briquettes prior to chemical analysis. The values of the standard are averages ($\pm$ variance) of four readings at the four positions described in Fig. 1.

Table C1. Tristimulus readings obtained on surface-chemistry briquettes of marble (dark area on groundward side)

Site	Briquette	Exposed Briquettes			Standard		
		Green	Amber	Blue	Green	Amber	Blue
NY	F15-06	74.2	74.3	73.3	70.0 ± 2.1	70.1 ± 2.3	67.8 ± 2.5
	F15-21	73.7	73.7	72.7			
	H16-12	75.0	75.0	73.7			
	G24-24	70.4	70.0	69.5			
	K14-08	72.5	72.0	69.9			
	M15-15	78.6	79.4	77.0			
	H11-18	48.4	48.2	52.5			
	L26-17	79.2	79.8	77.6			
	G24-02	73.7	72.2	72.0			
DC	H16-24	67.65	65.6	65.7	70.1 ± 2.2	70.3 ± 2.2	68.3 ± 2.6
	F15-14	65.2	65.4	61.3			
	H16-01	71.8	72.7	68.3			
	M15-20	61.7	63.6	55.2			
	H11-06	62.0	63.7	55.2			
	L26-05	64.2	66.2	57.0			
	K14-15	70.4	71.5	66.0			
	G24-23	65.3	65.0	60.6			
	G24-10	71.3	72.0	67.6			
OH	J36-20	64.0	69.6	59.2	70.5 ± 2.0	70.0 ± 2.1	68.4 ± 2.5
	L26-01	74.25	75.8	70.1			
	J36-23	70.3	72.3	65.7			
	K13-18	70.7	71.2	65.7			
	K14-21	72.6	74.1	67.5			
	G34-09	68.8	69.5	64.2			
	J36-18	72.3	72.3	68.2			
	I11-04	72.0	72.7	68.5			
	J36-19	71.3	72.3	67.2			
	K36-23	73.0	73.9	69.4			
	K14-19	74.2	75.8	70.0			
	G24-06	69.7	71.0	66.0			
	K14-03	72.85	73.8	68.5			
	G24-11	78.6	79.3	75.7			

Table C2. Tristimulus readings obtained on surface-chemistry briquettes of limestone (dark area on groundward side)

Site	Briquette	Exposed Briquettes			Standard		
		Green	Amber	Blue	Green	Amber	Blue
NY	JL6-18	48.6	52.0	40.1	46.1 ± .3	49.0 ± .3	37.8 ± .2
	JL5-02	51.2	54.5	42.0			
	EL3-14	51.3	54.4	43.3			
	GL6-14	50.5	53.9	42.05			
	HL2-06	42.3	45.9	32.3			
	HL8-24	42.25	46.1	31.2			
	HL4-14	41.8	45.7	32.5			
	IU5-17	43.7	47.0	33.8			
	GL2-24	42.2	45.8	31.4			
DC	JL6-16	35.0	38.7	25.1	46.1 ± .3	49.2 ± .3	37.8 ± .2
	EL3-07	35.7	40.0	26.3			
	JL5-18	36.0	39.8	26.3			
	HL8-22	33.65	37.1	22.35			
	GL6-19	33.7	37.25	22.7			
	HL8-08	34.3	38.1	23.3			
	HL4-24	30.0	33.35	19.75			
	IU5-08	31.8	35.3	21.7			
	GL7-17	29.3	32.3	18.7			
OH	GL8-21	37.5	41.1	26.6	46.1 ± .2	49.3 ± .3	37.9 ± .2
	ML3-05	37.9	41.4	26.3			
	GL8-23	41.0	44.9	29.9			
	GL3-12	41.2	45.3	30.4			
	IL8-11	39.6	43.4	28.3			
	HL2-08	40.1	44.0	29.6			
	GL8-22	47.8	51.0	38.5			
	ML3-11	42.0	45.3	32.2			
	GL8-19	42.5	45.5	33.6			
	ML3-19	33.1	36.4	23.8			
	GL6-12	40.2	43.8	30.15			
	HL8-02	39.0	42.7	28.95			
	GL6-04	40.85	44.55	30.7			
	HL8-05	42.0	44.5	31.3			

Table C3. Averaged color coordinates, and SO₂ and SO₄ data; fresh marble
(dark-spot data)

Site	Exp. Days	Briquette	Yellow	Corr. Factor	Color Coordinates			SO ₂ Ave. ppb	SO ₄ ppm Z Layer	SO ₄ Corr. ppm Z Layer	Remarks
					x	y	Y				
NY	NA	Standard	.033	NA	.313	.321	.700	NA	15	15	
	355	F15-06	.013	1.00	.311	.318	.742	2.1	836	836	
	355	F15-21	.014	1.00	.311	.318	.737	2.1	916	916	
	355	H16-12	.017	1.00	.311	.319	.750	2.1	78	78	
	1112	G24-24	.007	1.00	.310	.319	.704	2.8	1750	1750	
	1112	G24-02	.003	1.00	.308	.321	.737	2.8	955	955	
	1112	K14-08	.029	1.00	.312	.322	.725	2.8	709	709	
	2213	M15-15	.031	1.00	.313	.318	.786	2.8	80	80	
	2213	H11-18	-.089	1.00	.303	.306	.484	2.8	2390	2390	dark vein
	2213	L26-17	.028	1.00	.313	.319	.792	2.8	339	339	
DC	NA	Standard	.029	NA	.313	.320	.701	NA	15	15	
	330	H16-24	-.015	1.38	.307	.323	.677	12.0	2050	2829	dark vein
	330	F15-14	.063	1.36	.315	.325	.652	12.0	2270	3087	
	330	H16-01	.061	1.33	.316	.322	.718	12.0	3280	4362	
	1115	K14-15	.078	1.55	.318	.324	.704	12.0	3060	4743	
	1115	G24-23	.067	1.41	.315	.327	.653	12.0	2700	3807	
	1115	G24-10	.062	1.62	.316	.323	.713	12.0	3480	5638	
	2204	M15-20	.136	1.85	.324	.329	.617	11.0	6740	12469	
	2204	H11-06	.137	1.62	.324	.330	.620	11.0	7940	12863	
	2204	L26-05	.143	1.58	.325	.330	.642	11.0	9050	14299	

Table C3. (Contd.).

Site	Exp. Days	Briquette	Yellow	Corr. Factor	Color Coordinates			SO ₂ Ave. ppb	SO ₄ ppm Z Layer	SO ₄ Corr. ppm Z Layer	Remarks
					x	y	Y				
OH											
	NA	Standard	.023	NA	.311	.321	.705	NA	15	15	
	517	J36-20	.163	1.43	.331	.320	.640	21.5	5740	8208	
	517	L26-01	.077	1.29	.318	.323	.743	21.5	5230	6747	
	517	J36-27	.094	1.41	.320	.323	.703	21.5	5820	8206	
	1272	K13-18	.078	1.41	.317	.326	.707	21.9	7530	10617	
	1272	K14-21	.091	1.46	.319	.325	.726	21.9	6730	9826	
	1272	G36-09	.077	1.58	.317	.325	.688	21.9	4710	7442	
OHM											
	517	J36-18	.057	1.10	.315	.324	.723	21.5	3650	4015	dry
	517	I11-04	.058	1.10	.315	.322	.720	21.5	2460	4015	dry
	896	K14-19	.078	1.21	.318	.323	.742	22.4	4750	5748	dry
	896	G24-06	.072	1.33	.318	.322	.697	22.4	2980	3963	dry
	517	J36-19	.072	1.29	.317	.323	.713	21.5	4490	5792	sprayed
	517	K36-23	.062	1.23	.316	.322	.730	21.5	2980	3665	sprayed
	896	K14-03	.073	1.43	.317	.324	.729	22.4	3840	5491	sprayed
	896	G24-11	.046	1.25	.315	.321	.786	22.4	401	501	sprayed

Table C4. Averaged color coordinates, and SO₂ and SO₄ data; fresh limestone
(dark-spot data)

Site	Exp. Days	Briquette	Yellow	Corr. Factor	Color Coordinates			SO ₂ Ave. ppb	SO ₄ ppm Z Layer	SO ₄ Corr. ppm Z Layer	Remarks
					x	y	Y				
NY											
	NA	Standard	.243	NA	.337	.337	.461	NA	75	75	
	355	JL6-18	.245	1.15	.337	.336	.486	2.1	533	613	
	355	JL5-02	.244	1.17	.337	.337	.512	2.1	847	991	
	355	EL3-14	.216	1.23	.334	.334	.513	2.1	424	522	
	1112	GL6-14	.235	1.41	.336	.335	.505	2.8	1970	2778	
	1112	HL2-06	.322	1.52	.346	.344	.423	2.8	5620	8542	
	1112	HL8-24	.353	1.65	.350	.348	.423	2.8	5480	9042	
	2213	HL4-14	.316	6.05	.346	.341	.418	2.8	3330	20147	
	2213	IU5-17	.302	6.11	.343	.343	.437	2.8	3680	22485	
	2213	GL2-24	.341	8.47	.348	.347	.422	2.8	3520	29814	
DC											
	NA	Standard	.247	NA	.337	.337	.461	NA	75	75	
	330	JL6-16	.389	1.55	.354	.350	.350	12.0	17200	26660	
	330	EL3-07	.384	1.90	.355	.345	.357	12.0	8800	16720	
	330	JL5-18	.375	1.41	.353	.347	.360	12.0	13300	18753	
	1115	HL8-22	.438	1.27	.360	.359	.337	12.0	27800	35306	
	1115	GL6-19	.432	1.27	.359	.357	.337	12.0	20200	25654	
	1115	HL8-08	.431	1.38	.359	.356	.343	12.0	25500	35190	
	2204	HL4-24	.453	1.77	.362	.359	.300	11.0	27400	48498	
	2204	IU5-08	.428	1.41	.359	.355	.318	11.0	29800	42018	
	2204	GL7-17	.464	1.33	.362	.364	.293	11.0	39000	51870	

Table C4. (Contd.).

Site	Exp. Days	Briquette	Yellow	Corr. Factor	Color Coordinates			SO ₂ Ave. ppb	SO ₄ ppm Z Layer	SO ₄ Corr. ppm Z Layer	Remarks
					x	y	Y				
OH	NA	Standard	.247	NA	.337	.336	.461	NA	75	75	
	517	GL8-21	.387	1.10	.354	.352	.375	21.5	21400	23540	
	517	ML3-05	.398	1.13	.354	.355	.379	21.5	21100	23843	
	517	GL8-23	.366	1.10	.351	.349	.410	21.5	17900	19690	
	1272	GL3-12	.362	1.17	.351	.347	.412	21.9	28800	33696	
	1272	IL8-11	.381	1.19	.353	.351	.396	21.9	24900	29631	
	1272	HL2-Q8	.359	1.14	.353	.347	.401	21.9	22600	25764	
OHM	517	GL8-22	.262	1.09	.339	.339	.478	21.5	6970	7597	dry
	517	ML3-11	.312	1.13	.344	.344	.420	21.5	14900	16837	dry
	896	GL6-12	.340	1.01	.348	.346	.402	22.4	25500	25755	dry
	896	HL8-02	.353	1.01	.350	.347	.390	22.4	18400	18584	dry
	517	GL8-19	.280	1.13	.341	.341	.425	21.5	7940	8972	sprayed
	517	ML3-19	.381	2.30	.353	.350	.331	21.5	15200	34960	sprayed
	896	GL6-04	.339	1.14	.348	.345	.409	22.4	24100	27474	sprayed
	896	HL8-05	.314	1.41	.343	.350	.420	22.4	20100	28341	sprayed

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