Title

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Publication Date
2018-06-01

DOI
10.2172/1460338

Peer reviewed

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BERKELEY NATIONAL LABORATORY

Calibration of laboratory aging practice to replicate changes to roof albedo in a Chinese city

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June 2018

Heat Island Group
Energy Technologies Area

Report Number LBNL-2001005

Disclaimer

Acknowledgments

This work was supported by the Assistant Secretary for Energy Efficiency and Renewable Energy, Building Technologies Office, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. It was conducted in the framework of the US-China Clean Energy Research Center (CERC) for Building Energy Efficiency (BEE) between Lawrence Berkeley National Laboratory and the following Chinese research institutions: Research Institute of Standards and Norms (RISN) of the Ministry of Housing and Urban-Rural Development (MOHURD), Guangdong Provincial Academy of Building Research Group (GPABR), Shenzhen Institute of Building Research (Shenzhen IBR), Xiamen Academy of Building Research (XABR), Jiangsu Research Institute of Building Science (JRIBS), Sichuan Institute of Building Research (Sichuan IBR), China Building Materials Test & Certification Group (CTC), Shaanxi Provincial Academy of Building Research (SPABR), and Xinjiang Research Institute of Building Science (XRIBS).

Abstract

Highly reflective roofs can save building air conditioning energy, mitigate the urban heat island, and offset CO_2 emissions. A laboratory aging methodology was developed to simulate change in the solar reflectances of field-applied roof coatings induced by one year of natural exposure in Guangzhou (China). The approach compared the changes in solar reflectance spectra observed in the naturally exposed materials with those obtained following a three-stage laboratory procedure: conditioning, soiling, and weathering. The method was systematically modified over five trials by adjusting the key variables in laboratory aging: composition of the soiling mixture, deposition mass, and conditions of soiling application. Tests were performed using a small number of specimens. Simulating exposure in Guangzhou required a loading of soiling agents greater than that used to simulate US cites. In the final trial, the reduction in solar reflectance was reproduced to within an average of 0.02 for three of the six products, and to within an average of 0.06 for the remaining three products.

1 Introduction

Energy savings and environmental benefits associated with cool roofs are well documented. Highly reflective roofs can reduce the energy consumed by building air conditioning (Levinson et al. 2005; Levinson and Akbari, 2010; Gao et al., 2014), contribute to mitigation of the urban heat island (Rosenfeld et al. 1998), and offset of CO_2 emissions (Akbari et al. 2012). However, the potential benefits of cool roofs can be severely reduced by aging upon exposure in the environment (Sleiman et al. 2011). The initially high albedo of cool roofs can be reduced by soiling and weathering processes. By soiling we refer to the deposition of atmospheric constituents such as elemental (black) carbon, mineral dust particles, organic chemicals, and inorganic salts. In addition to these soiling constituents, significant microbiological soiling has been observed in humid climates, associated with the growth of fungi, bacteria, and other microorganisms. Weathering processes comprise physical and chemical damages to the material associated with contact with moisture, ultraviolet (UV) radiation, and temperature cycles (Sleiman et al., 2014).

Changes associated with soiling and weathering take place over several years of continuous exposure. Usually there is an initial period of rapid change during the first months of exposure, followed often by a slower rate of change that could be modulated by seasonality (e.g., increased atmospheric deposition during a dry season, or partial cleaning during a rainy season). However, some materials continue to experiment significant changes in solar reflectance even after the first year in the field, and may never reach a steady-state. Solar reflectance changes have been documented over the initial 3 years of exposure for a wide range of products used in the United States (US) (Sleiman et al. 2011; and Berdahl et al. 2008). Due to the slow nature of aging processes, the Cool Roof Rating Council (CRRC) and the US EPA Energy Star program have established an exposure period of 3 years prior to measuring aged (long-term) values of solar reflectance and thermal emittance (CRRC, 2017; US EPA, 2017).

The 3-year aging period creates a technical barrier for the development and commercialization of innovative, better performing cool roofing materials. It not only delays obtaining a product rating from the CRRC and/or the US EPA, but also makes it slower and more onerous for manufacturers to evaluate the performance of new prototypes. Therefore, LBNL has developed a laboratory aging methodology that replicates the effects of 3 years of natural exposure within a few days. This is achieved through a three-stage process in which a clean specimen is conditioned through exposure to UV light, heat, and moisture in a standard weathering apparatus; sprayed with a calibrated soiling mixture; and then weathered through a second round of exposure in the weathering apparatus (Sleiman et al. 2014). The methodology has been approved as ASTM Standard D7897-15 (ASTM, 2015), and is currently implemented by the US CRRC in its Rapid Ratings Program, which lets participating manufacturers obtain interim laboratory-aged ratings until 3-year field-exposed ratings are available (CRRC, 2017).

The aging methodology developed by LBNL applies only to products aged in the US, as soiling mixtures had been calibrated to reproduce reflectance changes observed in the three CRRC-specified aging sites of Phoenix (AZ, hot and dry), Miami (FL, hot and humid) and Cleveland (OH, a temperate weather with more polluted atmosphere). A fourth soiling mixture was calibrated reproduce the three-site average reflectance change, which is the quantity used for rating. Such calibration was carried out by adjusting the composition of the soiling mixture and the spraying regime to reproduce the 3-year aged solar spectral reflectances of a variety of US roof products. While most soiling constituents can be found in every climate, we observed that the spectra of aged materials could be reproduced by adjusting the composition of the mixture.

Black carbon and organic matter are major components of soot particles emitted during the combustion of fossil fuel and biomass. Black carbon absorbs strongly solar radiation across the entire solar spectrum (Kirchstetter and Thatcher, 2012). By contrast, particulate organic matter absorbs sunlight mostly in the visible region of the spectrum (Andrae and Geleneser, 2006). It contains highly polymerized humic-like substances that are predominantly brown in color. Mineral dust is a relatively weak absorber (Kinne et al. 2003). Finally, inorganic salts do not contribute significantly to the reflectance spectrum, but can have a major impact on how other constituents suspend in water and spread onto surfaces (Covert et al. 1972). The amount of soiling material present on surfaces depends on transport by wind, dissolution in water, and/or runoff.

Microbial growth can be significant in humid climates, sometimes even becoming the main soiling constituent. Microbial soiling may comprise lichen, algae, fungi and/or bacteria. As colonies die over time, remaining melanins and polysaccharides become persistent constituents of microbial soiling due to their resistance to photooxidation and other degradation processes. The complexity and composition of microbial colonies are site-specific and depend on the nature of the surface material, rainfall, and relative humidity (Cheng et al., 2011).

While the final outcome of the laboratory aging methodology developed by LBNL is specific to the US, the same approach can be applied elsewhere, as long as a good set of naturally aged materials is available to perform the calibration. For example, a team in Italy is currently developing an aging procedure to reproduce conditions in Europe (Paolini et al. 2016). Here, we describe initial efforts carried out to calibrate LBNL's laboratory aging practice for roofing materials to replicate changes in solar reflectance of samples exposed in Guangzhou, China.

2 Exposure of specimens in Guangzhou (China)

2.1. Background

Soiling results from the deposition of atmospheric particulate matter and growth of microorganisms. Weathering of materials occurs with exposure to water, sunlight, and temperature change. Both phenomena are site-specific and should be characterized by exposing product specimens in natural conditions, to understand how the solar reflectances of cool materials change over time. In December 2014, Changqing LIN from the Research Institute of Standards and Norms (RISN) of Ministry of Urban-Rural Development (MOHURD) initiated natural exposure trials of cool roof coatings in China with the participation of eight Chinese research institutions and five roofing manufacturers. The nine exposure sites span most of the major climate zones in China (see Table 1 and Figure 1).

Table 1. Research institutions testing materials in each climate zone.

City	Province	Responsible research institutions
Hot summer warm winter zone		
Guangzhou	Guangdong Province	Guangdong Provincial Academy of Building Research Group (GPABR)
Shenzhen	Guangdong Province	Shenzhen Institute of Building Research (Shenzhen IBR)
Xiamen	Fujian Province	Xiamen Academy of Building Research (XABR)
Hot summer cold winter zone		
Changzhou	Jiangsu Province	Jiangsu Research Institute of Building Science (JRIBS)
Shanghai Municipality		Jiangsu Research Institute of Building Science (JRIBS)
Chengdu	Sichuan Province	Sichuan Institute of Building Research (Sichuan IBR)
Cold and severe cold zones		
Beijing Municipality		China Building Materials Test & Certification Group (CTC)
Xi’an	Shaanxi Province	Shaanxi Provincial Academy of Building Research (SPABR)
Ürümqi	Xinjiang Province	Xinjiang Research Institute of Building Science (XRIBS)

Figure 1. Research institutes, roofing manufacturers, and test sites in natural exposure study.

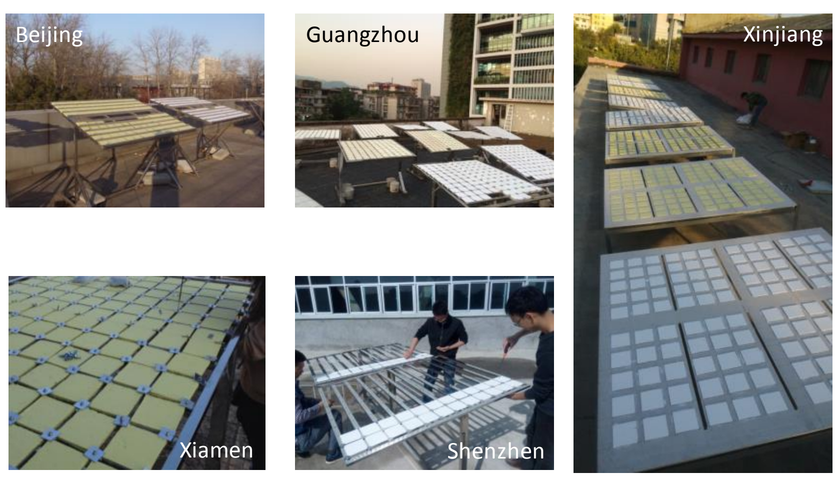


Figure 2. Images of exposure racks at five of the nine exposure sites.

2.2 Methodology

The natural exposure study is scheduled to run for five years (2014 – 2019). Product specimens are exposed in racks inclined 2% or 20% from horizontal, facing south. Each product is a yellow or white roof coating, and is applied to two substrates: a concrete tile, and an aluminum panel. Each rack has 120 specimens of each product (2 substrates × 4 retrievals/year × 5 years × 3 replicates = 120 specimens) at one slope (Figure 2). Three specimens will be retrieved every three months in years 1 - 3, and every six months in years 4 - 5. The solar reflectance, thermal emittance, and color of each retrieved specimen are measured and recorded to assess how those coatings age in natural conditions.

Exposure in Guangzhou is carried out by the Guangdong Provincial Academy of Building Research (GPABR). Their responsibilities include:

1. Prepare, expose, retrieve and measure specimens
2. Calibrate testing equipment
3. Participate in data collection and analysis

A complete description of instrumentation and measurement protocols used by GPABR can be found in Appendix A of Levinson et al. 2017. In summary, a solar spectrophotometer was used to measure solar reflectance and a portable emissometer was used to measure thermal emittance. Solar ("Sol"; 300 nm – 2500 nm), ultraviolet ("UV"; 300 nm – 400 nm), visible ("Vis"; 400 nm – 700 nm), and near-infrared ("NIR"; 700 nm – 2500 nm) reflectances were calculated by weighting spectral reflectance with an air mass 1 global horizontal (AM1GH) solar irradiance (Levinson et al. 2010a,b).

The products exposed are coded CG-a, CG-b, DC-a, DC-b and SC-a, where "CG", "DC", and "SC" are manufacturer labels assigned by RISN, and "a" and "b" indicates colors white and yellow, respectively. Specimens were prepared as follows:

- Before making the specimen, the concrete tile substrate was dried and cleaned; there was no rust on top of aluminum alloy base board.
- Coating was applied with roller or sprayer, 12-13 L/m².
- Applied primer once, and solar reflective thermal insulation coating twice. (Huarun coating for dirt resistance is applied twice.)
- Applied the coating again after 24 hours.
- Stored finished specimen indoor for 14 days, or stored specimen indoor for 4 days with 2 additional days in 40 °C oven.
- Applied 2 layers of primers. Dry film ≥ 60 µm, wet film ≥ 120 µm.

- Can choose to not dilute, or to dilute with 10% to 20% clean water.

A summary of coating properties is presented in Table 2.

Table 2. Participating manufacturers and coating application guidelines, prepared by RISN.

Participating manufacturers (coded)		**(DC)**	**(CG)**	**(SC)**
Coating 1	Color	White	White	White
	Initial solar reflectance	0.83	0.85	0.80
	Initial thermal emittance	0.85	0.86	-
	Product code	DC-a	CG-a	SC-a
Coating 2	Color	Light yellow	Light yellow	Light yellow
	Initial Solar reflectance	0.65	0.60	0.60
	Initial thermal emittance	0.75	0.86	-
	Product number	DC-b	CG-b	SC-b

2.3. Progress and issues

As of September 2016, the experiment has run for one and half years. Lawrence Berkeley National Laboratory (LBNL) has received 12 months (Dec 2014 to Dec 2015) of natural exposure experiment data from Guangzhou (Table 3), as well as the exposed specimens from that site. The variations with time of the solar reflectances of specimens from Guangzhou exposed at a 2% incline facing south are shown in Figure 3. The solar spectral reflectance of one of the coating products is plotted in Figure 4.

LIN from RISN reports that the natural exposure trials have experienced major data quality issues. For example, some naturally exposed specimens were washed in contravention of the protocol, and had to be excluded from the study. Out of the nine exposure sites, only three—Guangzhou, Beijing, and Chengdu—have yielded high quality data.

After examining data from Guangzhou, Beijing, and Chengdu and the physical condition of the exposed specimens from Guangzhou, the following major issues were identified by LBNL researchers.

1. All products from one of the manufacturers cracked shortly after the natural exposure trials began.

2. Spectrophotometer calibration errors are evident in spectral reflectance measurements performed by GPABR, as shown in Figure 5.
3. The color of a set of unexposed yellow specimens appeared to have faded white in a manner that the coating manufacturer considers atypical.

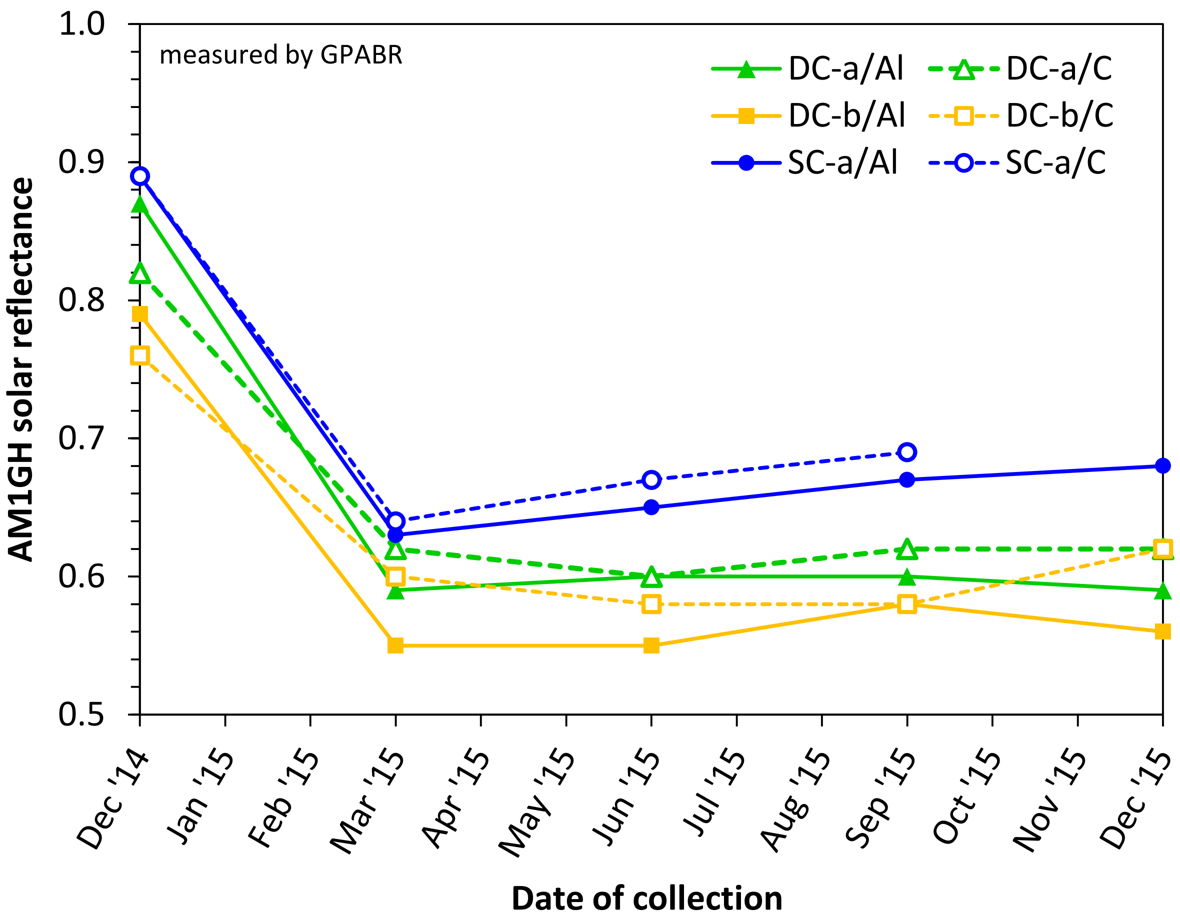


Figure 3. Variation with time of the solar reflectances of six specimens exposed in Guangzhou at a 2% slope facing south. Refer to Table 4 for an explanation of specimen nomenclature.

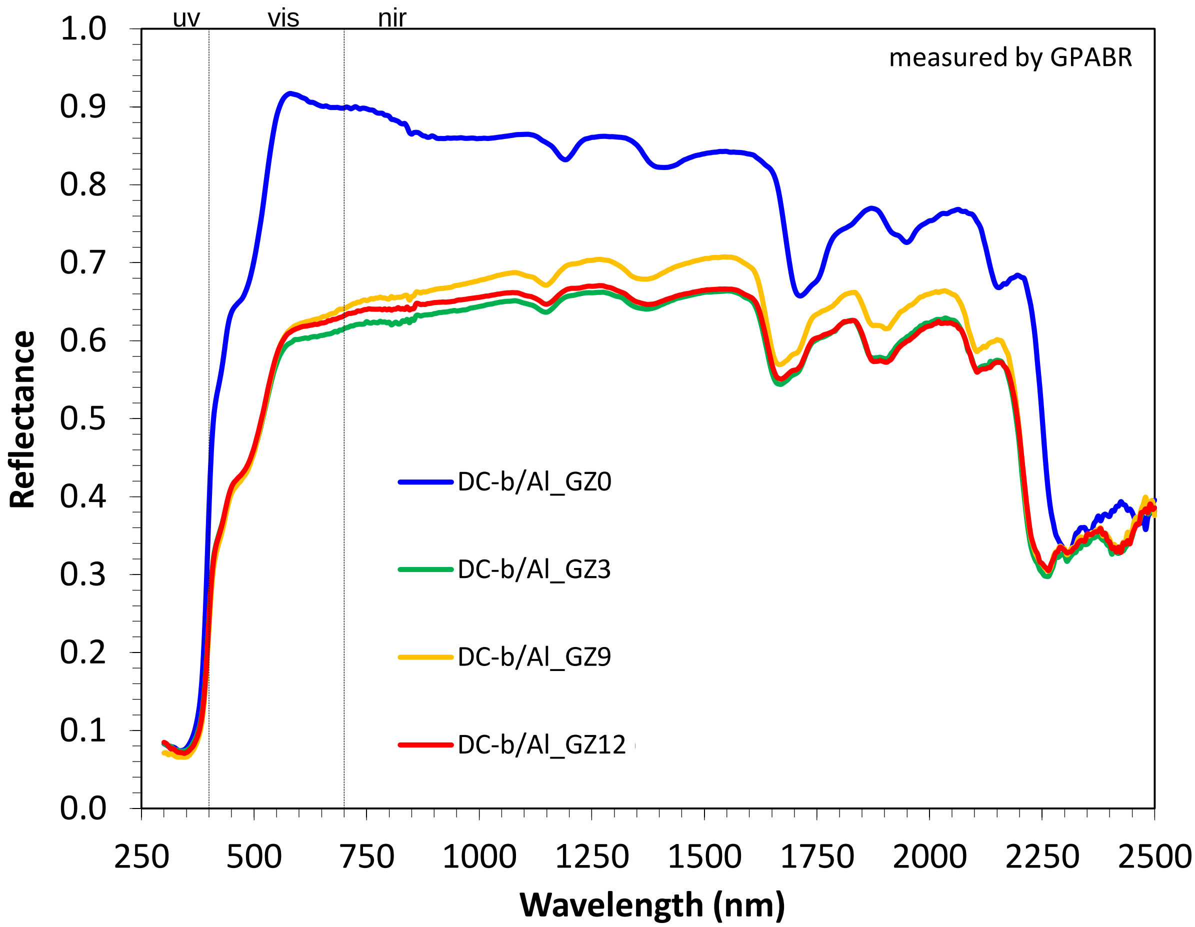


Figure 4. Solar spectral reflectances of one of the products exposed in Guangzhou ("GZ") at 0 months, 3 months, 9 months, and 12 months. The product is a yellow coating ("b") from manufacturer "DC" applied to a south-facing aluminum ("Al") panel tilted 2% from horizontal.

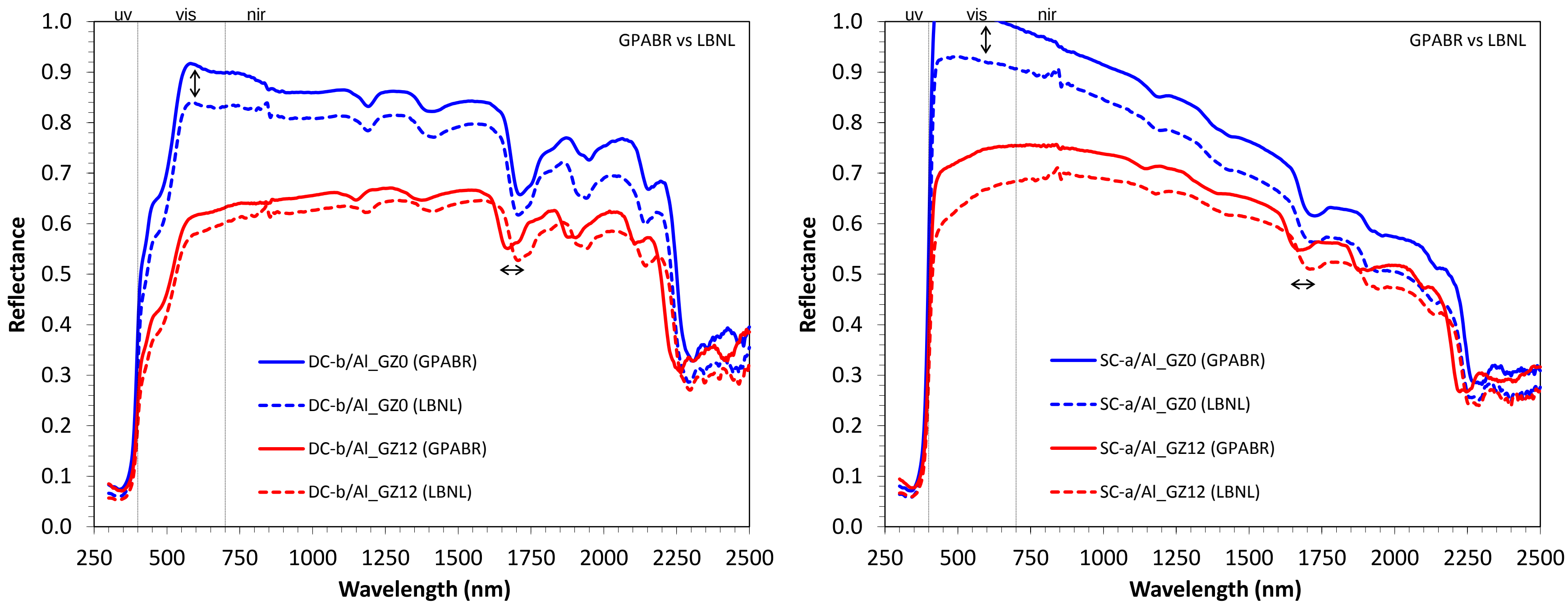


Figure 5. Comparison of GPABR and LBNL reflectance measurements of the same specimens, illustrating spectral differences that may result from instrument calibration errors at GPABR (e.g., use of a soiled reflectance standard, or mis-registration of wavelength). The two panels illustrate this effect for two different products. Note that the discontinuity at 860 nm in the LBNL measurements is an artifact of detector change at this wavelength.

Table 3. GPABR-reported initial and aged specimen solar reflectances and thermal emittances of specimens exposed in Guangzhou. The set of data reported here encompasses 3, 6, and 9 months of exposure for specimens tilted at 2% and 20% from horizontal.

Coating color	Product code	Product code (alt.)	Substrate (aluminum or concrete)	Solar reflectance							Thermal emittance							Notes
				Initial (Dec-2014)	3 months (Mar-2015)		6 months (Jun-2015)		9 months (Sep-2015)		Initial (Dec-2014)	3 months (Mar-2015)		6 months (Jun-2015)		9 months (Sep-2015)		
					2%	20%	2%	20%	2%	20%		2%	20%	2%	20%	2%	20%	
White	CG-a	I-a	Al	0.83	0.59	0.57	0.64	0.62	0.71	0.69	0.86	0.87	0.87	0.85	0.87	0.87	0.88	Cracking observed with natural exposure
			C	0.85	0.62	0.62	0.67	0.67	0.74	0.72	0.88	0.86	0.86	0.84	0.85	0.86	0.88	Cracking observed with natural exposure
Yellow	CG-b	I-b	Al	0.78	0.55	0.56	0.60	0.60	0.67	0.66	0.87	0.86	0.86	0.86	0.86	0.88	0.87	Cracking observed with natural exposure
			C	0.80	0.60	0.61	0.64	0.64	0.70	0.70	0.86	0.86	0.86	0.86	0.84	0.86	0.86	Cracking observed with natural exposure
White	DC-a	II-a	Al	0.87	0.59	0.58	0.60	0.57	0.60	0.58	0.90	0.89	0.88	0.87	0.87	0.88	0.87	
			C	0.82	0.62	0.61	0.60	0.58	0.62	0.63	0.88	0.86	0.88	0.86	0.87	0.88	0.89	
Yellow	DC-b	II-b	Al	0.79	0.55	0.53	0.55	0.55	0.58	0.56	0.89	0.89	0.89	0.87	0.88	0.89	0.88	
			C	0.76	0.60	0.59	0.58	0.56	0.58	0.62	0.86	0.87	0.87	0.86	0.87	0.86	0.88	
White	SC-a	III-a	Al	0.89	0.63	0.62	0.65	0.64	0.67	0.66	0.89	0.86	0.88	0.87	0.87	0.89	0.88	
			C	0.89	0.64	0.63	0.67	0.65	0.69	0.68	0.88	0.87	0.87	0.86	0.86	0.86	0.88	

3 Aging of specimens in the laboratory

A laboratory aging methodology was developed to simulate field exposure in Guangzhou, using a limited number of coating samples applied over two different substrates. The approach was comparable to our previous development of a laboratory aging methodology for the three US CRRC sites (in Florida, Arizona and Ohio), and their average (Sleiman et al. 2014). In order to refine the methodology, the field-exposed results were compared with the laboratory-aged results for each material. Five iterative trials were performed (labeled A-E), in which the method was systematically modified by adjusting the key variables in laboratory aging: composition of the soiling mixture, duration and conditions of soiling application. Conditions used in each trial were modified in order to improve the performance of the method.

3.1 Samples used for laboratory aging

Of the ten products field-exposed in Guangzhou, the six that did not exhibit cracking were used in the laboratory aging experiment. This set of six comprised three field-applied coatings (white SC-a, white DC-a, yellow DC-b) applied over two substrates (aluminum and concrete). In 2016, GPABR prepared a fresh batch of these six products (15 replicates per product, each 10 cm by 10 cm) for usage in LBNL's laboratory aging trials. In this section we describe these specimens as batch "L" in order to distinguish them from batch "GZ", the set of specimens prepared in 2014 and used in the Guangzhou field exposure experiment. It is important to note that this is a break from recommended procedure; good experimental practice calls for the specimens used in both field-exposure and laboratory aging trials to originate from the same batch. In this case, however, not enough specimens had been prepared in batch GZ to accommodate the needs of the laboratory aging trials.

In preparation for the laboratory aging experiment, GPABR shipped LBNL in 2016 the following materials:

- Guangzhou field-exposed specimens representing 0, 3, 6, 9, and 12 months of exposure at 2% and 20% slope (1 replicate per condition). Note that these specimens originated from batch GZ.
- Unexposed specimens from batch L (15 replicates each).

Our laboratory aging development work focused on replicating the changes in reflectance observed in these six products after 12 months of field-exposure in Guangzhou at 2% slope. All spectral reflectance measurements (250 – 2,500 nm @ 5 nm) presented in this section were made at LBNL in 2016 using a Perkin-Elmer Lambda 900 UV-vis-NIR spectrometer outfitted with a 150 mm Labsphere integrating sphere per ASTM E903-12: Standard Test Method for Solar Absorptance, Reflectance, and Transmittance of Materials Using Integrating Spheres (ASTM, 2012). Measurements are identified with both a product and condition code, separated by an underscore. The naming conventions are summarized in Table 4 and Table 5.

Table 4. Naming convention used to identify the specimens used in field exposure and laboratory aging experiments.

Specimen code	Coating product	Color	Substrate
SC-a/Al	SC-a	white	aluminum
SC-a/C	SC-a	white	concrete
DC-a/Al	DC-a	white	aluminum
DC-a/C	DC-a	white	concrete
DC-b/Al	DC-b	yellow	aluminum
DC-b/C	DC-b	yellow	concrete

Table 5. Naming convention used to describe the condition of specimens used in field exposure and laboratory aging experiments.

Condition code	Meaning
GZ0	Specimen prepared in 2014, unexposed (1 available)
GZ12	Specimen prepared in 2014, field-exposed 12 months in Guangzhou
L0	Specimen prepared in 2016, unexposed (15 replicates available)
A0	Specimen prepared in 2016 and used in lab aging trial A, initial condition
A1	Specimen prepared in 2016 and used in lab aging trial A, after step 1
A2	Specimen prepared in 2016 and used in lab aging trial A, after step 2
A3	Specimen prepared in 2016 and used in lab aging trial A, after step 3
BX-Y through EX-Y	Specimen prepared in 2016 and used in lab aging trials B through E X denotes condition (“0”: initial; “1”: after step 1; “2”: after step 2; “3”: after step 3) Y identifies specimen replicate where applicable, arbitrarily designated 1 – 3 (“1”: specimen 1; “2”: specimen 2, “3”: specimen 3) *Example: “E2-3” refers to replicate #3 of a trial E specimen at step 2 in the lab aging process*

Soon after the laboratory aging work began, it became apparent that specimens from batch L and batch GZ were not equivalent. Worryingly, we observed differences in visual appearance and spectral reflectance between unexposed batch L specimens (hereafter “L0”) and the unexposed batch GZ specimens used in Guangzhou (hereafter “GZ0”). The spectral differences for various specimens are shown in Figure 6 through Figure 8.

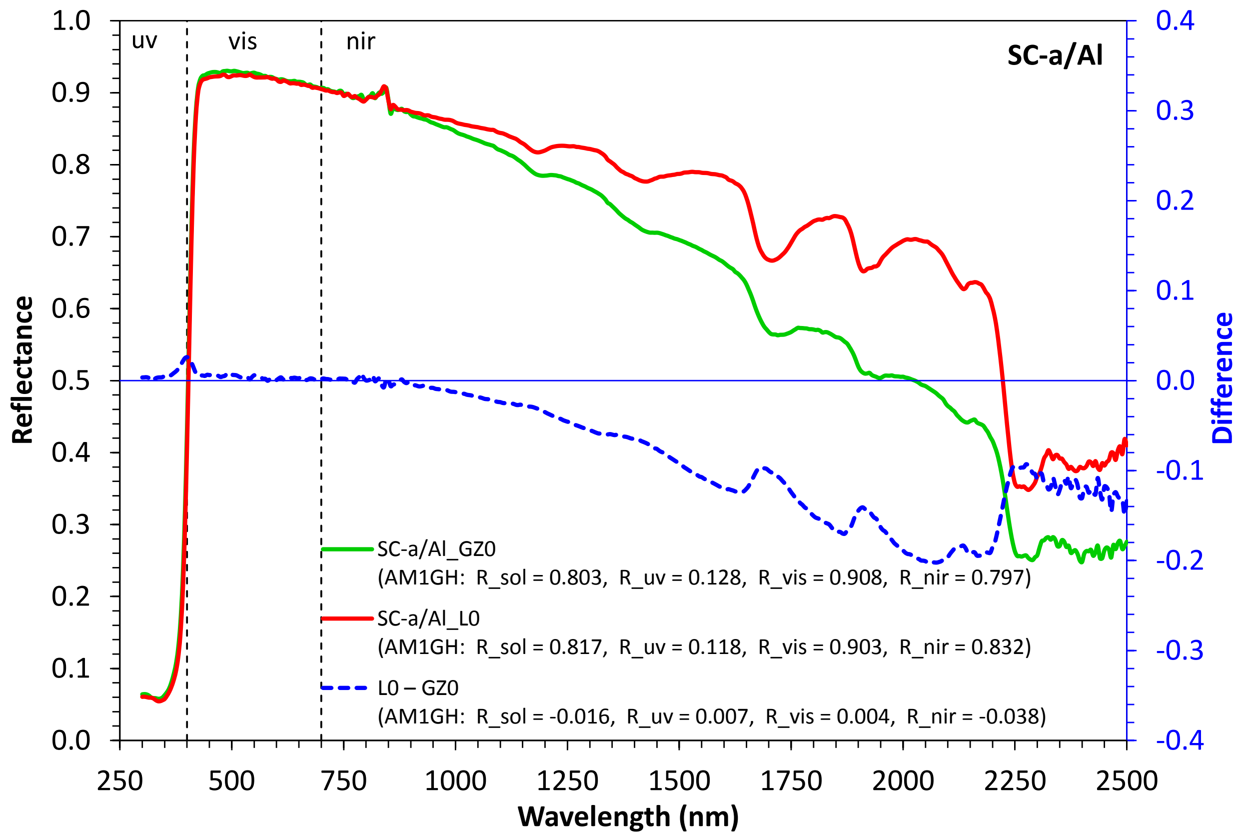


Figure 6. Solar spectral reflectances of one unexposed SC-a/Al sample from batch L (L0) and batch GZ (GZ0). Spectral reflectance difference (L0 – GZ0) is plotted on the right axis.

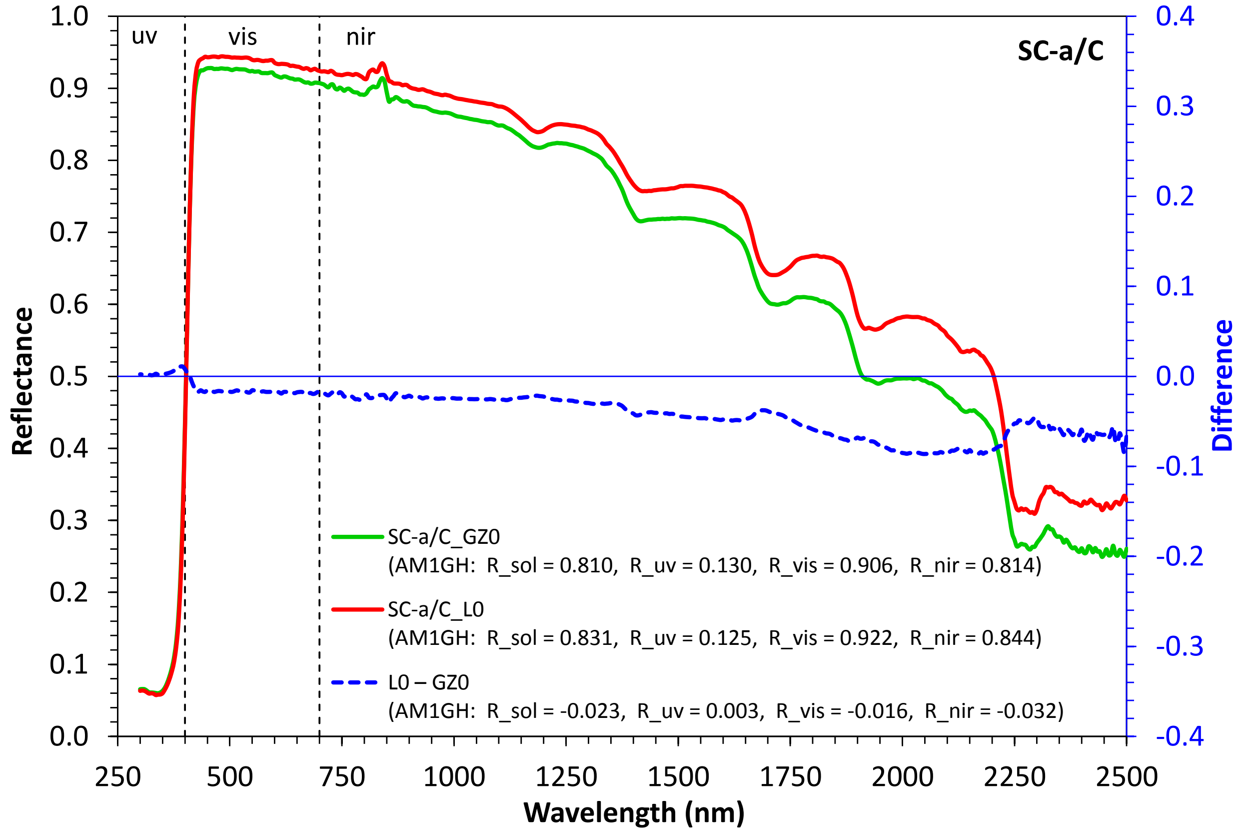


Figure 7. Solar spectral reflectances of one unexposed SC-a/C sample from batch L (L0) and batch GZ (GZ0). Spectral reflectance difference (L0 – GZ0) is plotted on the right axis.

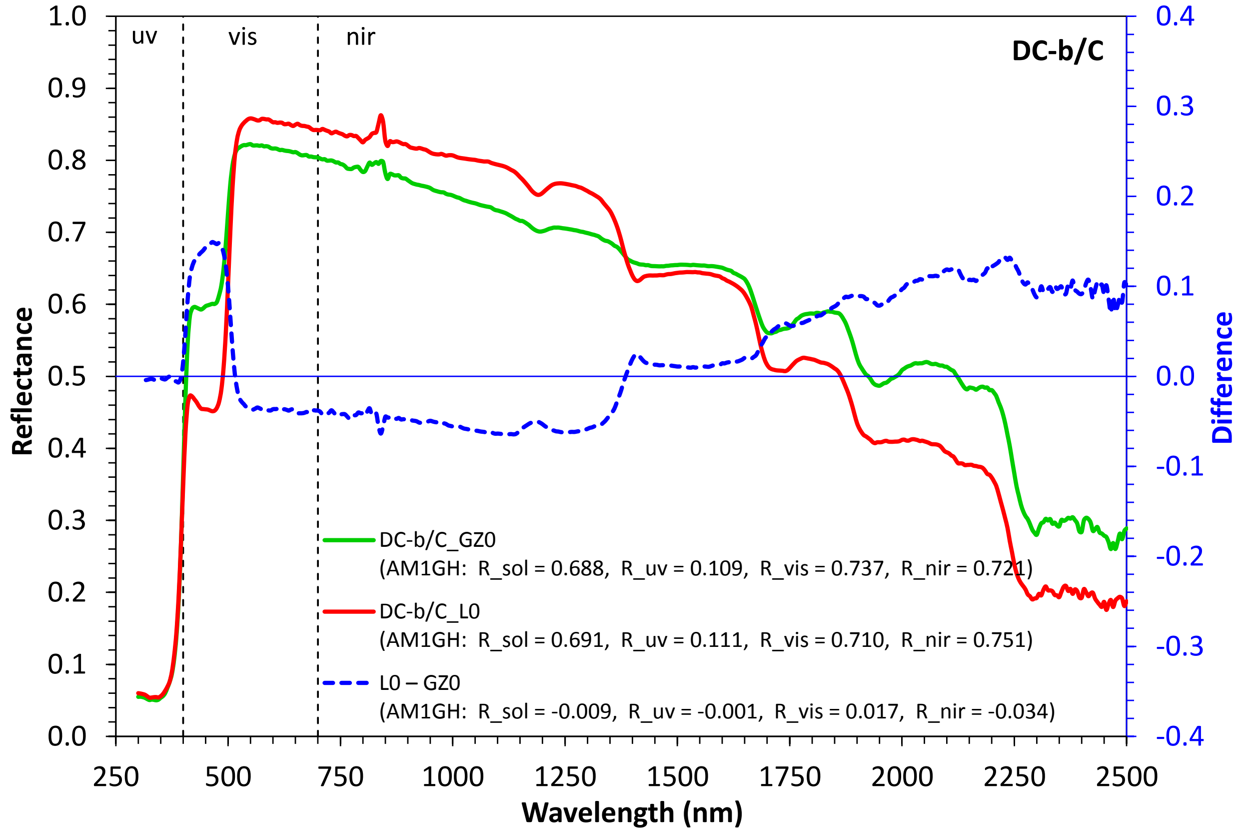


Figure 8. Solar spectral reflectances of one unexposed DC-b/C sample from batch L (L0) and batch GZ (GZ0). Spectral reflectance difference (L0 – GZ0) is plotted on the right axis.

Using an electronic coating thickness gauge, we measured the dry film coating thicknesses of the GZ0 and L0 specimens with an aluminum substrate (SC-a/Al, DC-a/Al, and DC-b/Al). The results are presented in Table 6, and show that the GZ0 coating thicknesses are more than twice that of the L0 specimens. Coatings typically achieve opacity at thicknesses greater than 100 microns. Hence, we suspect that the L0 specimens tested do not have opaquely thick coatings, which is consistent with some of the spectral differences we observed (Figure 9).

Table 6. Dry film thicknesses (µm) of coatings on unexposed specimens from batch GZ and batch L. Values represent the average of 5 spot measurements on one GZ0 and one L0 specimen.

Product/substrate	GZ0	L0
SC-a/Al	119	44
DC-a/Al	146	60
DC-b/Al	122	91

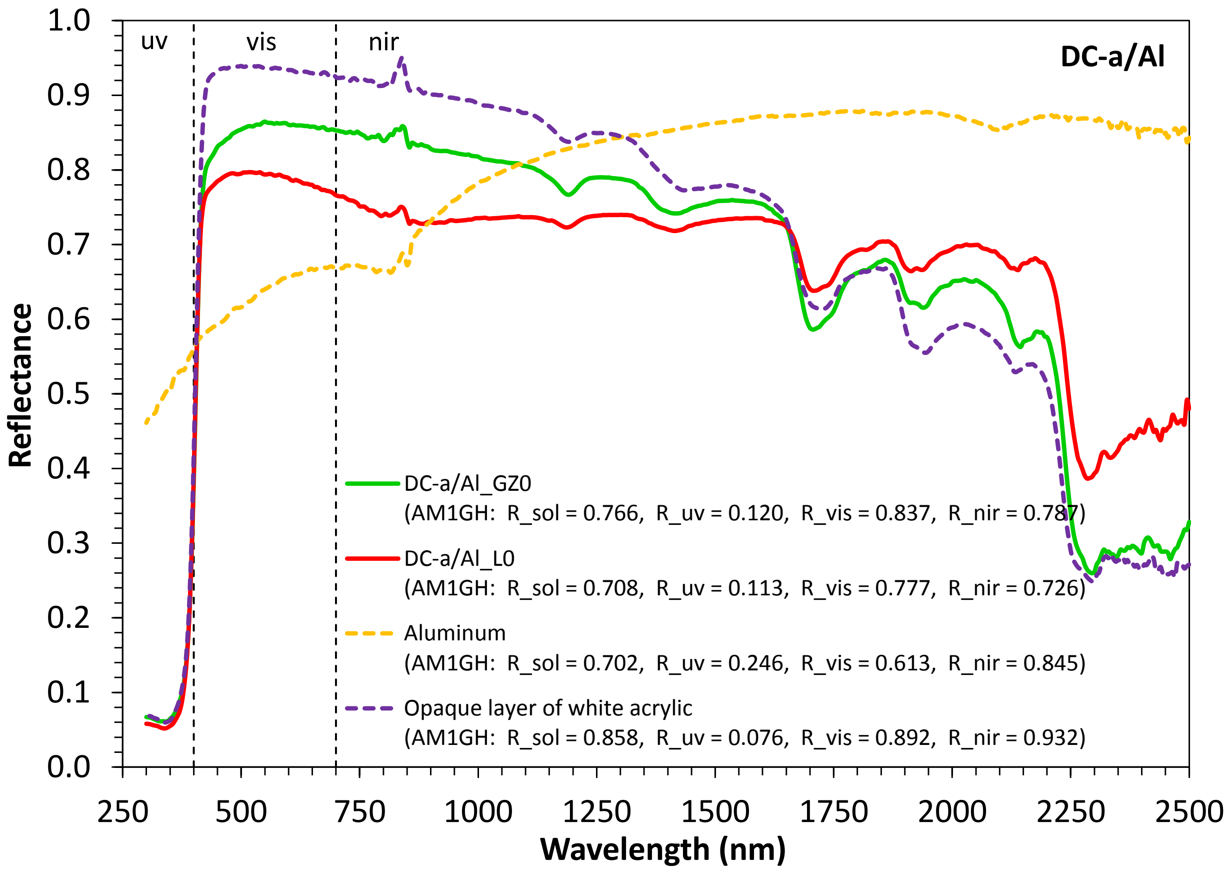


Figure 9. Reflectance spectra of one unexposed DC-a/Al sample from batch L (L0) and batch GZ (GZ0). Also shown are the spectral reflectances of bare aluminum and an opaque layer of white acrylic paint. These curves show that the spectral differences between L0 and GZ0 specimens are consistent with the differences in coating thickness.

In addition to discrepancies in initial spectral reflectance and coating thickness, the L0 and GZ0 specimens also differed in coating surface texture on a scale visible to the naked eye. It should be noted that the aluminum and concrete substrates are themselves quite smooth, so we do not believe the coating texture to be a result of any substrate roughness. We describe below our subjective assessments of the textural differences, referencing photographs shown in Figure 10.

- **SC-a/Al:** The surface of GZ0 is rough while that of L0 is very smooth.
- **SC-a/C**: The surface of GZ0 has a slightly rougher surface than does L0.
- **DC-a/Al:** The surface of GZ0 is slightly smoother than that of L0. Overall, GZ0 looks glossier than does L0.
- **DC-a/C:** L0 has a slightly smoother surface than GZ0.
- **DC-b/Al:** The surface of GZ0 is smooth while that of L0 is rougher. Overall, GZ0 looks glossier than does L0.
- **DC-b/C:** GZ0 has a significantly rougher surface than does L0. Overall, L0 looks glossier than does GZ0.

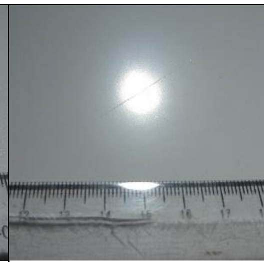
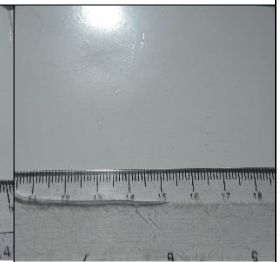
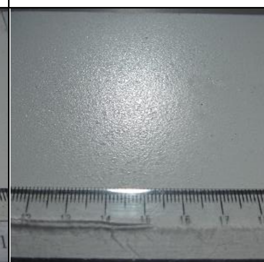
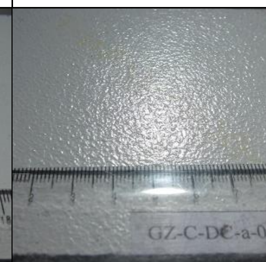
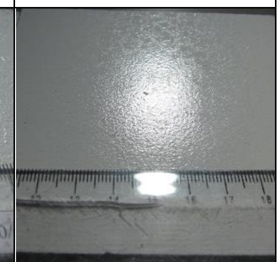
Sample	Al substrate		C substrate	
SC-a				
	GZ0	L0	GZ0	L0
DC-a				
	GZ0	L0	GZ0	L0
DC-b				
	GZ0	L0	GZ0	L0

Figure 10. Images of one unexposed sample from batch GZ (GZ0) and batch L (L0), showing differences in surface texture. These photographs were taken in a dark room using a single lamp as the light source, with both the specimens and camera mounted in a fixed position. A centimeter ruler is included in each photograph for scale.

Table 7 summarizes the information shown in Table 6 and Figures Figure 6 through Figure 10. It reports differences in coating thickness, solar reflectance, and surface texture between the L0 and GZ0 specimens. Note that, unless otherwise specified, all values reported in the table represent the measurements of one specimen. The column of L0 SR ranges is the one exception, and instead reports a range of values representing 3-6 specimens. It shows that even amongst replicate specimens of the same batch—in this case, batch L—differences in SR can be as high as 0.07.

Table 7. Summary of differences between L0 and GZ0 specimens.

Product	Color	Substrate	Coating thickness (µm)		AM1GH SR		Surface texture (subjective)	
			GZ0	L0	GZ0	L0 [a]	GZ0	L0
SC-a	white	aluminum	119	44	0.803	0.80–0.83	slightly rough	smooth
SC-a	white	concrete	-	-	0.810	0.80–0.84	slightly rough	smooth to slightly rough
DC-a	white	aluminum	146	60	0.766	0.67–0.74	smooth to slightly rough	slightly rough
DC-a	white	concrete	-	-	0.743	0.74–0.76	rough	slightly rough
DC-b	yellow	aluminum	122	91	0.708	0.67–0.69	smooth	slightly rough
DC-b	yellow	concrete	-	-	0.688	0.68–0.71	rough	smooth to slightly rough

[a] This column reports a value range, representing 3-6 L0 samples randomly chosen from the set of 15. In contrast, the GZ0 solar reflectance column represents measurements of just 1 sample, since GPABR did not ship LBNL any replicates.

In combination, these significant differences lead us to suspect that batch L and batch GZ may have been prepared using either different coating formulations or different coating preparation procedures. This invites the possibility that L0 specimens will not perform equivalently to GZ0 specimens when subjected to the same lab aging protocols. We therefore note that the laboratory aging protocol developed in this section can only serve as a proof-of-concept, rather than as a useful protocol that can be used to simulate 1 year of field-exposure in Guangzhou for a wide selection of roofing products.

For the reasons described previously, the development of a laboratory aging process for Guangzhou is less straightforward than our work on US-exposed samples (Sleiman et al. 2014). Because there are discrepancies in initial solar reflectance between the GZ0 and L0 specimens, it is not meaningful to focus protocol development on matching the solar reflectances of the GZ12 specimens. Instead, the success of a protocol is determined by whether it achieves the same reflectance loss (initial – lab aged) as 1 year of field-exposure in Guangzhou (GZ0 – GZ12).

The nomenclature used to identify the different lab aging tests is described in Table 5.

3.2 Data from field-exposed specimens

GPABR sent LBNL one specimen per product/condition. Photographs shown in Figure 11 to Figure 13 compare the L0, GZ0 and GZ12 samples.

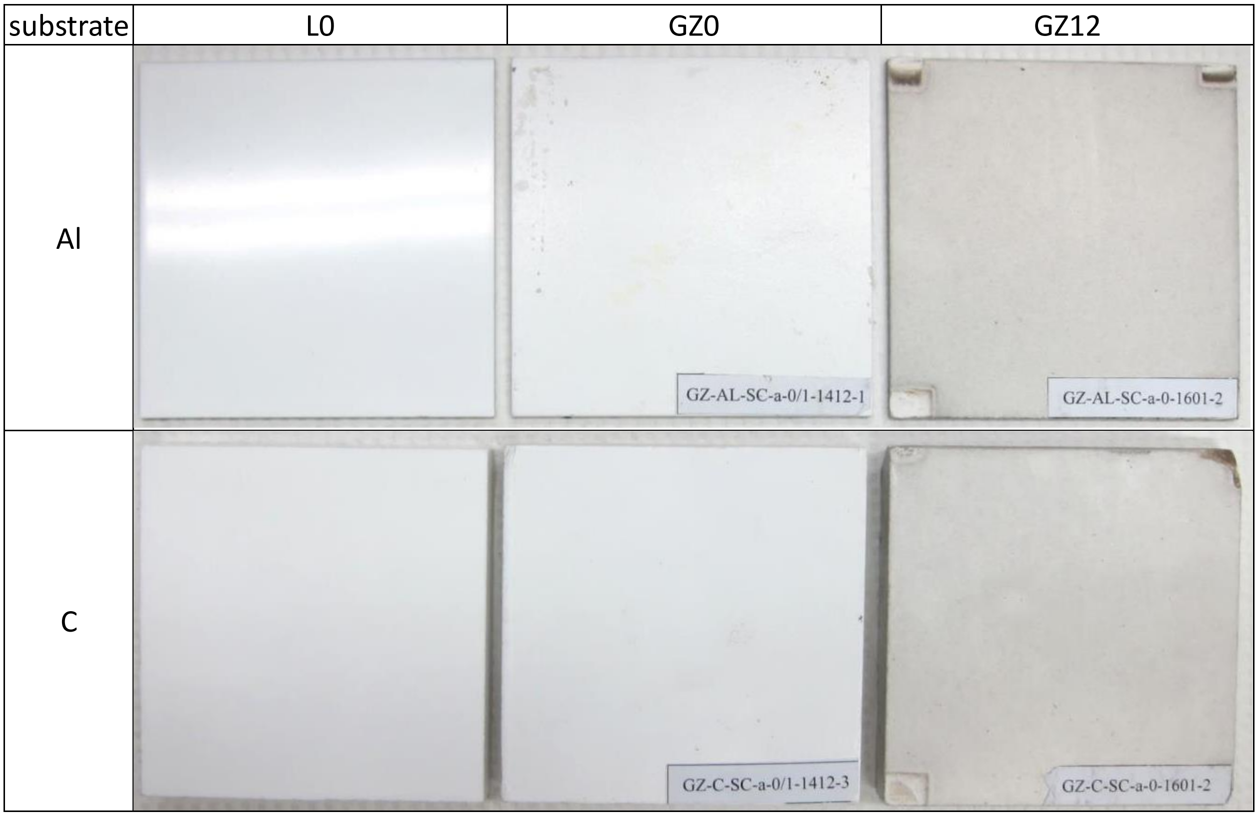


Figure 11. Images of coating product SC-a applied over aluminum (Al) and concrete (C) substrates. From left to right: batch L unexposed (L0), batch GZ unexposed (GZ0), and batch GZ exposed for 12 months in Guangzhou (GZ12).

substrate	L0	GZ0	GZ12
Al			
C			

Figure 12. Images of coating product DC-a applied over aluminum (Al) and concrete (C) substrates. From left to right: batch L unexposed (L0), batch GZ unexposed (GZ0), and batch GZ exposed for 12 months in Guangzhou (GZ12).

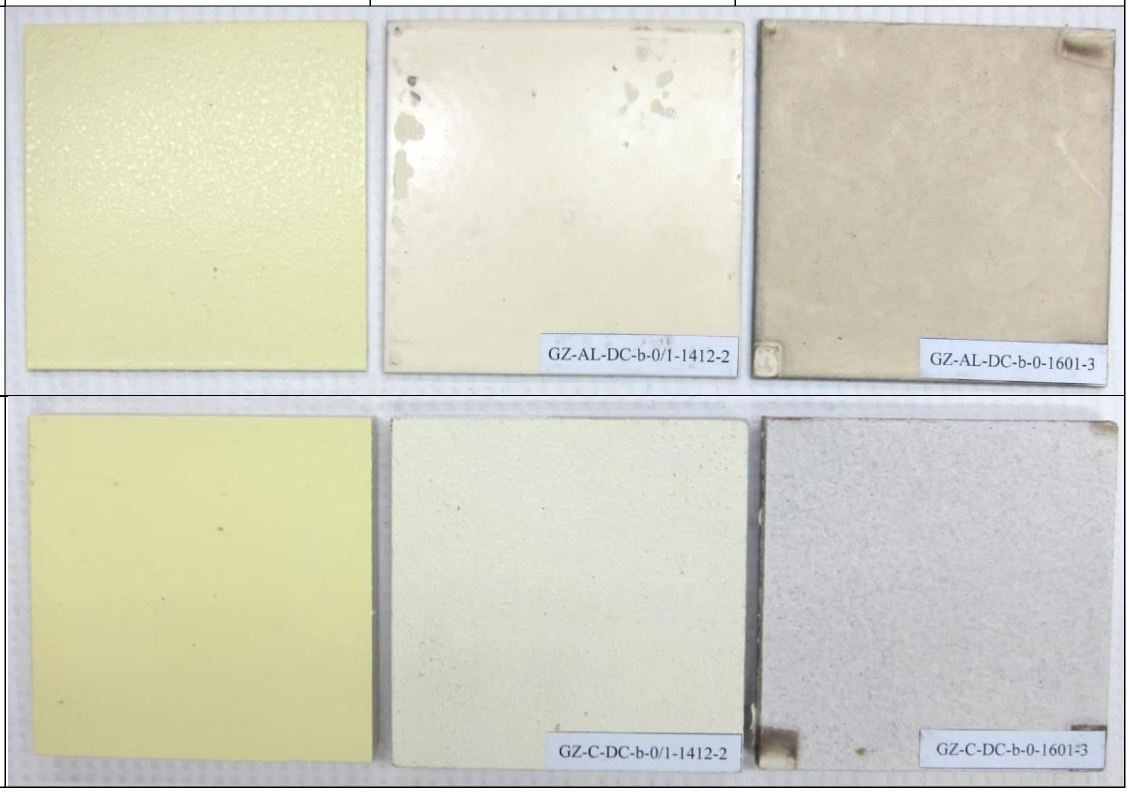
substrate	L0	GZ0	GZ12
Al			
C			

Figure 13. Images of coating product DC-b applied over aluminum (Al) and concrete (C) substrates. From left to right: batch L unexposed (L0), batch GZ unexposed (GZ0), and batch GZ exposed for 12 months in Guangzhou (GZ12).

Figure 14 presents the initial and aged spectral reflectances (averaged across three spots per specimen) for one of the products (SC-a/Al). Figure 15 and Table 8 summarize 12-month natural exposure results for all tested samples. The spectral differences between GZ0 and GZ12 for all products are presented in Figure 16 and Figure 17.

To guide the formulation of a lab aging procedure simulating 1 year in Guangzhou, we referenced past experiments of white field-applied coatings that had been lab-aged per ASTM D7897 (see Figure 18). Comparing the Guangzhou field-exposure results to these lab-aged results, we made the following observations and assumptions:

- We observed a greater reflectance loss in the GZ12 samples than in the lab-aged (ASTM D7897) field-applied coatings. Hence, we expected the Guangzhou version of the lab aging process to involve greater soiling deposition during Step 2.
- Interestingly, the GZ12 concrete specimens did not lose as much reflectance as their aluminum substrate counterparts. We wondered if this effect could be replicated in the laboratory aging trials.
- The spectral differences (initial – aged) in the GZ12 specimens (Figure 16 and Figure 17) showed higher visible reflectance loss than the ASTM D7897 lab-aged specimens (panel

(c) of Figure 18). This led us to believe the Guangzhou soiling mixture should contain a higher level of humic acid than the US average mixture.

- The field-applied coatings in past ASTM D7897 experiments did not undergo a significant change in reflectance in Step 3 (post-soiling weathering apparatus exposure). This led us to assume the six Guangzhou products would behave in a similar manner, and that the soiling deposition in Step 2 would be responsible for a bulk of the total reflectance loss during the lab aging procedure. We would later find this assumption to be erroneous.

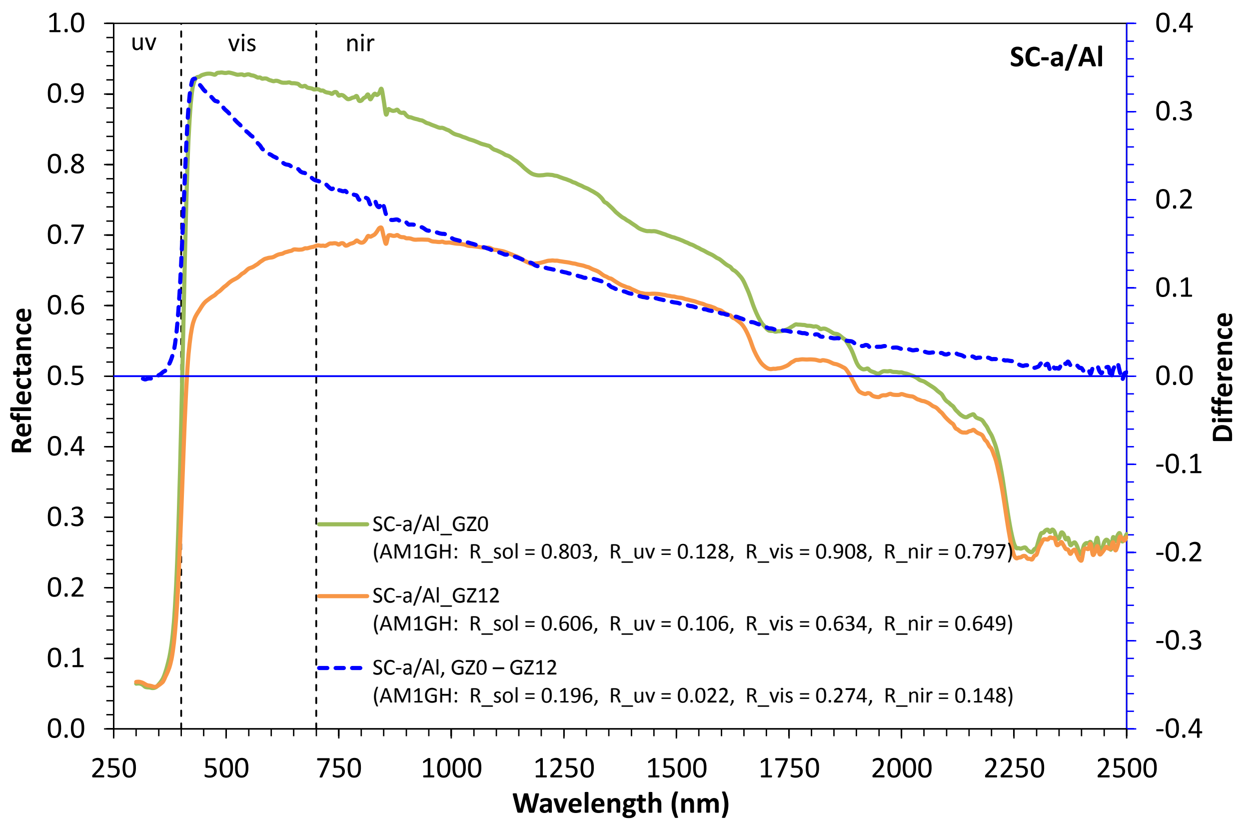


Figure 14. Spectral reflectances of SC-a/Al at t=0 (GZ0), exposed in Guangzhou at t=12 months (GZ12), and the corresponding spectral difference (GZ0 – GZ12).

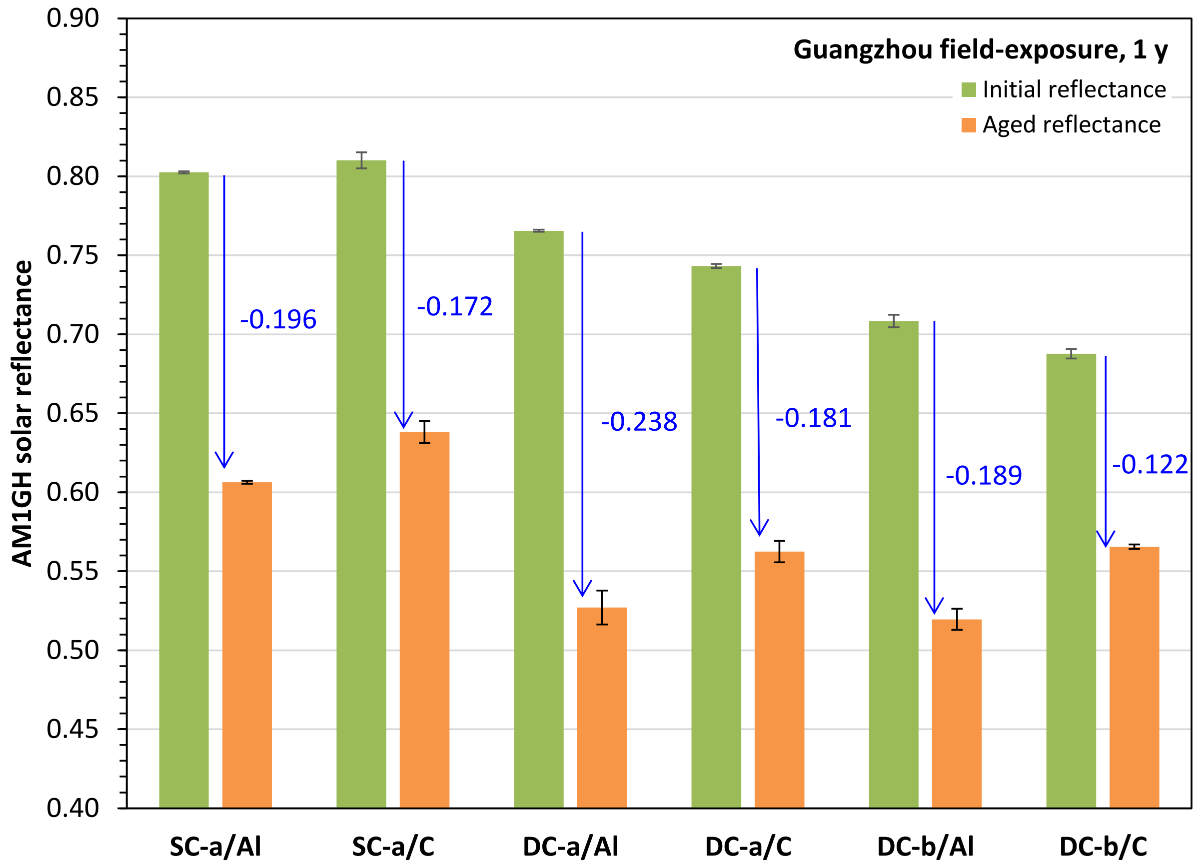


Figure 15. Solar reflectances of samples exposed in Guangzhou at t=0 (GZ0, initial) and t=12 months (GZ12, aged). Numbers next to arrows report absolute differences (GZ12 – GZ0). Error bars represent the standard deviation of the three spot measurements.

Table 8. Summary of solar, visible, and near-infrared reflectances measured in samples exposed in Guangzhou at t=0 (GZ0) and t=12 months (GZ12), and their corresponding absolute differences.

AM1GH:	**sol (300 nm – 2500 nm)**			**vis (400 nm – 700 nm)**			**nir (700 nm – 2500 nm)**		
Specimen	**GZ0**	**GZ12**	**GZ12-GZ0**	**GZ0**	**GZ12**	**GZ12-GZ0**	**GZ0**	**GZ12**	**GZ12-GZ0**
SC-a/Al	0.803	0.606	-0.196	0.908	0.634	-0.274	0.797	0.649	-0.148
SC-a/C	0.810	0.638	-0.172	0.906	0.664	-0.242	0.814	0.687	-0.127
DC-a/Al	0.766	0.527	-0.238	0.837	0.521	-0.317	0.787	0.592	-0.195
DC-a/C	0.743	0.562	-0.181	0.830	0.589	-0.241	0.748	0.598	-0.151
DC-b/Al	0.708	0.520	-0.189	0.709	0.484	-0.225	0.789	0.611	-0.178
DC-b/C	0.688	0.566	-0.122	0.737	0.595	-0.142	0.721	0.598	-0.123

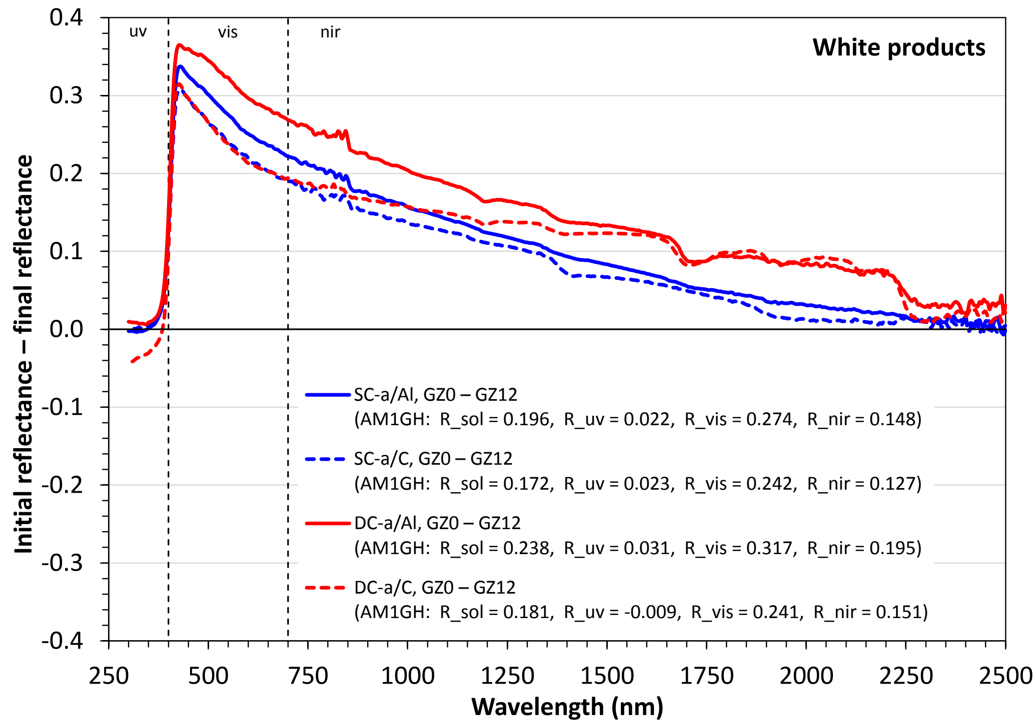


Figure 16. Spectral differences observed after 1 year of field-exposure in Guangzhou for white products SC-a and DC-a.

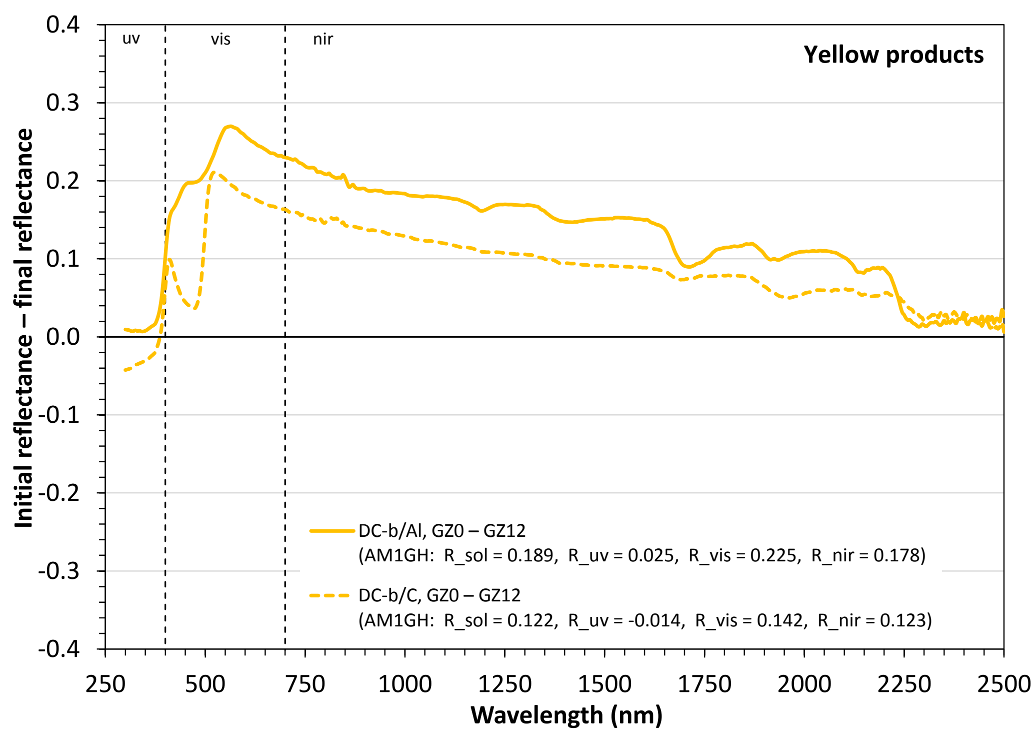


Figure 17. Spectral differences observed after 1 year of field-exposure in Guangzhou for yellow product DC-b.

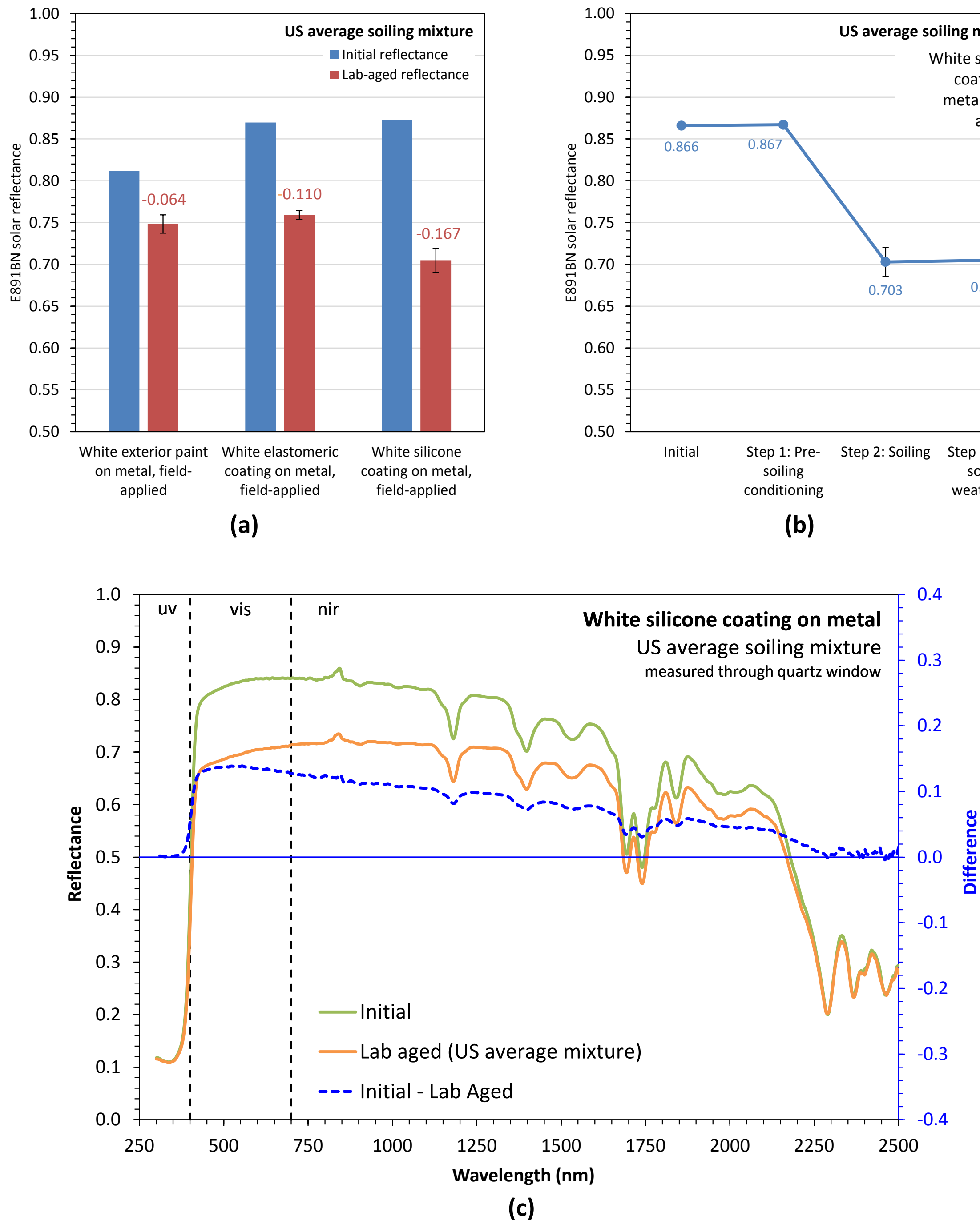


Figure 18. Past experimental data of various commercial white field-applied products lab-aged per ASTM D7897. Panel (a) shows the initial and lab-aged solar reflectances for 3 products. Panels (b) and (c) focus on one of the products, showing the reflectance changes at each step of the process and the initial – aged spectral differences, respectively.

3.3 Laboratory aging tests

In total, we performed five iterative laboratory aging trials as described in the following subsections, incrementally optimizing the Step 2 soiling mixture formulation and spraying protocol until the lab-aged results reproduced the reflectance losses observed in 12 months of aging in Guangzhou. In order to conserve the limited number of L0 specimens (15 replicates per product) available to work with, each trial utilized just one or two replicates for each of the six products. We note here that in an ideal situation, three or more specimens of each product would have been tested in each trial, to ensure results had statistical significance.

Initial trials focused only on the four white products. The stark differences in color suggested that the yellow L0 specimens either had different coating formulations than their GZ0 counterparts, or were otherwise defective. In order to achieve the desired soiling agent loadings during Step 2, we varied two parameters:

a) the concentration of the soiling mixture, or

b) the spray duration/wet mass deposited.

There was no clear argument for one over the other, so both were arbitrarily varied individually or simultaneously, based on what was most convenient. However, tests always remained within the following boundaries:

- The spray duration chosen did not produce spots more than 5 mm in size on the reference specimen. We found the small spot sizes specified in ASTM D7897 difficult to achieve with the amount of soiling deposition needed to reach the target lab-aged reflectance losses.
- The concentration of large particles in the soiling mixture was not high enough to induce frequent clogging in the spray nozzle.

The soiling mixtures and spray protocols tested in each of the five trials are summarized in Table 9 and Figure 19. Note that soiling spray application is described in terms of wet mass (of soiling mixture) and dry mass (of each soiling agent) deposited on a reference specimen during spray calibration.

Table 9. Composition and application of soiling mixtures used in Trials A – E, including soiling agent mass fractions, soiling agent concentrations, wet soiling mass deposited on reference specimen, and dry soiling mass coverage on reference specimen.

Trial	**US average (ASTM D7897)**	**Trial A**	**Trial B**	**Trial C**	**Trial D**	**Trial E**
Step 2 soiling mixture composition						
Dust (mass %)	47	13	6	4	3	3
Salts (mass %)	20	5	2	2	1	1
POM (mass %)	28	75	88	92	94	94
Soot (mass %)	5	7	3	2	2	1
Dust concentration (g L^{-1})	0.575	0.161	0.322	0.322	0.322	0.410
Salts concentration (g L^{-1})	0.250	0.062	0.124	0.124	0.124	0.157
POM concentration (g L^{-1})	0.35	0.93	4.6	7.6	10.6	12.5
Soot concentration (g L^{-1})	0.065	0.087	0.174	0.174	0.174	0.177
Total concentration (g L^{-1})	**1.24**	**1.24**	**5.22**	**8.22**	**11.22**	**13.24**
Step 2 soiling mixture deposition on reference specimen						
Wet mass (g)	0.9 ± 0.1	1.6 ± 0.1	1.2 ± 0.1	1.2 ± 0.1	1.4 ± 0.1	1.15 ± 0.1
Dry dust coverage (μg cm^{-2})	5.2	2.6	3.8	3.8	4.5	4.7
Dry salts coverage (μg cm^{-2})	2.2	1.0	1.5	1.5	1.7	1.8
Dry POM coverage (μg cm^{-2})	3.1	14.9	54.9	90.5	146.8	141.9
Dry soot coverage (μg cm^{-2})	0.6	1.4	2.1	2.1	2.4	2.0
Total coverage (μg cm^{-2})	**11**	**20**	**62**	**98**	**155**	**150**

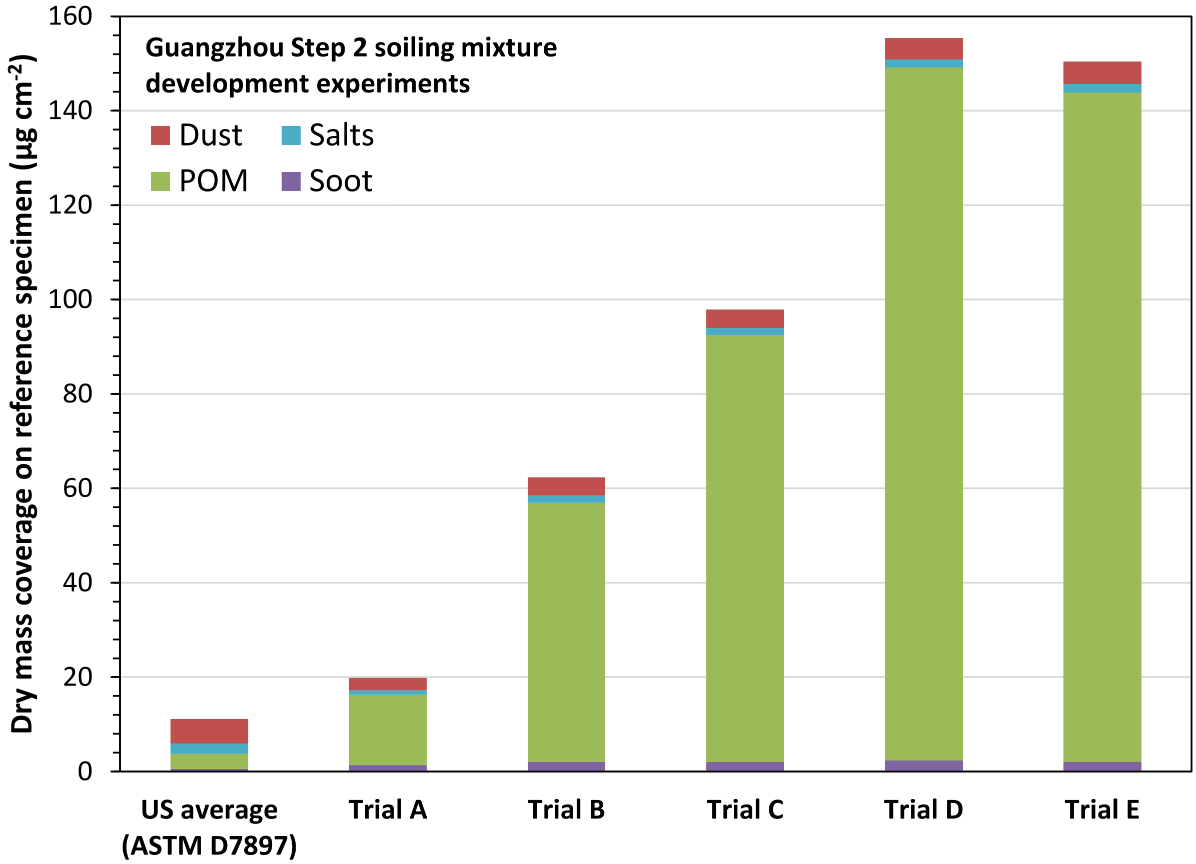


Figure 19. Step 2 soiling mixture deposition in each of Trials A – E, reported in terms of dry soiling mass coverage deposited on the reference specimens used to calibrate spray.

3.3.1 Trial A

For the initial Trial A, the total soiling agent concentration was arbitrarily kept at a level equivalent to the US average mixture (1.24 g/L). Based on what we learned from comparing past ASTM D7897 lab aging data to the Guangzhou field exposure results, it was decided that the soiling agent ratios for this trial would be: 13% dust, 5% salts, 75% POM, and 7% soot—essentially the US average mixture with POM and soot increased, and dust and salts decreased accordingly. Two specimens per product were tested in Trial A. The resultant spectral changes are illustrated in Figure 20. The solar reflectance, visible reflectance and near-infrared reflectance losses are presented in Figure 21. We observed that:

- Only one out of four products (DC-a/Al) exhibited any consistency in results between the two replicates.
- Only one replicate of one product (SC-a/Al replicate #1) achieved close to the target reflectance loss.
- Visual inspection suggested the surprisingly low reflectance losses arose from some of the soiling deposition in Step 2 washing off during the weathering apparatus exposure of Step 3.

In conclusion, we assumed that the one successful result was probably an outlier, and that in subsequent trials both the POM and soot loading should be increased.

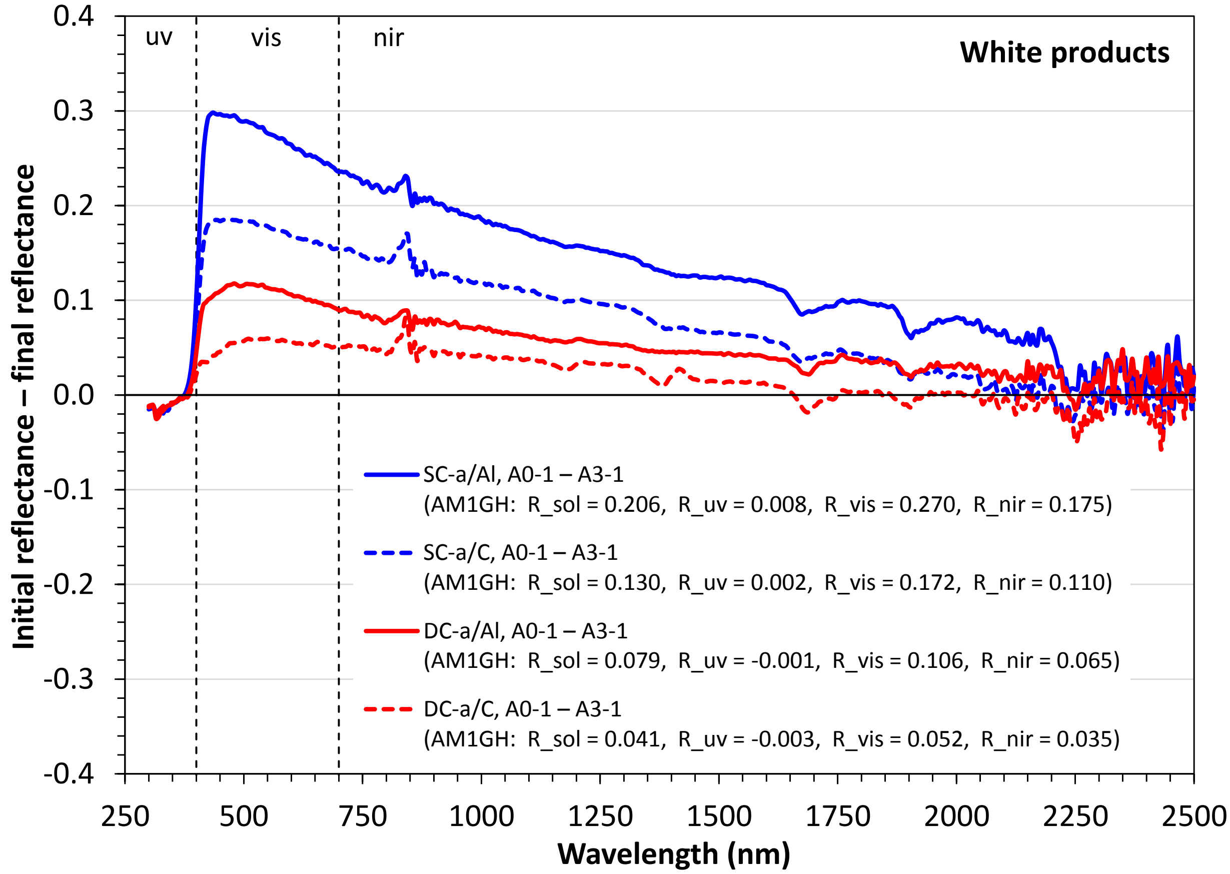


Figure 20. Spectral differences (lab-aged – initial) observed in Trial A.

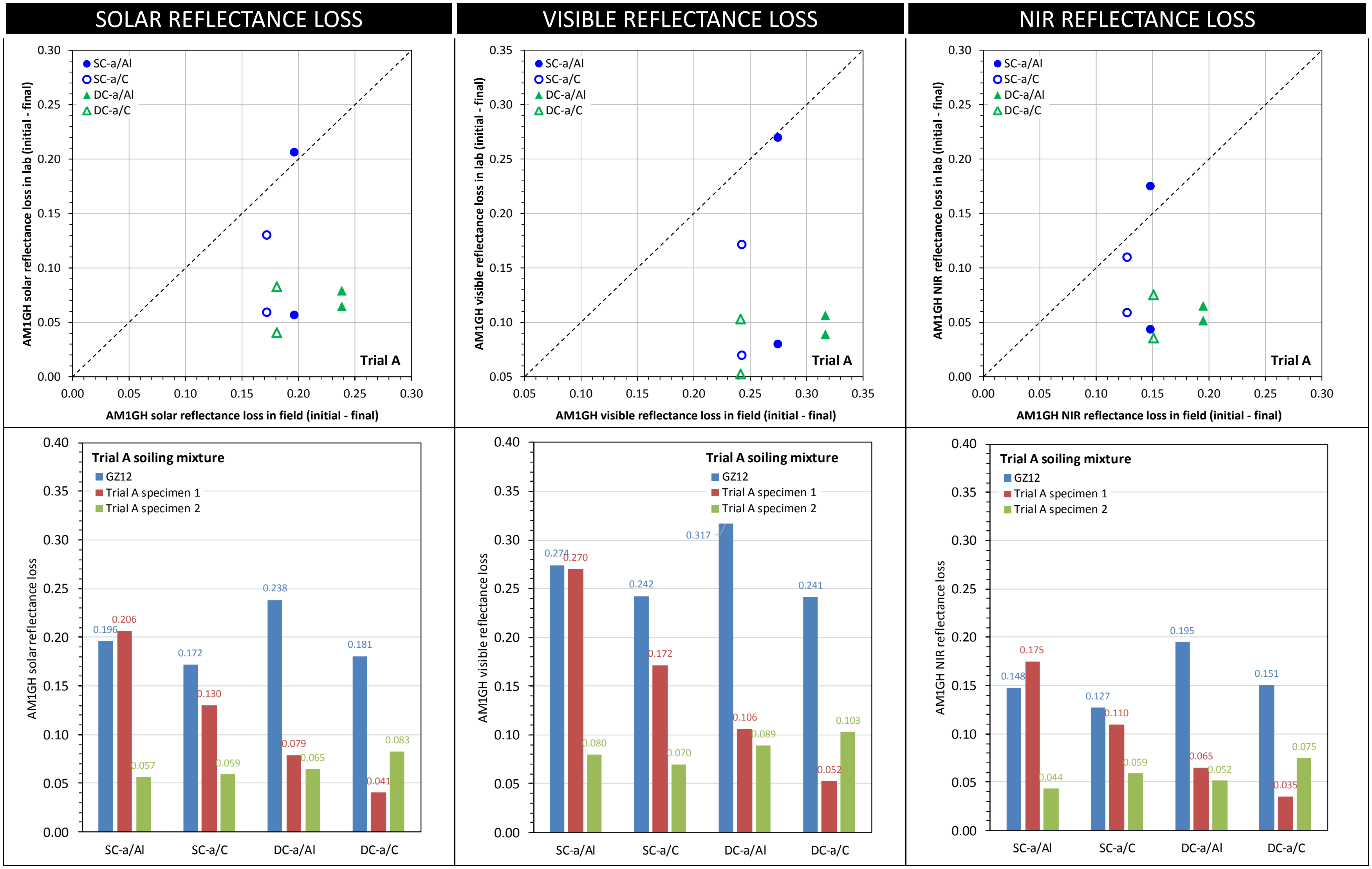


Figure 21. Comparison of broadband reflectance losses observed in Trial A and in Guangzhou field-exposure. The top row displays this information in a scatter plot; the bottom row displays this information in the form of a column plot.

3.3.2 Trial B

Given that the results of Trial A proved wrong our original assumption there no cleaning would occur during Step 3, we decided to increase the POM and soot loading in Trial B. This trial tested only one specimen each of the four white products. The changes in spectral reflectance at each step of the trial are shown in Figure 22, and the comparison of broadband reflectance losses between Guangzhou field-exposure and Trial B are shown in Figure 23.

Overall, the solar reflectance losses in the four tested products were still too low. In comparing the visible and near-infrared reflectance losses of the field-exposed and lab-aged specimens, it was determined that subsequent trials should further increase the Step 2 loading of POM, and slightly increase the Step 2 loading of soot. We also noted that DC-a/Al sample experienced significantly less cleaning in Step 3 than all other specimens.

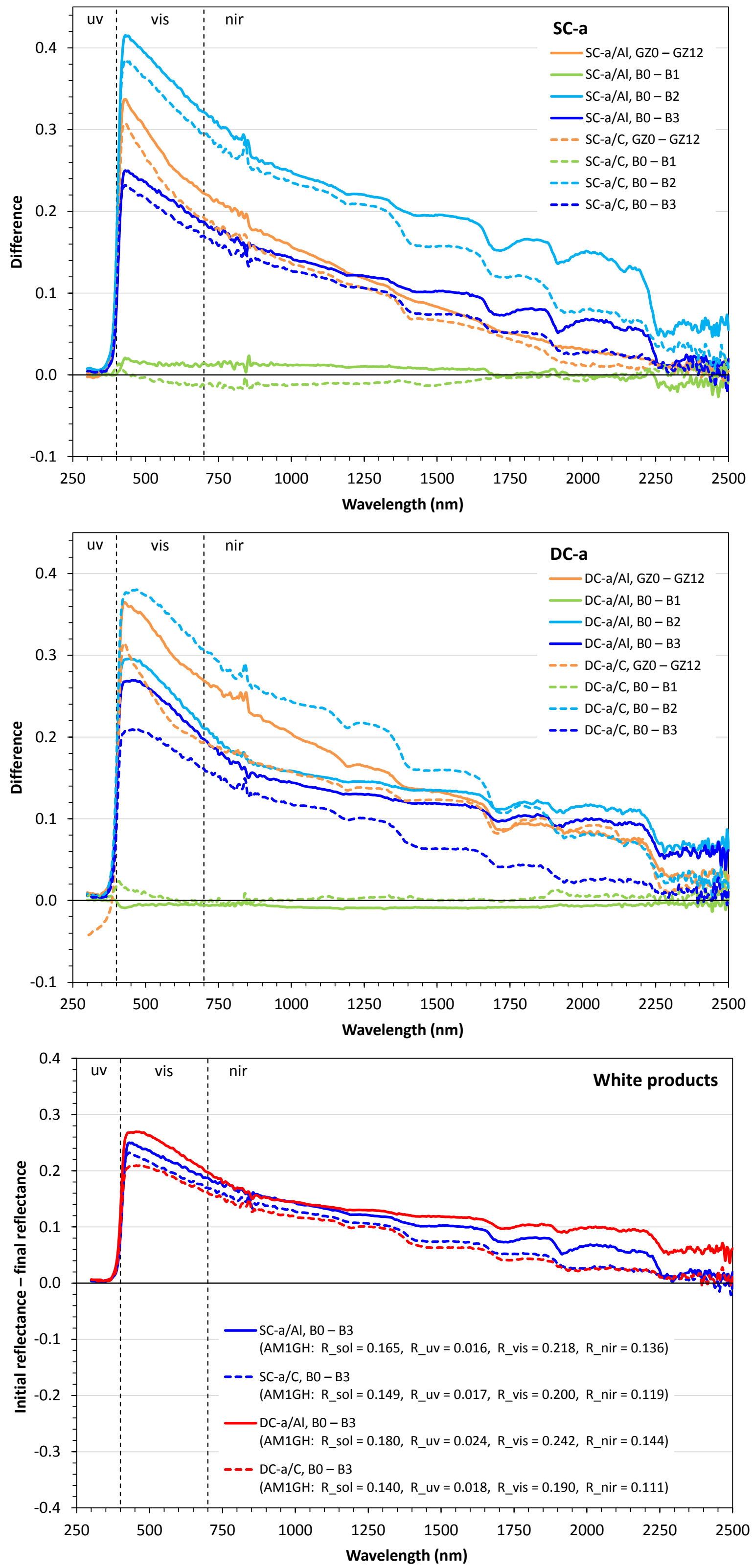


Figure 22. Spectral differences observed at each step of the lab aging process in Trial B.

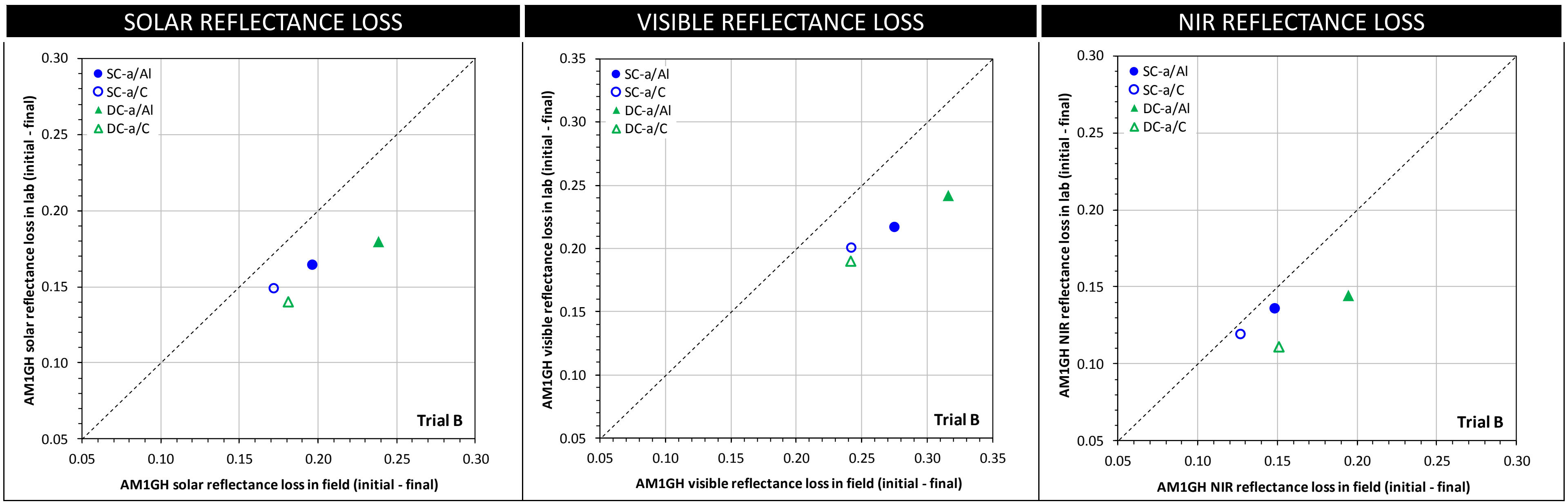


Figure 23. Comparison of broadband reflectance losses observed in Trial B and in Guangzhou field-exposure.

3.3.3 Trial C

In this trial, the amount of POM deposited in Step 2 was increased with respect to Trial B, as shown in Table 9. Trial C tested one specimen each of the four white products. Figure 24 presents the spectral changes observed at each step of the lab aging process, and Figure 25 presents a comparison of broadband reflectance losses between Guangzhou field-exposure and Trial C.

In this trial, two of the four tested products (SC-a/C and DC-a/Al) registered a success. The results were somewhat puzzling, however, as the successes and failures did not correspond neatly to either a coating product or a substrate type. In comparing the visible and near-infrared reflectance losses of the field-exposed and lab-aged specimens, it was concluded that subsequent trials should further increase the Step 2 loading of POM. Once again, we noted that DC-a/Al experienced significantly less cleaning in Step 3 than all other specimens.

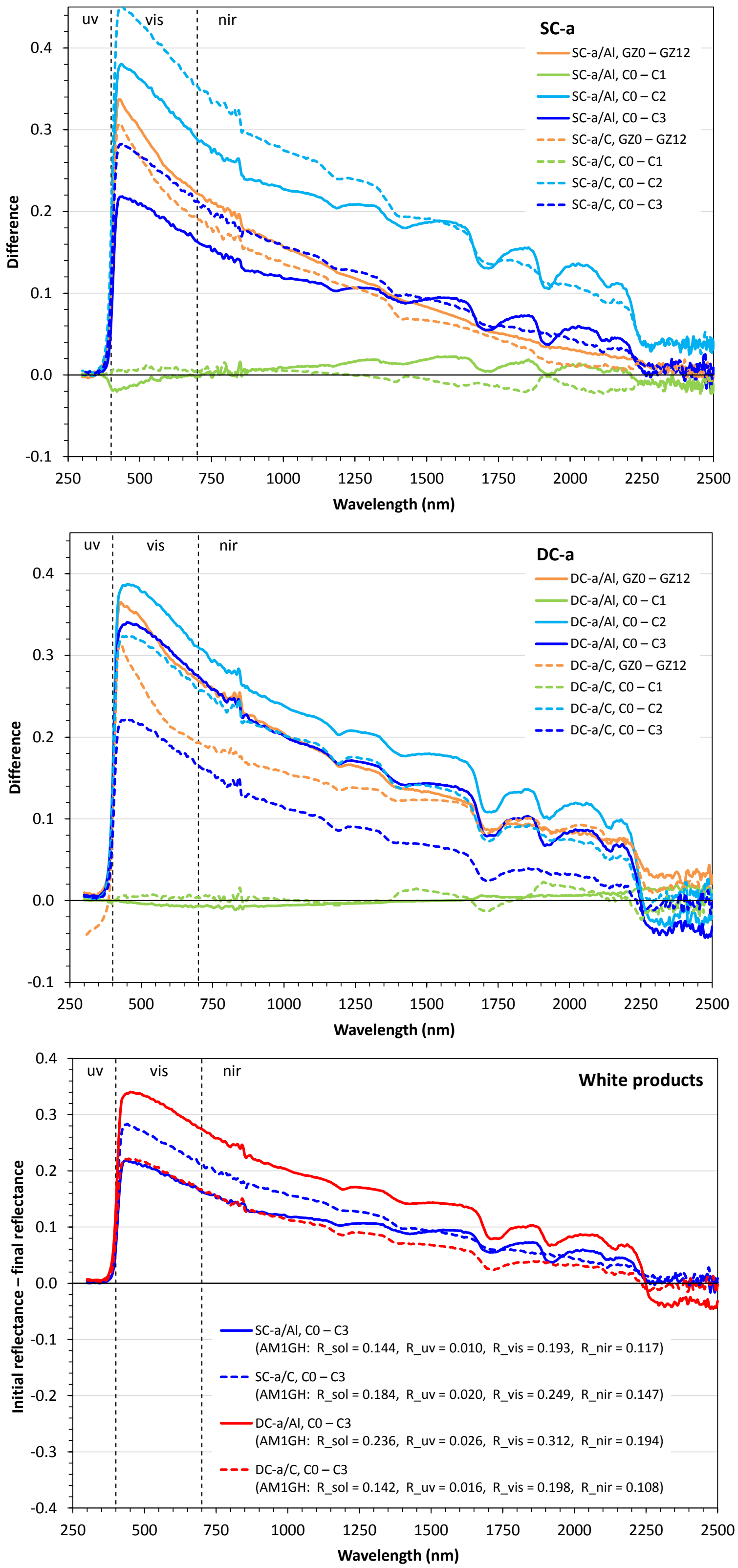


Figure 24. Spectral differences observed at each step of the lab aging process in Trial C.

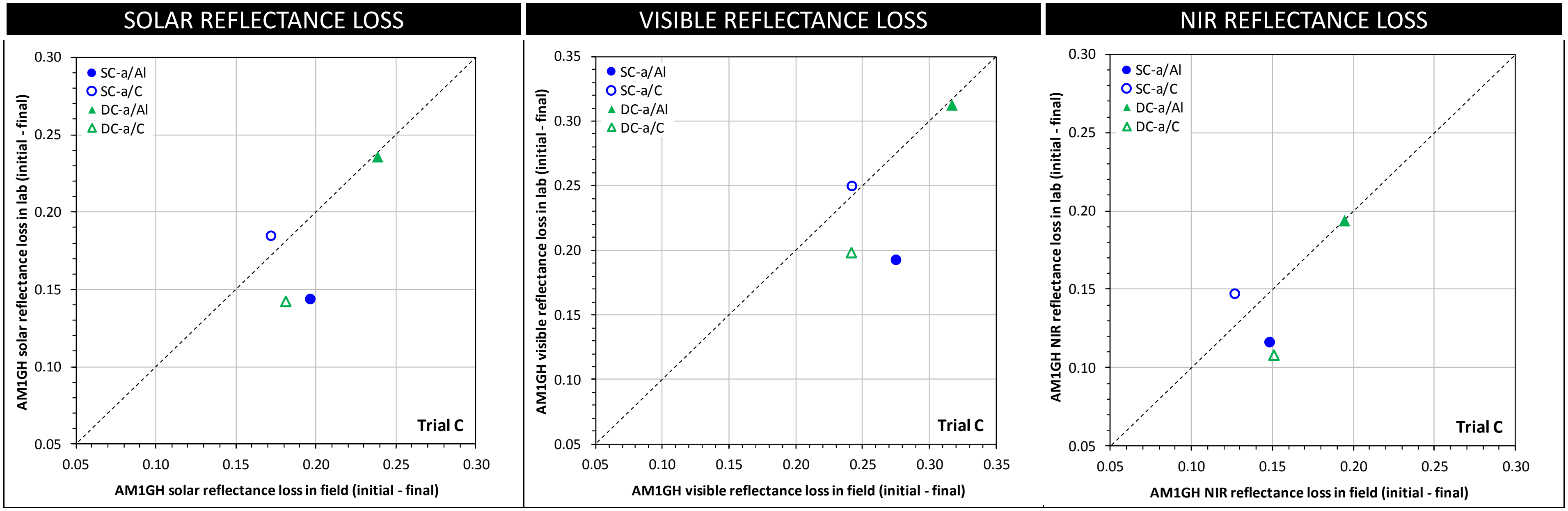


Figure 25. Comparison of broadband reflectance losses observed in Trial C and in Guangzhou field-exposure.

3.3.4 Trial D

In the previous trial it was determined that Trial D should employ a higher loading of POM in Step 2. To make a more informed estimate of the amount of increase necessary, we performed a short sub-trial examining spectral changes from soiling two of the white products (SC-a/C and DC-a/C) with varying amounts of humic acid. After we factored in the expected magnitude of Step 3 cleaning based on what we observed in previous trials, we finally concluded that a Step 2 POM loading on the order of 120 – 160 µg cm^{-2} was the target (see Figure 26).

Table 9 summarizes the soiling mixture composition and spray protocol used in Trial D, which once again tested one specimen each of the four white products. Figure 27 presents the spectral changes observed at each step of the lab aging process, and Figure 28 presents a comparison of broadband reflectance losses between Guangzhou field-exposure and Trial C. Specimen photographs are shown in Figure 29.

Overall, the solar, visible, and NIR reflectance losses observed in this trial exceeded the target. It was determined that in subsequent trials both the POM and soot loading should be decreased somewhat. We observed in this trial that the DC-a/Al sample curiously experienced slightly more Step 3 cleaning than in previous trials.

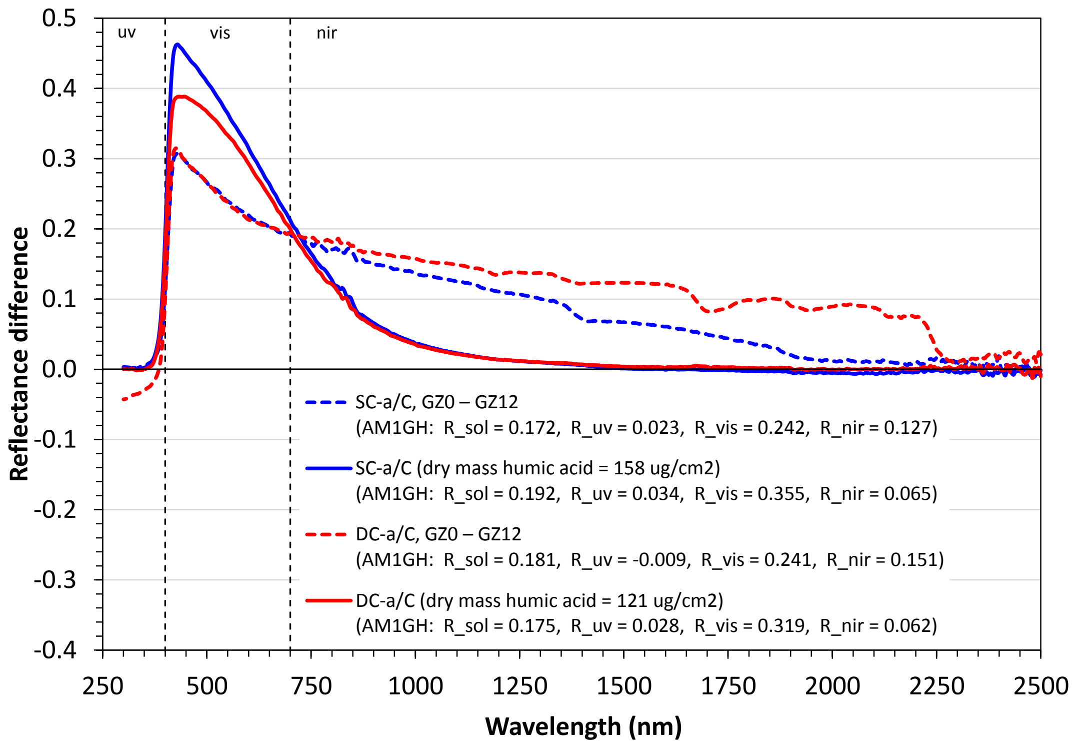


Figure 26. Spectral changes observed after soiling SC-a/C and DC-a/C with humic acid at a dry mass loading of 160 and 120 µg cm^{-2}, respectively. Solid lines represent initial reflectance – POM-soiled reflectance. Dashed lines represent initial reflectance – Guangzhou field-exposed reflectance, and are provided for reference.

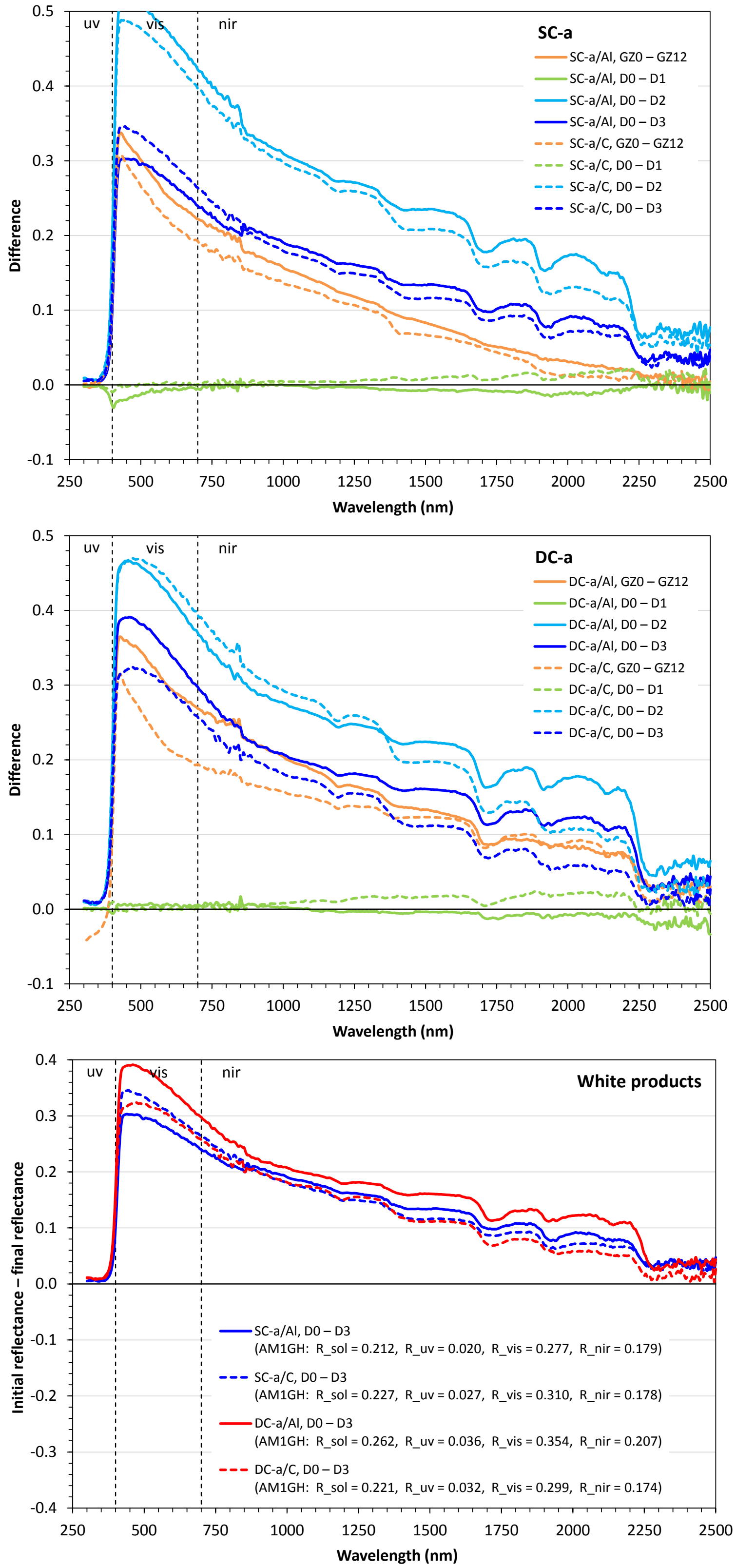


Figure 27. Spectral differences observed at each step of the lab aging process in Trial D.

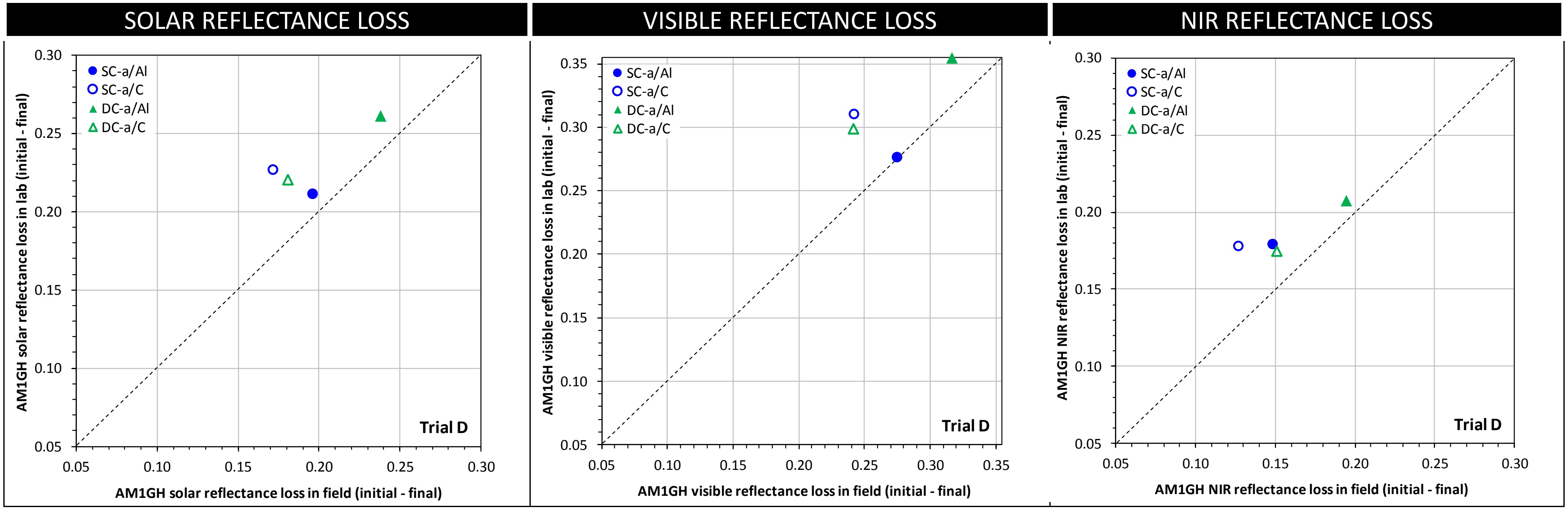


Figure 28. Comparison of broadband reflectance losses observed in Trial D and in Guangzhou field-exposure.

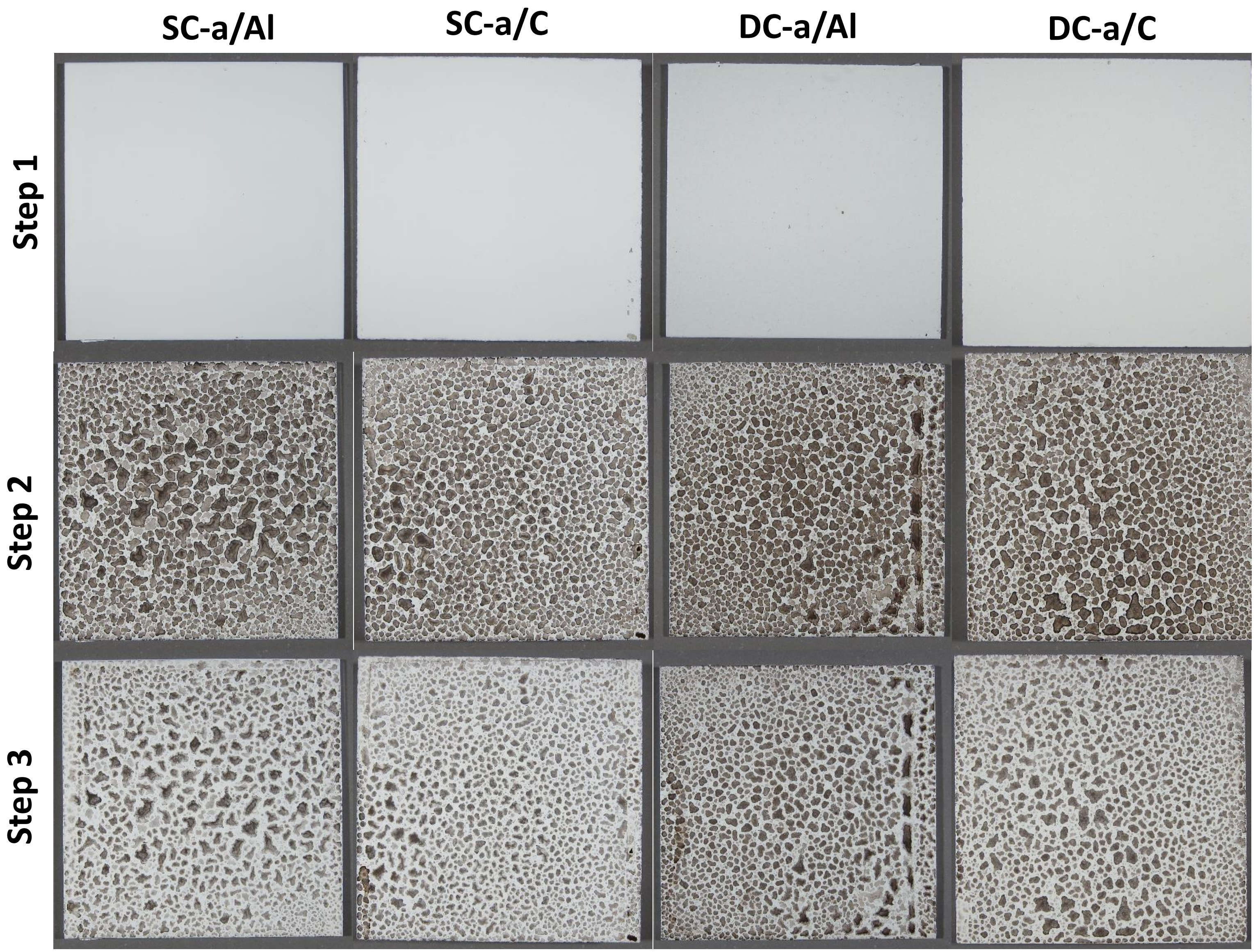


Figure 29. Photographs of Trial D specimens at each step of the lab aging procedure.

3.3.5 Trial E

In Trial E, the Step 2 loadings of POM and soot were reduced slightly, as shown in Table 9. Initially, we tested one specimen for each of the six products. When this first round produced 3 successes in the 6 products tested, an additional two replicates per product were subjected to Trial E to improve the statistical significance of the results.

All three rounds of Trial E reproduced quite closely the Guangzhou 1-year field-exposure reflectance losses in three out of six products. Interestingly, these three successes (SC-a/Al, DC-a/Al, and DC-b/Al) occurred in products with an aluminum substrate, and correspondingly the failures occurred in products with a concrete substrate. We decided to designate Trial E as the winner. Results are presented in Figure 30, Figure 31, Figure 32, Figure 33, and Table 10.

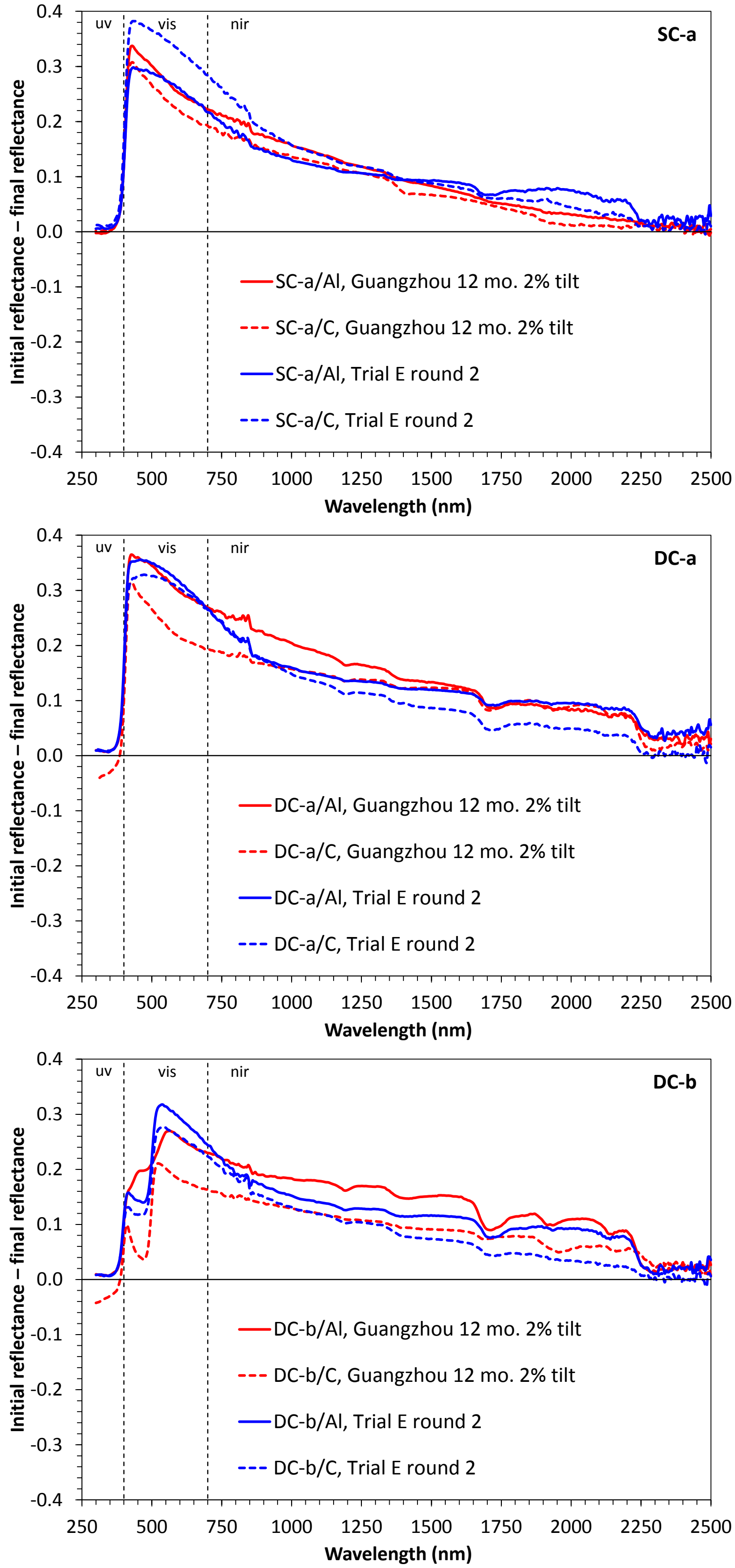


Figure 30. Spectral differences observed after 12-months of field-exposure in Guangzhou at 2% slope (red curves) compared to those achieved in Trial E round 2 (blue curves).

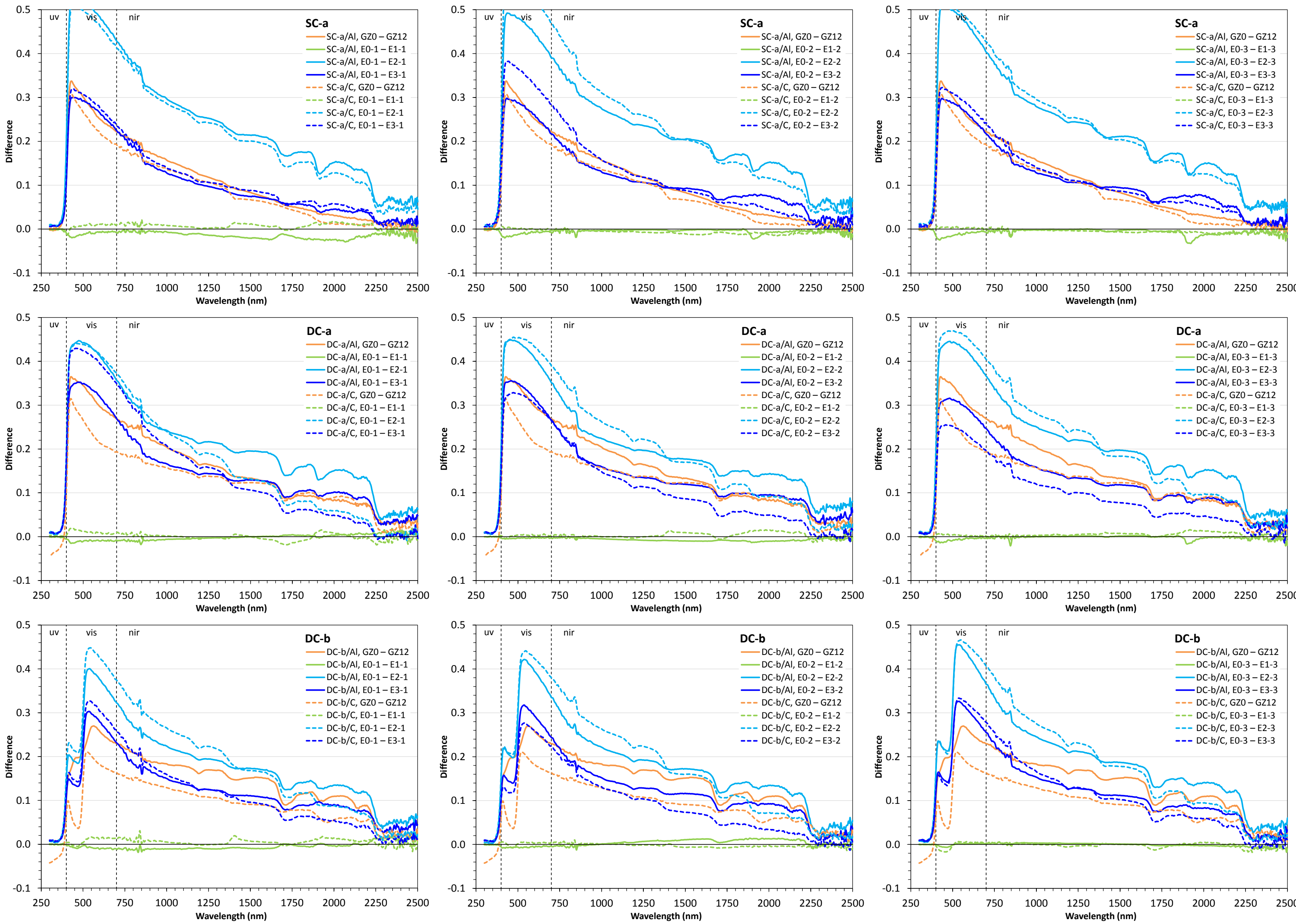


Figure 31. Spectral differences observed at each step of the lab aging process in Trial E. The data for all three rounds of Trial E are represented here.

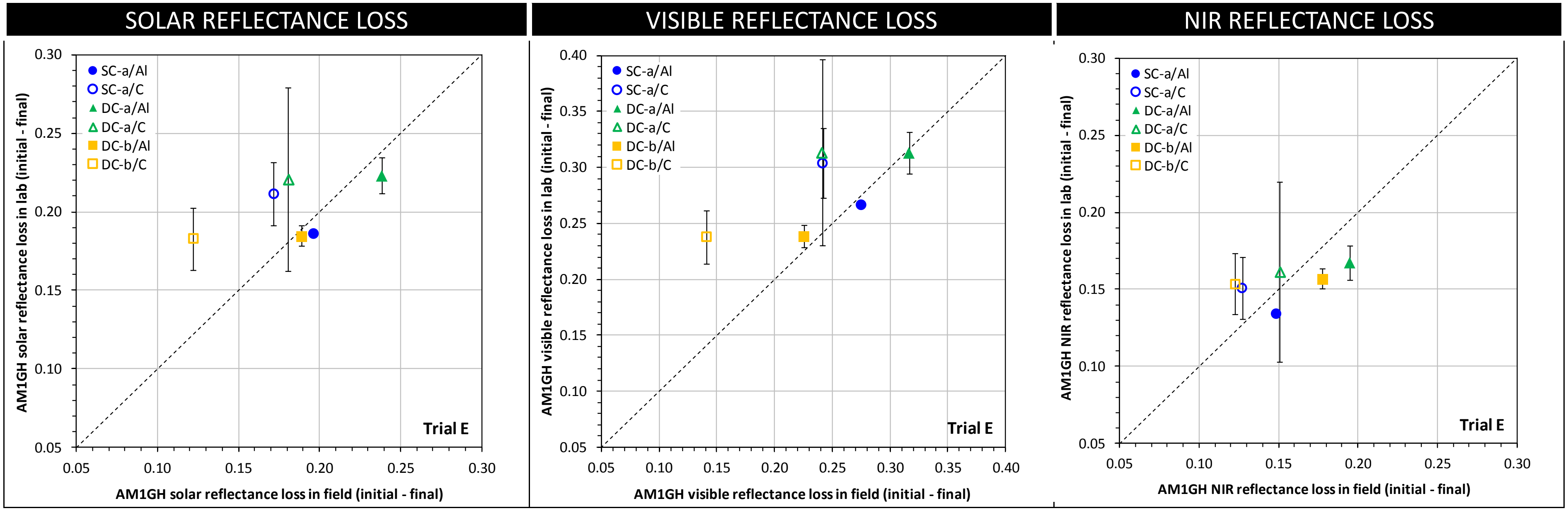


Figure 32. Comparison of broadband reflectance losses observed in Trial E and in Guangzhou field-exposure. Error bars represent the standard deviation (S.D.) of the three rounds in Trial E.

Table 10. AM1GH solar reflectances and reflectance losses of specimens tested in three rounds of Trial E, reported at each step of the laboratory aging process. Equivalent data for the Guangzhou field-exposed specimens is provided in the left three columns for comparison.

Specimen	GZ0	GZ12	GZ0 - GZ12	E0-1	E0-2	E0-3	E1-1	E1-2	E1-3	E2-1	E2-2	E2-3	E3-1	E3-2	E3-3	E0-1 – E3-1	E0-2 – E3-2	E0-3 – E3-3	E0 – E3 (avg)	E0 - E3 (S.D.)
SC-a/Al	0.803	0.606	**0.196**	0.815	0.811	0.826	0.824	0.818	0.833	0.449	0.473	0.478	0.627	0.625	0.640	0.187	0.187	0.185	0.186	0.001
SC-a/C	0.810	0.638	**0.172**	0.831	0.836	0.839	0.824	0.838	0.839	0.481	0.440	0.479	0.633	0.602	0.638	0.198	0.234	0.201	0.211	0.020
DC-a/Al	0.766	0.527	**0.238**	0.745	0.702	0.739	0.752	0.706	0.743	0.437	0.399	0.427	0.514	0.473	0.529	0.231	0.228	0.210	0.223	0.011
DC-a/C	0.743	0.562	**0.181**	0.741	0.759	0.757	0.736	0.757	0.754	0.439	0.437	0.420	0.459	0.545	0.592	0.282	0.214	0.166	0.221	0.058
DC-b/Al	0.708	0.520	**0.189**	0.692	0.684	0.686	0.700	0.685	0.685	0.443	0.426	0.406	0.515	0.498	0.496	0.177	0.186	0.190	0.184	0.007
DC-b/C	0.688	0.566	**0.122**	0.698	0.694	0.711	0.691	0.693	0.712	0.415	0.412	0.413	0.508	0.534	0.513	0.190	0.160	0.198	0.183	0.020

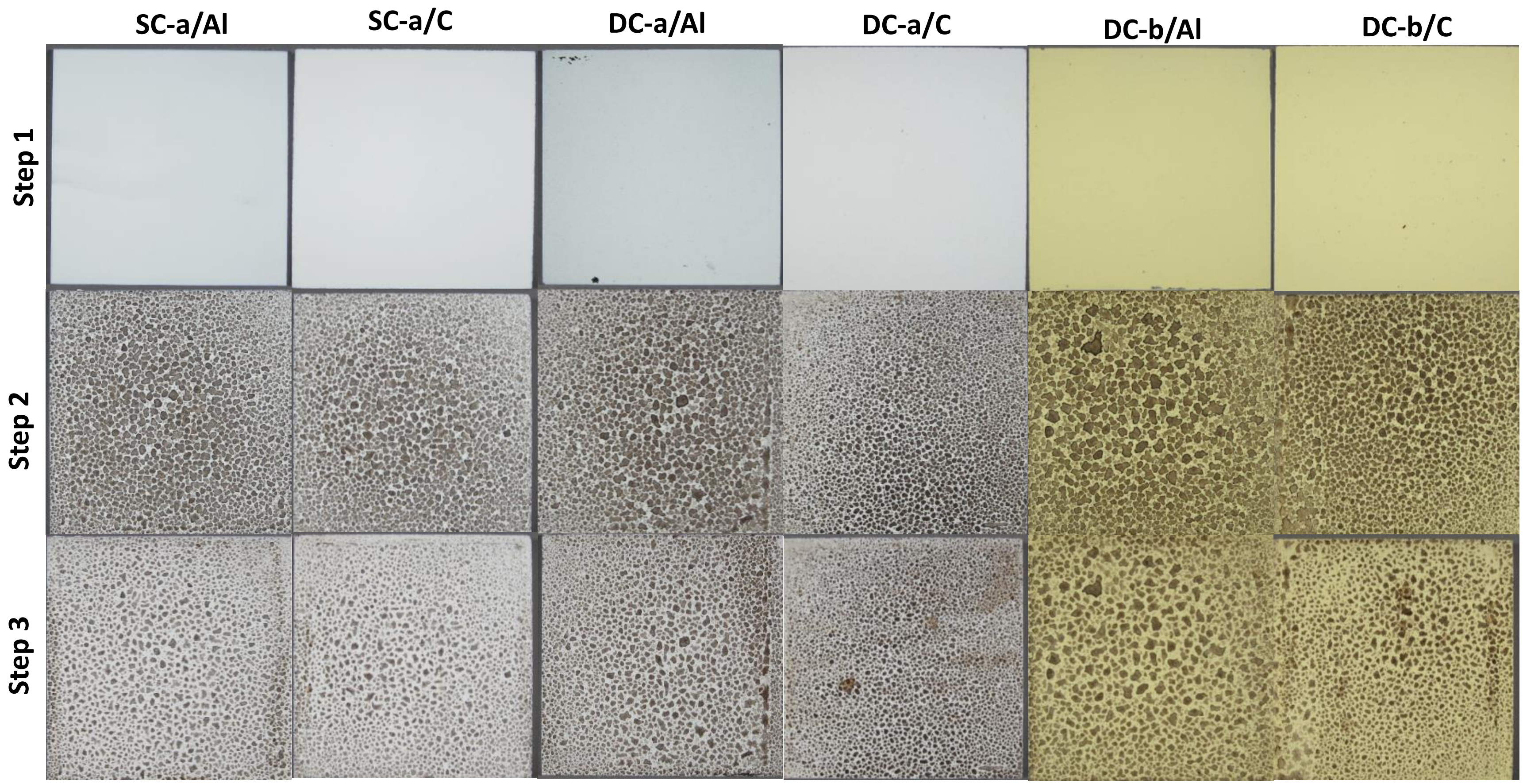


Figure 33. Photographs of Trial E (round #1) specimens at each step of the laboratory aging process.

4 Discussion

This study describes a limited number of tests conducted with using a small set of specimens. For that reason, the conclusions reached should be subject to additional verification using more samples and under a broader range of experimental conditions. The following observations were made to assess the effectiveness of laboratory aging in reproducing field aging results:

- Opposite to what was observed for the field-exposed GZ specimens, all three concrete specimens aged in Trial E experienced a greater amount of reflectance loss than did the aluminum specimens.
- In Trial E, all three aluminum (Al) substrate specimens achieved a lab-aged solar reflectance loss within ~0.015 of GZ0 – GZ12.
- In Trial E, all three concrete (C) substrate specimens exhibited a lab-aged solar reflectance loss no closer than ~0.04 of GZ0 – GZ12.
- The lab aging protocol represented in Trial E worked best for L0 specimens with an aluminum substrate.
- Results showed a different experimental uncertainty depending on the materials. For example, in the lab vs. field plot, DC-a/C had larger error bars than other samples. By contrast, the SC-a/Al sample had smaller error bars.
- It was interesting to note that in early trials, DC-a/Al experienced very little cleaning during Step 3, while in Trial E its Step 3 cleaning was on par with the other specimens. This may be due to lack of uniformity across the specimens, or variability inherent to the process.
- We do not have a very good explanation for the unexpected results in Trial A.

5 Challenges and lessons learned

Our team identified the following observations as lessons learned from this process that can help streamline similar efforts in the future:

- The variability observed between materials exposed in the field and the laboratory impeded accurate comparisons. In an ideal situation, specimens used to develop lab aging procedure should match those exposed in the field (e.g., come from the same manufacturing lot). Another limitation stemmed from the limited range of products and also the small number of specimens available for each product. In the reported study, using fifteen specimens was not enough to rigorously evaluate each trial. Results could be much more statistically significant with three specimens per trial, instead of only one.
- Ideally, a variety of products should be used in the development of any lab aging soiling mixture. The set of six products used here represented only one product category (field-applied coatings) and two manufacturers. It is worth noting that the lab aging procedure we developed in Trial E assumes that cleaning will occur during Step 3—as was observed in this set of six products—which may not occur with other product types.
- During the development process our team focused primarily on soot and humic acids (surrogates for particulate organic matter, POM), which are the two main agents affecting the reflectance spectra. But less attention was paid to other soiling agents such as salts and dust, which do not significantly impact reflectance compared to POM and soot. However, there should be a method prescribed to determine the amount of dust and salts needed in a soiling mixture, whether it be a default amount or a calculation.
- The development process could have been more streamlined if we had made better initial assumptions. To wit:
    - We should not have assumed there would be no cleaning during Step 3.
    - There was no compelling reason to have limited the soiling mixture to a total concentration of 1.24 g/L for the first few trials.
    - Given that the amount of dust and salts in Trial E was arbitrary, we should have at least kept the loading the same as with the US average ASTM D7897 procedure.
- To reduce iteration, we could have performed a set of pre-trials to investigate how much Step 1 and Step 3 are expected to change the reflectance of the product. Step 1 can affect the product's reflectance if it ages with UV/moisture exposure (some products turn yellow). Step 3 can affect the product's reflectance by washing off some of the soling accumulated in the surface during Step 2. Performing additional pre-trials would allow for more informed formulations of the first trial mixtures.

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