

Disintegration of lunar samples over time: A test

L. A. TAYLOR^{1,†}, J. V. HOGANCAMP², L. A. WATTS³, S. J. WENTWORTH⁴, P. D. ARCHER⁴,
R. A. ZEIGLER⁵, and A. BASU^{6,*}

¹Department of Earth and Planetary Sciences, Planetary Geoscience Institute, University of Tennessee,
Knoxville, Tennessee 37996, USA

²Geocontrols Systems—Jacobs JETS Contract, NASA Johnson Space Center, Houston, Texas 77058, USA

³Barrios Technology—Jacobs JETS Contract, NASA Johnson Space Center, Houston, Texas 77058, USA

⁴Jacobs, NASA Johnson Space Center, Houston, Texas 77058, USA

⁵Astromaterials Research and Exploration Science Division, NASA Johnson Space Center, Houston, Texas 77058, USA

⁶Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, Indiana 47405, USA

*Corresponding author. E-mail: basu@indiana.edu

(Received 17 March 2017; revision accepted 16 January 2018)

Abstract—Lunar samples provide ground-truth for all planetary exploration. Lunar soils, especially their <1 mm fraction, constitute the only primary standards for remotely sensing the composition of small airless planetary bodies. Therefore, maintaining the integrity, especially of the <1 mm fraction, takes on a much larger, big picture responsibility. A possibility has been suggested that lunar soils may disintegrate (to smaller grain sizes) if exposed to the Earth's moist atmosphere, thus losing some of their intrinsic value to science. We have tested that possibility by multiple, independent reanalyses with three techniques (wet-sieving in water and in alcohol, and laser diffractometry) using a fresh allocation of Apollo 17 “orange soil,” 74220. Our results are very similar to each other despite repeated soaking–drying in water, and also to those originally determined in the 1970s. We have also used a laser diffractometry technique to reanalyze the grain sizes of ~50 mg splits of eight soils that were initially analyzed three to four decades ago. The results are randomly different from previous measurements, which we attribute to nonrepresentative subsampling of very small amounts from previous allocations; ~50 mg is too small for obtaining representative aliquots. The results of grain-size analyses presented and discussed in this study indicate that the integrity of the lunar soil 74220, and indeed, all lunar soils, has not been physically compromised in the last four decades.

INTRODUCTION

One of the most important methods of planetary exploration is remote sensing *sensu lato*. Use of reflectance and thermal-emission spectra in the ultraviolet–visible–infrared (UV-VIS-IR) range of the electromagnetic spectrum has been, and is currently, the principal mode of assessing the surface chemistry and mineralogy of a planetary body. The spectra of minerals, rocks, and chemical compounds obtained in laboratories were used to calibrate and interpret those of extraterrestrial bodies, especially before the return of samples of the lunar regolith principally by the Apollo

astronauts (e.g., Binder and Cruikshank 1966; Aronson et al. 1967; Greenman et al. 1967; McCord 1969). The lunar regolith was formed primarily by impact processes and thermo-mechanical disintegration that resulted in mineralogical and chemical fractionation in different grain-size ranges, especially in the <1 mm fraction. Micrometeoritic impacts also produced small amounts of melt that aggregated mineral and lithic fragments together into agglutinates. Some of these impact melts reached temperatures high enough (2000–3000 °C) to selectively vaporize chemical components of the soil that subsequently condensed as thin veneers (i.e., 10s of nm) of silica-rich glass with myriads of single-domain, nanophase metallic iron—np-Fe⁰ (Keller and McKay 1993, 1997); this np-Fe⁰ substantially

[†]Deceased.

affects reflectance properties (Pieters et al. 1993; Noble et al. 2007). Indeed, reflectance spectra of various size fractions of lunar soils are quite distinct from each other (Taylor et al. 2001, 2010). All surficial processes, impact as well as bombardment by solar wind, which change the properties of freshly excavated material on the surface of the Moon and other small airless bodies, have been collectively termed “space weathering” (e.g., Zeller and Ronca 1967; Pieters et al. 1993, 2000; Hapke 2001; Bennett et al. 2013). All remote sensing of airless bodies is of space-weathered material, and it is the lunar soil that is the primary extraterrestrial ground-truth we have for inferring the mineralogical and chemical compositions of rocky planetary bodies, especially the small airless bodies. It follows that the integrity of the only extraterrestrial ground-truth we have—i.e., samples of the lunar regolith—be preserved as best as possible for future generations of planetary scientists. This is a time-honored charge for the Astromaterials Acquisition and Curation Office at the NASA Johnson Space Center.

On the basis of a recent determination of grain-size distributions of 20 lunar soils using a laser diffractometry technique, however, Cooper et al. (2015) suggested that the average grain sizes of lunar soils exposed to Earth’s moisture-laden atmosphere while stored in the laboratories of principal investigators (PIs), have been reduced by two to four times relative to previous measurements on original subsamples. They assign the cause of such mechanical disintegration of lunar soils to the reaction between terrestrial moisture (“water”) and the glass phases in lunar soils, which cement soil grains (e.g., agglutinatic glass). By extrapolation, their results could imply that the integrity of all lunar soils in storage may have been lost forever (for a dissent, see Taylor et al. 2016). Given the seriousness of possible compromises in all future lunar sample analyses, we deemed it essential to test if lunar (soil) samples have indeed disintegrated while in curatorial storage. We conducted our tests in two batches of grain-size analyses. The first batch consisted of one fresh ~2 g allocation of the 74220 “orange soil” from the Lunar Sample Curator. The second batch consisted of samples—actually subsplits—from the same allocations originally made to PIs McKay and Taylor. All of these samples, but different subsplits, had been analyzed by Cooper et al. (2015); the new results provide data for direct comparison.

The fresh allocation of soil 74220 was exposed to terrestrial atmospheric moisture, as we independently determined its particle size distribution (PSD) by three different methods, each measured by a different analyst. They are (1) wet-sieving in ultrapure water (UPW), (2) wet-sieving in isopropyl alcohol (IPA), and (3) laser

particle size analysis. The total time between the date of allocation and the date of the last PSD was about 6 months. The results of each different method are very similar to each other, consistent with results obtained in earlier studies of sample 74220 back in the 1970s.

The second batch consists of subsplits of samples of eight lunar soils (12001, 12030, 12033, 14260, 15041, 15071, 61141, and 67481) drawn from the same PI allocations that were analyzed by Cooper et al. (2015). We conducted PSDs of these samples by laser diffractometry using the same instrument in the Astromaterials Research and Exploration Science (ARES) facility at the Johnson Space Center in Houston and following the same standard operating procedure (SOP) of Cooper et al. (2015). The results are different.

Major PSD Techniques Applied to Lunar Soils

“The basic descriptive element of all sedimentary rocks [*and sediments*] is the grain size” (Tucker 2001). Within months of the return of Apollo samples, the grain-size distribution of Apollo 11 soils was reported in NASA’s Apollo 11 Preliminary Science Report (1969), and subsequently published by Duke et al. (1970). The principal methodology for determining PSD was sieving in air (dry) or using water (wet) or Freon (wet) and expressed in weight percent (Duke et al. 1970; Butler and King 1974; McKay et al. 1974). The PSD of the <20 μm fraction was obtained through variations of the Coulter counter method of dispersing soil grains in an electrolyte or alcohol and measuring the size of interference or dispersion of an optical beam (including laser) to estimate their model-dependent volume and conversion to weight (Lindsay 1972; McKay et al. 1974). It is to be noted that sieving <20 μm particles is neither efficient nor reproducibly accurate, largely because smaller sieve-openings become clogged and also because manufacturing uniform <10 μm sieves is a challenge. In many fields (e.g., soil science, clay mineralogy, and medicine), PSD determination in <10 μm sizes is essential, and it is routine to use laser diffraction methods (e.g., as by Microtrac©), along with free-fall sedimentation of particles in a dispersant (e.g., SEDIGRAPH; Buchan et al. 1993). In general, laser diffraction methods depend strongly on particle shape, causing distortion of PSD of nonspherical, irregular, and especially tabular particles (such as clay minerals). For data-based comparisons of different methods, see for example, Beuselinck et al. (1998) and Molinaroli et al. (2011). The ARES Division NASA-JSC has chosen to use the Microtrac© system (Plantz and Davis 2007) and their FLEX© software to correct for shape and other physical variabilities of particles.

METHODS USED FOR PSD

We performed three independent determinations of PSDs of three separate splits of the submillimeter fraction of the lunar soil 74220, carried out by three different analysts, using three different liquid dispersants. Care was taken by the lunar curatorial personnel to allocate a 2 g representative sample for these endeavors following the well-established curation procedures that have been practiced for decades.

Laser Particle Size Determination

Laser particle size analysis is a method in which lasers are directed at dispersed particles, producing scattered light patterns, which are used to estimate particle sizes. This method is based on the principle that large particles scatter light at small angles relative to the incident beam, whereas small particles scatter light at large angles relative to the incident beam (Fig. 1). We used a Microtrac Bluewave instrument consisting of a Sample Delivery Controller (SDC) and a S3500 Microtrac Bluewave laser diffraction system (LDS). The SDC delivers a well-dispersed sample in a carrier liquid (IPA, in this case) to the LDS, which then directs three stationary lasers at the dispersed particles. The tri-laser system changes the incident angle of the laser beam, which increases the number of detectors capable of detecting scattered light. The Microtrac Bluewave system utilizes a primary red laser and two shorter wavelength blue lasers, which are used to detect $<1 \mu\text{m}$ and nanoscale particles. This instrument has the ability to detect particles ranging from 0.01 to 2800 μm .

The specific operating procedure is created by each user and depends on the nature of the sample (e.g., particle shape, refractive index; the fluid carrier and its refractive index). In passing, we note that “Assumed values for absorption (the degree to which sediment grains absorb the light) can have a profound effect on grain-size results” (Sperazza et al. 2004). The procedure used in this study matched the procedure used by Cooper et al. (2015) and was tested using JSC-1A., a lunar soil simulant. The following parameters were utilized: run time of 60 s, transparent transparency, particle refractive index of 1.81, irregular particle shape, IPA as the carrier fluid, fluid refractive index of 1.375, and flow rate of 50%. Two aliquots of lunar soil 74220 (52.3 mg of split,894; and 43.1 mg of split,912) were separately homogenized in Wheaton glass vials using the “roll and tumble” method, i.e., by rotating the vial on its side. Approximately 50 mg of the homogenized sample was measured on weighing paper and transferred to a 5 mL glass beaker. A small amount (~1 mL) of 99.9% purity IPA was added to the lunar

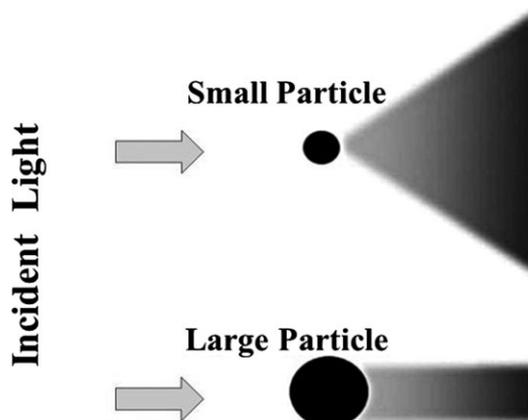


Figure 1. Small particles scatter light at larger angles relative to the incident beam compared to large particles (adapted after Malvern Instrumentation Ltd. 2013).

material and gently stirred with a metal spatula. The soil sample/IPA slurry was gently poured into the sample loading area of the SDC. The beaker was rinsed with a small amount of additional IPA, and this was poured into the sample loading area to assure complete sample transfer.

The first run was started immediately after loading the sample. Each run consisted of three 60 s measurements, the results of which could be viewed separately or averaged. Each sample was run nine times total, with 5-min intervals between the end of each run and start of the subsequent run. At the end of the third run, three drops of the dispersant Triton X100 (octylphenoxy polyethoxyethanol; 5% in DDI) was added to the sample loading area. After the sixth run ended, the sample was recirculated for 4–5 min and then ultrasonicated for 30 s before immediately starting the next run. This ultrasonication process was repeated after the seventh and eighth runs.

The same procedure was followed for the small (~50 mg) subsamples of eight soils in the second batch.

Wet-Sieving at NASA’s Curatorial Facilities in Houston

We resurrected, updated, and followed NASA-JSC’s SOP (Schwarz 2015) to wet-sieve 0.893 g of the lunar soil sample 74220,911. Briefly, the sieving operation was performed on a clean, laminar flow bench. The sieve stack was assembled using six sieves: 500, 250, 150, 90, 45, and 20 μm and a collecting pan at the bottom. According to the SOP, the airflow was turned off when the sample was uncovered in order to prevent loss of the finest fraction. The sample was transferred from its container to the top sieve on the stack. Antistatic equipment and a vibrator were used to gently remove all grains from the container. Using UPW from a

Teflon squeeze bottle, the sides and mesh of the sieves were carefully and repeatedly flushed through the sieve stack, which also aided in flushing finer particles from the surfaces of larger grains. The water level in the catch pan was monitored. Every time the pan was nearly full, it was placed on a hot plate at a setting of ~ 65 °C to evaporate.

The sieve stack was covered and placed on a sieve shaker to gently move the grains on the mesh. Vibration was set as low as possible and run for 15–20 min to prevent damage to the grains. The washing and vibrating of the sieves was repeated several times until only the largest grains remained. The top sieve was carefully placed on glassine paper and checked under a binocular microscope. If fine grains or dust were observed adhering to the sieve or to other grains, the process was repeated. When all the grains were disaggregated and clean, they were transferred to a folded glassine paper and then into a preweighed vial. The empty sieve was then placed on the stack and rinsed thoroughly, the water running through the sieves below and into the collecting pan. The underside of the sieve, in a tilted position above the stack, was rinsed with squirts of water and gently wiped using a sable brush. The brush and the stack were thoroughly rinsed several times until no dust was observed. If any grain fell through the sieve, they were transferred from the glassine paper to the next sieve in the stack.

The above procedure was repeated for each sieve in the stack. When all the sieves, down to 20 μm , were cleaned, the catch pan was set on the hot plate, and the water was evaporated without loss of sample. A sable-hair brush was used to transfer the <20 μm fraction to a folded glassine paper. Each sieve fraction was weighed, and then transferred to a preweighed vial for final storage. About 79 wt% recovery was achieved. In comparison with previous grain-size analyses of lunar soils, this recovery is low. It is likely that more of the finer sizes were lost relative to coarser sizes. Results show that this particular analysis records a slightly larger size for the soil 74220 (Table 1; Fig. 2 upright triangles), but not deviating much from other results.

This method not only exposes the sample to the Earth's moist atmosphere but actually soaks and dries the sample repeatedly. For the analysis of the subsample of 74220 reported here, the sieving process took over a month.

Wet-Sieving Using Alcohol

Wet-sieving of aliquot 74220,894 (915.1 mg) of the soil using IPA was conducted at UT, Knoxville. The procedure was essentially the same as that described above for the water wet-sieving, however with some

different sieve sizes. Six, 3 inch diameter sieves (sizes 500, 250, 125, 75, 45, and 20 μm) were used. Two pans with depths of 2 and 5 cm, depending on the anticipated amount of IPA–sediment mix, were utilized to catch the alcohol and the finer particles. A mild shaker apparatus was applied to the sieves and pans during the sieving. A stiff, 1 cm wide sable brush was utilized to mix and stir and move the soil particles through each sieve. After each wash through the sieves, the bottoms of each sieve were washed and brushed with alcohol and collected in the pans in order to have the pan contain only the soil smaller than the last sieve size and the IPA. The brush was washed repeatedly to remove as many particles as possible. The IPA in this pan was allowed to evaporate to dryness in the laboratory, assisted by a warm plate of ~ 45 °C.

The sieves were utilized two at a time, along with a bottom pan. The 500 and 250 μm sieves were the easiest to use, and only a 2 cm deep pan was sufficient to catch all the pass-through soil and IPA. This pan was allowed to dry, and this remaining soil was carefully brushed into the top of the next sieve—i.e., 125 μm . The 125 and 75 μm sieves required much more IPA, and the 5 cm deep pan was used. This pan was allowed to dry overnight, and the dry pan was carefully brushed to remove the <75 μm soil. The deep pan was used again for the 45 and 20 μm sieves. Each dried size fraction was collected in preweighed, tare-corrected glass vials and subsequently weighed. About 94 wt% recovery was achieved.

RESULTS

Results of the new grain-size analyses of the <1 mm fraction of sample 74220 are presented in Table 1. This also includes results of all previous analyses of the <1 mm fraction of the soil 74220 as compiled by Graf (1993) and checked against original publications. We provide a ready conversion at the bottom of Table 1 because many results of the old analyses and derived statistics were reported in the ϕ -scale (e.g., Frondel et al. 1971; Butler et al. 1973). The results of the analysis conducted by Cooper et al. (2015) are much finer than all other data conducted between 1971 and 2017 (Table 2). The weight percent of the <20 μm fraction and the median grain size reported by Cooper et al. (2015) are 85 wt% and 9.5 μm , respectively, but the corresponding median grain size (Md) and average of all other analyses are 28 wt% and 44 μm , respectively. All data are plotted as cumulative curves in Fig. 2. Except for one outlier (i.e., Cooper et al. 2015), all other results for 74220—although not identical—plot together as a bundle in a restricted area.

Results of the new laser diffraction PSD of the second group of eight different soil samples are

Table 1. Results of grain-size analysis of lunar soil 74220 (<1 mm fraction).

Cumulative % of recovery										
Analyst ^c		DSM	DSM	B&K	WVE	BLC	LAT ^b	LAW ^b	H&A ^b	H&A ^b
Year		1974	1974	1974	1976	2015	2016	2016	2017a	2017b
Split #		,6	,82	,84	,121	,791	,894	,912	,912	,894
Size (µm)	Phi ^a (φ)									
1	9.97	100.00	100.00	99.82	100.00	99.74	100.00	100.00	100.00	100.00
3.9	8.00	98.7	99.5			87.72				
7.8	7.00	93.4	95.4			61.51				
10	6.64			89.12	88.9					
15.6	6.00	82.1	80			26.22				
20	5.64	72.8	69.9	76.02	68.6	15.25	67.45	73.8	72.65	70.56
30	5.06			65.22	58.2	5.14				
37	4.76			50.22						
44	4.51			46.72						
45	4.47	46.1	43.4		44.7	1.73	48.39	52.5	41.37	39.19
53	4.24			40.52						
60	4.06				36.3	0.85				
75	3.74	31.2	28.5	32.72		0.52	32.75		22.08	22.89
90	3.47	26	24		25	0.37		32.2		
105	3.25			23.62						
125	3.00			18.82			18.71		11	13.25
150	2.74	12.6	11.8	14.02		0.17		17.2		
177	2.50			10.82						
200	2.32				8.6			9.3		
250	2.00	4.9	4.2	5.92		0.08	4.47		2.15	3.83
420	1.25			2.22						
500	1.00	0	0.1		1.5	0	0.26	3.3	0	0.33
841	0.25			0.02						

^aφ = -(log mm)/(log 2).

^bUnpublished new analyses.

^cDSM = McKay et al.; B&K = Butler and King; WVE = Engelhardt et al.; BLC = Cooper et al.; LAT = Taylor et al.; LAW = Watts; H&A = Hogancamp and Archer.

presented in Table 3. For ready reference, primary sources of all published data are included and the current results are listed under Hogancamp and Archer, 2017 (year of analyses). For comparison, only the graphical mean grain size (M_Z ; Folk and Ward 1957) of the size distributions is listed. These new results, in general, are different from previous results; M_Z and M_d values are higher, lower, or the same at random.

DISCUSSION

Reproducibility

Many lunar soils have been sieved multiple times, by different groups for different purposes, using different methods and with very small allocations that may not have been representative of the bulk soil (McKay et al. 1991; Graf 1993). Given such variability, identical results are not to be expected, but general agreement between the results of different groups does exist. Model-dependent corrections to volume-based PSD by the laser diffraction techniques commonly, but not necessarily, overestimate

the <10 µm fraction of terrestrial samples, in which platy phyllosilicates, i.e., clay minerals, are abundant (Blott and Pye 2006; Di Stefano et al. 2010). Note that a variation of the laser diffraction PSD has been advocated specifically for 1–10 µm fraction for clayey samples on Earth (Sperazza et al. 2004). However, results of laser diffraction determination of PSD are usually self-consistent and not too different from those obtained by sieving or sedimentation (Blott and Pye 2012). For the specific case of Apollo soil 74220, consider the results of laser diffraction PSDs of split numbers ,894 and ,912 (Tables 1 and 2). Despite using barely 50 mg samples, both results are almost identical (Table 2). Sieving results are also close to each other. For 74220, the median grain size and the <20 µm fraction in sieving results from the 1970s were 49 µm and 28%, respectively; those obtained herein were 43 µm and 28%, respectively (Table 2). Because the <20 µm fractions are identical in both, it appears that the minor discrepancy is possibly a result of variability in allocations.

A comparison of the results of the new laser diffraction PSD of eight separate samples with those

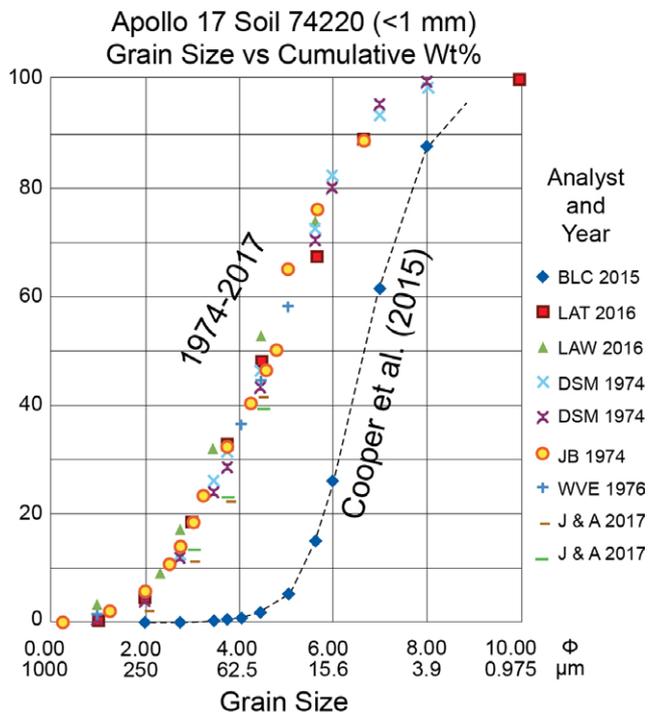


Figure 2. Cumulative grain-size frequency distributions of the submillimeter fraction of the Apollo 17 soil 74220 as determined by various analysts between 1974 and 2017.

that have been analyzed previously provides a new insight into lunar sample storage under terrestrial conditions, PI handling, and subsampling of very small amounts (Table 3). There is no systematic relationship between the new results (column C) and those initially obtained by others in the period between 1971 and 1981 (column A). Hence, the cause of the difference is likely to be random.

Disintegration of Lunar Samples Over Time

The results of grain-size analyses presented and discussed above indicate that *the integrity of the lunar soil 74220 has not been compromised in the last four decades* while stored in dry nitrogen ambience in Johnson Space Center. Similarly, additional results of grain-size analysis of Apollo 11 soil 10084, by King et al. (1971; dry sieving) and that by Basu et al. (2001, 2002; wet-sieving in *water*) three decades later are nearly identical. Mean grain size and standard deviation of the aforementioned PSD are 52 and 25 μm , and 51 and 24 μm , respectively. This demonstrates that the integrity of lunar soil 10084, also stored in dry nitrogen ambience in Johnson Space Center, was not compromised in 30 yr. Wet-sieving that required complete soaking and drying in multiple cycles also did not disintegrate soil 10084 in any measurable way.

Table 2. Median grain size and wt% for the $<20 \mu\text{m}$ portion of the lunar soil 74220 ($<1 \text{ mm}$ fraction).

Analyst, year	Split #	Median (μm)	$<20 \mu\text{m}$ wt%
McKay et al. (DSM; 1974)	,6	50.8	27%
McKay et al. (DSM; 1974)	,82	46.1	30%
Butler & King (B&K; 1974)	,84	50.1	24%
Englehardt et al. (WVE; 1976)	,121	48.0	31%
Cooper et al. (BLC; 2015)	,791	9.5	85%
Taylor et al. (LAT; 2016)	,893	43.0	33%
Watts et al. (LAW; 2016)	,911	43.0	26%
Hogancamp (H&A; 2017)	,912	35.9	27%
and Archer (H&A; 2017)	,894	35.4	29%

Thin sections of lunar material—mostly of the regolith, large grains, and rock slices—all without any cover, have been stored under terrestrial atmosphere since 1969. None of them have cracked or fractured or disintegrated. Thin sections in the Lunar Educational Packages, also without any cover, which have been used in many colleges and universities over decades are the best examples of having survived intact.

Discrepancies Between the Results of Different PSDs

As mentioned above, the results from our PSD by three techniques, collectively, differ by a factor of 2 to >4 from those obtained by Cooper et al. (2015), who used the same laser diffraction instrument at the JSC facilities. The differences are real, not only for 74220 and the second group of eight samples but also for the 11 other lunar soils analyzed by Cooper et al. (2015). These vast differences are present despite using the same equipment and presumably the same procedure used in the present study. It is possible that the very small soil samples (about 25 mg) used for each of their PSD were not sufficient to be statistically representative. However, that does not explain why all 20 soils would appear to be so much finer (see Table 3; results in column A and C are similar but those in column B are consistently much finer).

CONCLUSIONS

Three new grain-size analyses of the $<1 \text{ mm}$ fraction of the Apollo 17 lunar soil 74220 and previous analyses of Apollo 11 soil 10084 indicate that storage and handling in the curatorial facilities of NASA-JSC has maintained the soil's integrity for more than four decades, despite prolonged (weeks-long) wet-sieving in water, which required multiple soaking–drying cycles.

Results of new laser diffraction PSD of eight subsamples from the same parent splits allocated to PI McKay and PI Taylor and exposed to terrestrial atmosphere for many years also see no systematic

Table 3. Comparison of graphic mean (M_Z) and median (M_d) grain sizes of eight lunar soils.

A Sieve (1971–1981) Sample, split ^a			B Laser PSD (2015) Sample, split ^b			C Laser PSD (2017) Sample, split ^c		
M_Z (μm)	M_d (μm)	<20 (μm)	M_Z (μm)	M_d (μm)	<20 (μm)	M_Z (μm)	M_d (μm)	<20 (μm)
61141,3			61141,17			61141,17		
75.3	80.2	19%	31.7	35.3	36%	96.88	65.9	22%
15071,19			15071,37			15071,37		
52.1	64.1	24%	43.6	48.7	26%	78.08	58.91	16%
12033,37			12033,40			12033,62		
60.7	72.8	16%	35.8	36.9	20%	34.21	28.57	38%
12030,34			12030,182 & 183			12030,182		
110.1	107.1	12%	33.9&20.2	39.8&23.5	32%&43%	101.42	53.97	24%
67481,23			67481,89-90			67481,89-90		
109.5	119.9	16%	38.1	42.7	33%	110.06	67.5	23%
14260,4			14260,91-92			14260,91-92		
82.6	71.4	19%	40.9	47.9	27%	65.49	55.33	18%
15041,34			15041,56			15041,56		
36.8	47.4	29%	28.9	34	34%	81.18	54.34	19%
12001,average			12001,925			12001,925		
66.4	58.2	19%	42.8	47.7	27%	54.5	43.54	24%

^a12001 (averaged from Frondel et al. 1971; King et al. 1971; Wood et al. 1971); 12030,34 and 12033,37 (King et al. 1971); 14260,4 (McKay et al. 1972); 15041,34 and 15071,19 (Basu et al. 1981); 61141,3 and 67481,23 (Heiken et al. 1973).

^bCooper et al. (2015).

^cHogancamp and Archer (new, 2017).

degradation of grain size. Instead, variations observed for small subsamples are random (larger, smaller, or similar mean grain size) relative to early analyses. It is likely that the very small sizes of the splits (~50 mg) do not accurately sample soil grain-size distribution for the bulk soil originally returned by Apollo astronauts. Regardless, the new results are not compatible with the suggestion that exposure to atmospheric moisture disintegrates lunar soils.

It is not possible to explain why Cooper et al. (2015) consistently found smaller M_Z and smaller M_d than the original.

Thus, given the present state of knowledge, it is concluded that lunar soils have not disintegrated over time because of exposure to terrestrial atmospheric moisture; therefore, these soils will continue to provide critical insights into the bombardment history of the Moon and the effects of space weathering on airless bodies in the inner solar system.

Acknowledgments—The Lunar Curatorial Personnel are appreciated for preparing the sample of orange soil 74220, allocated by CAPTEM. ARES at Johnson Space Center provided support for SJW, DA, and JVH. The senior author (LAT) also is most thankful for the major support of the two SSERVI organizations at Brown-MIT (SEED) and University of Central Florida (CLASS).

We thank two anonymous reviewers and especially AE Carle Pieters who have kindly provided insightful feedback and have allowed us to improve earlier versions

of the manuscript. Finally, we are immensely grateful to Dong-Hwa Taylor for her constant support and help throughout the process that resulted in this paper.

Editorial Handling—Dr. Carle Pieters

REFERENCES

- Apollo 11 Preliminary Science Report. 1969. NASA SP-214, Library of Congress Cat. No. 77-603770. 204 p.
- Aronson J. R., Emslie A. G., Allen R. V., and McLinden H. G. 1967. Studies of the middle- and far-infrared spectra of mineral surfaces for application in remote compositional mapping of the Moon and planets. *Journal of Geophysical Research* 72:687–703.
- Basu A., McKay D. S., Griffiths S., and Nace G. 1981. Regolith maturation on the earth and the moon with an example from Apollo 15. Proceedings, 12th Lunar and Planetary. Science Conference. pp. 433–449.
- Basu A., Wentworth S. J., and McKay D. S. 2001. Submillimeter grain-size distribution of Apollo 11 soil 10084. *Meteoritics & Planetary Science* 36:177–181.
- Basu A., Wentworth S. J., and McKay D. S. 2002. Erratum: Submillimeter grain-size distribution of Apollo 11 soil 10084. *Meteoritics & Planetary Science* 37:145.
- Bennett C. J., Pirim C., and Orlando T. M. 2013. Space-weathering of solar system bodies: A laboratory perspective. *Chemical Reviews* 113:9086–9150.
- Beuselinck L., Govers G., Poesen J., Degraer G., and Froyen L. 1998. Grain-size analysis by laser diffractometry: Comparison with the sieve-pipette method. *Catena* 32:198–208.
- Binder A. B. and Cruikshank D. P. 1966. Lithological and mineralogical investigation of the surface of Mars. *Icarus* 5:521–525.

- Blott S. J. and Pye K. 2006. Particle size distribution analysis of sand-sized particles by laser diffraction: An experimental investigation of instrument sensitivity and the effects of particle shape. *Sedimentology* 53:671–685.
- Blott S. J. and Pye K. 2012. Particle size scales and classification of sediment types based on particle size distributions: Review and recommended procedures. *Sedimentology* 59:2017–2096.
- Buchan G. D., Grewal K. S., Claydon J. J., and McPherson R. J. 1993. A comparison of Sedigraph and Pipette methods for soil size analyses. *Soil Research* 31:407–417.
- Butler J. C. and King E. A. 1974. Analysis of grain size-frequency distributions of lunar fines. Proceedings, 5th Lunar Science Conference. pp. 829–841.
- Butler J. C., Greene G. M., and King E. A. 1973. Grain size frequency distributions and modal analyses of Apollo 16 fines. Proceedings, 4th Lunar Science Conference. pp. 267–278.
- Cooper B. L., Thaisen K., Chang B. C., Lee T. S., and McKay D. S. 2015. Disintegration of Apollo lunar soil. *Nature Geoscience* 8:657–658.
- Di Stefano C., Ferro V., and Mirabile S. 2010. Comparison between grain-size analyses using laser diffraction and sedimentation methods. *Biosystems Engineering* 106:205–216.
- Duke M. B., Woo C. C., Bird M. L., Sellers G. A., and Finkelman R. B. 1970. Lunar soil: Size distribution and mineralogical constituents. *Science* 167:648–650.
- Engelhardt W. v., Hurrle H., and Luf E. 1976. Microimpact-induced changes of textural parameters and modal compositions of the lunar regolith. Proceedings, 7th Lunar Science Conference. pp. 373–392.
- Folk R. L. and Ward W. C. 1957. Brazos river bar: A study in the significance of grain size parameters. *Journal of Sedimentary Petrology* 27:3–26.
- Frondel C., Klein C. Jr., and Ito J. 1971. Mineralogical and chemical data on Apollo 12 fines. Proceedings, 2nd Lunar Science Conference. pp. 719–726.
- Graf J. C. 1993. *Lunar soils grain size catalog*. Houston, Texas: NASA Reference Publication 1265. 488 p.
- Greenman N. N., Burkig V. W., and Young J. F. 1967. Ultraviolet reflectance measurements of possible lunar silicates. *Journal of Geophysical Research* 72:1355–1359.
- Hapke B. 2001. Space weathering from Mercury to the asteroid belt. *Journal of Geophysical Research Planets* 106:10,039–10,073.
- Heiken G. H., McKay D. S., and Fruland R. M. 1973. Apollo 16 soils: Grain size analyses and petrography. Proceedings, 4th Lunar Science Conference. pp. 251–265.
- Keller L. P. and McKay D. S. 1993. Discovery of vapor deposits in the lunar regolith. *Science* 261:1305–1307.
- Keller L. P. and McKay D. S. 1997. The nature and origin of rims on lunar soil grains. *Geochimica Cosmochimica Acta* 61:2331–2340.
- King E. A. Jr., Butler J. C., and Carman M. F. 1971. The lunar regolith as sampled by Apollo 11 and Apollo 12: Grain size analyses, modal analyses, and origins of particles. Proceedings, 2nd Lunar Science Conference. pp. 737–746.
- Lindsay J. F. 1972. Sedimentology of clastic rocks returned from the Moon by Apollo 15. *Geological Society of America Bulletin* 83:2957–2970.
- Malvern Instruments Ltd. 2013. Overview of important particle characterisation techniques. <http://www.azom.com/article.aspx?ArticleID=9937>. Accessed February 2017.
- McCord T. B. 1969. Color differences on the lunar surface. *Journal of Geophysical Research* 74:3131–3142.
- McKay D. S., Heiken G. H., Taylor R. M., Clanton U. S., Morrison D. A., and Ladle G. H. 1972. Apollo 14 soils: Size distribution and particle types. Proceedings, 3rd Lunar Science Conference. pp. 983–994.
- McKay D. S., Fruland R. M., and Heiken G. H. 1974. Grain size and evolution of lunar soils. Proceedings, 5th Lunar Science Conference. pp. 887–906.
- McKay D. S., Heiken G. H., Basu A., Blanford G., Simon S., Reedy R., French B. M., and Papike J. J. 1991. The lunar regolith. In *Lunar sourcebook*, edited by Heiken G. H., Vaniman D., and French B. M. Cambridge, UK: Cambridge University Press. pp. 285–356.
- Molinaroli E., de Falco G., Matteucci G., and Guerzoni S. 2011. Sedimentation and time-of- transition techniques for measuring grain-size distributions in lagoonal flats: Comparability of results. *Sedimentology* 58:1407–1413.
- Noble S. K., Pieters C. M., and Keller L. P. 2007. An experimental approach to understanding the optical effects of space weathering. *Icarus* 192:629–642.
- Pieters C. M., Fischer E. M., Rode O., and Basu A. 1993. Optical effects of space weathering: The role of the finest fraction. *Journal of Geophysical Research Planets* 98:20,817–20,824.
- Pieters C. M., Taylor L. A., Noble S. K., Keller L. P., Hapke B., Morris R. V., Allen C. C., McKay D. S., and Wentworth S. 2000. Space weathering on airless bodies: Resolving a mystery with lunar samples. *Meteoritics & Planetary Science* 35:1101–1107.
- Plantz P. E. and Davis P. 2007. Recalculation of Microtrac data to emulate data from sieves, sedimentation and other methods: Microtrac Application Note SL-AN-24 Revision A. 6 p.
- Schwarz C. M. 2015. Office of the Curator Sample Processing Procedure SPP 19 (July 16, 2015; last update).
- Sperazza M., Moore J. N., and Hendrix M. S. 2004. High-resolution particle size analysis of naturally occurring very fine-grained sediment through laser diffractometry. *Journal of Sedimentary Research* 74:736–743.
- Taylor L. A., Pieters C. M., Keller L. P., Morris R. V., and McKay D. S. 2001. Lunar mare soils: Space weathering and the major effects of surface-correlated nanophase Fe. *Journal of Geophysical Research Planets* 106:27,985–27,999.
- Taylor L. A., Pieters C. M., Patchen A., Taylor D.-H. S., Morris R. V., Keller L. P., and McKay D. S. 2010. Mineralogical and chemical characterization of lunar highland soils: Insights into the space weathering of soils on airless bodies. *Journal of Geophysical Research Planets* 115: <https://doi.org/10.1029/2009JE003427>.
- Taylor L. A., Liu Y., and Lofgren G. 2016. Integrity of lunar soil samples. *Nature Geosciences* 9:87. <https://doi.org/10.1038/ngeo2637>.
- Tucker M. E. 2001. *Sedimentary petrology*, 3rd ed. Oxford: Blackwell. 262 p.
- Wood J. A., Marvin U. B., Reid J. B., Taylor G. J., Bower J. F., Powell B. N., and Dickey J. S. 1971. Mineralogy and Petrology of the Apollo 12 Lunar Sample. SAO Special Report 333. 272 p.
- Zeller E. J. and Ronca L. B. 1967. Space weathering of lunar and asteroidal surfaces. *Icarus* 7:372–379.