STUDIES ON THE DEPOSITION AND REMOVAL OF RADIOACTIVE SOIL

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The rate of deposition and removal of a radioactive milk soil from several surfaces have been studied. The nature of the surface exhibited a small but measurable effect on the rate of deposition of a radioactive milk soil on the surfaces tested. However, the surfaces showed no significant effect on the rate of soil removal. The build-up that took place on all surfaces due to repeated soilings without washing was not a simple additive accumulation of residue but appeared to be a selective deposit of a residue slowly laid down over a period of time and difficult to remove. As the accumulated residue increased, a point was reached beyond which there was no significant difference among surfaces in the rate of soil build-up. Subsequent washing removed only part of the soil and even repeated washings had little effect unless special heavy duty cleaning was applied.

The importance and problem of cleaning food equipment have been emphasized by the many studies reported in recent years. In general, these studies were concerned with cleaning materials and methods, bacteriological cleanability of various types of surfaces or the amount of soil remaining on these surfaces.

The objective of this investigation was to obtain data on the rate of soil build-up, and the rate of soil removal from surfaces under several test conditions.

REVIEW OF LITERATURE

If a surface is free of all organic and inorganic residues, then it is necessarily free of microbiological contamination; however, a surface may be free of microbiological contamination and still be covered with organic or inorganic residues.

Kaufmann *et al.* (9) observed that there was no apparent bacteriological build-up accompanying a visual soil build-up on the walls of the experimental bulk tank after 12 soilings using water rinses with no detergent followed by chlorine sanitization; at the expiration of this time the surfaces complied with bacterial requirements as recommended by Standard Methods (1) at least 96% of the times tested. Holland *et al.* (5) in a study of in-place cleaning of sanitary equipment found no correlation between film deposits and bacterial counts. They found some of the lowest bacterial counts where heavy films were readily visible.

Radioactive tracer methods according to Harris

(4) are the most sensitive measure of residues at the present time and are in general ten times as sensitive as the next best method. Cucci (3) used P^{32} in the form of phosphoric acid, which was mixed with milk, to study deposition of radioactive soil on rubber, glass and tygon tubing. Jennings et al. (6) in a preliminary experiment to their extensive project on circulation cleaning compared in vivo labeled milk, prepared by injecting a P³² solution into a cow, with in vitro P³² labeled milk, prepared by adding a radioactive phosphorous solution to homogenized milk, in their cleaning regimens and found no significant difference between in vivo and in vitro labeled milk. They concluded that the use of in vitro labeled milk was justified as an index of both organic and inorganic residues.

GENERAL EXPERIMENTAL PROCEDURE

This study of radioactive soil deposition and removal consisted of four groups of experiments. All soiling and washings (except to clean plates before a test or replication) were carried out with all plates in the rack shown in Figure 1 so each plate would be subject to the same treatment.

Radioactive milk soil was prepared by mixing fresh pasteurized skim milk, obtained from the University Dairy Plant, with P³² as phosphoric acid and with formaldehyde added as a preservative². The solution was thoroughly mixed in a lucite container which was placed in a small, top-opening refrigerator



Figure 1. Rack with plates.

¹Journal article No. 2857 from the Michigan Agricultural Experiment Station, East Lansing, Michigan.

TABLE	1-Рво	FILES ^a	OF	THE	SURFA	CE	FINISH	OF	THE	16
Sta	INLESS	STEEL	Pı	ATES	USED	IN	RADIO	лст	IVE	
		7	Гва	CEB	STUDIE	s				

ai	Profile	Profile in microinches		
plate surface	Average	Range		
2B	18.00	18 ^b		
3	22.75	22-24		
4	18.25	18-19		
7	15.75	14-17		

^aMeasured using a Brush Surface Analyzer, Manufactured by the Brush Instruments Co., Cleveland 14, Ohio. ^bAll four plates had the same profile.

adjusted to maintain the radioactive milk soil at $36^{\circ}F$.

The plates were new at the beginning of these experiments and were cleaned initially by the same procedure used by Kaufmann *et al* (8).

The plates were soiled by submerging the rack of plates in the radioactive milk. (This dipping operation was carried out without removing the container of radioactive milk from the refrigerator.) The plates were allowed to remain in the radioactive milk solution for about 3 seconds; they were then removed, placed in a vertical position, allowed to drain, and dried prior to radioactive counting.

The dipping procedure used to soil the plates was constant throughout the experiment; however, drying times, temperatures and washing procedures varied among groups of tests.

Plates

Sixteen type 302 stainless steel plates, four of each of the four finishes (Nos: 2B, 3, 4 and 7) were used throughout this study. These 16 plates were produced by five different manufacturers; the four plates of one finish were from at least two manufacturers. The plates were 8 inches square and each plate was cut from a whole stainless steel sheet. The roughness of the finish (profile) was measured as the root mean square of the deviations of the peaks and valleys from the mean in micro-inches. A summary of the profiles of the 16 stainless steel plates is listed in Table 1. Four polished glass plates were included in the Group I and II studies for general interest purposes; the data for the glass plates are included in the results, however, a discussion of these data are beyond the scope of this study.

Radioactive counting

In general, the radioisotope technique outlined by Comar (2) was used. A Sugarman type Geiger-Miller disintegration counter with a Nuclear Instrument and Chemical Corporation, Model 161A scaling unit was used to count the radioactivity of the residue retained by the plates. The plates were placed in a jig, shown in Figure 2, with the head of the counter placed above the plate and a 1-minute count was made on each of four areas on the plate. The window of the counter had a weight per unit area of 1.4 mg per sq cm and was 2.8 cm in diameter.



Figure 2. Jig for counting plates, counter and scaling unit.

A counting time of one minute was selected following the analysis of the counts of a standard C¹⁴ disk for 25 successive 1-minute periods. The counts showed a normal distribution with a mean of 480 counts per minute and a coefficient of variation of 0.042. A like test was done on one spot on a 2B plate and on a glass plate, both of which had been soiled with radioactive milk. The mean for the 2B plate was 1901 counts per minute, the coefficient of variation was 0.024; the mean for the glass plate was 1229 counts per minute, the coefficient of variation was 0.036.

Two 10-minute background radioactive counts were made each day the plates were counted, one before starting a series of countings and the other at the conclusion of the countings. They ranged from 19 to 23 counts per minute; the average was used as the correction for background count.

The counting system was standardized daily by counting a C^{'4} source of known radioactivity. All counts were first corrected for background and this net count was then corrected for time decay back to the time of known radioactive level. Therefore, the results for the four Groups of tests can be compared after correcting the counts for differences in the initial amount of P^{s2} and the volume of solution.

GROUP I STUDIES

Experimental

Group I was an accumulative soiling study followed by a repeated cleaning. The objective of the

² P³² was obtained from the Oak Ridge National Labtoratory, Oak Ridge, Tenn.



Figure 3. Results of Group I soiling test.

soiling study was to evaluate the effect of surface finish on the rate of soil build-up. The objective of the repetitive washing study was to evaluate the effect of the surface on the rate of soil removal.

The 16 type 302 stainless steel plates and four polished glass plates were tested using a radioactive soil made by mixing 1.0 ml of phosphoric acid solution containing 0.9 mc P^{32} and 7 ml formaldehyde with 6.5L of skim milk.

The plates were repeatedly dipped and dried without washing for a total of 30-dipping cycles. The plates were allowed to dry in a ventilated hood at room temperature $(70^{\circ}F)$ for at least 30 minutes between dippings.

The plates with the residue accumulated through 30 dippings were subjected to 10 washing cycles using the following procedure: the plates were soaked for 1 minute in 160° F water containing chlorinated alkaline detergent³ (1 ounce per 5 gallons of water); then washed by lifting the plate holder containing the plates up and down five times as rapidly as possible. The plates were allowed to drain for approximately 1-minute, then rinsed by dipping three times in tap water at 120° F and then allowed to stand vertically and dry. The radioactivity of the residue remaining on the plate was counted

after each washing cycle. The plates dried for at least an hour between washings.

Results

The counts after 6, 14, 22 and 30 dippings were subjected to an analysis of variance (10) which showed no difference a mong plates of the same finish after soiling or after washing. (P was greater than 0.25)³. The counts on the plates with the same finish were combined and these sums are shown in Figure 3 as a function of the number of dippings (The count for dipping number one is taken from Group III and IV studies and is the average corrected to the same initial radioactive level as Group I test.) An analysis of variance of the radioactive



Figure 4. Results of Group I washing test.

counts after the 30 dippings suggested differences among finishes (P less than 0.005). Duncan's Multiple Range Test indicated that the three groupings are: 2B, 3; 4, 7; glass.

The results of the repetitive washing study are

³Klenzade Mfg. Co., Inc., Beloit, Wisconsin.

³P is the chance probability of occurrence on an F-value as large as or larger than that observed (see ref. 10)

shown graphically in Figure 4 where the sum of the counts on the four plates of the same finish are shown as a function of the number of washings. An analysis of variance of the percent reduction in radioactive count after washing showed no difference among stainless steel finishes (P was greater than 0.25).

GROUP II STUDIES

Experimental

Group II was an accumulative soiling study similar to Group I except: (a) the radioactivity of the soil was larger by a factor of 2.56 since 3.0 ml of phosphoric acid solution containing 2.3 mc P³² and 7 ml. formaldehyde were added to 6.5L of skim milk; (b) the number of dippings in radioactive soil was increased to 44 with radioactive counts being made after 5, 10, 15, 21, 27, 33, 39 and 44 dippings, and (c) the number of washing cycles was increased to 18.

Results

An analysis of variance of the radioactive counts after 5, 10, 15, 21 and 44 dippings showed no significant difference among plates with the same finish after soiling or after washing (p greater than 0.25). The counts on the plates having the same finish were combined and these sums as a function of the number of dippings are shown in Figure 5 (the count for dipping number one is taken from Group III and IV studies and is the average corrected to the same initial radioactive level as the Group II Tests).

The radioactive counts on the five surfaces appear to be different after 5 and 10 dippings but not different after 15, 21 and 44 dippings (for 5 and 10 dippings P is less than 0.005).

The results of the repetitive washing study are shown graphically in Figure 6 where the sum of the counts on the four plates having the same finish are shown as a function of the number of washings. An analysis of variance of the percent reduction in radioactive counts resulting from the 18 washings showed no difference among finishes (P greater than 0.25).

GROUP III STUDIES

Experimental.

The Group III studies were designed to evaluate the soil deposition characteristics of a clean surface and the rate of soil removal from a one-time-soiled surface.

In this study, the 16 stainless steel plates were tested. The plates were arranged in the rack according to a series of random numbers. A different random order was used for each of the 10 replications. A single test consisted of a heavy duty wash-



Figure 5. Results of Group II soiling test.



Figure 6. Results of Group II washing test.

ing to remove all soil (radioactive count at background level), dipping in radioactive soil, drying, radioactive counting of the residue, one controlled washing (described under Group I washing study), radioactive counting of the residue. The following procedure was used to clean the plates before starting each test: The plates were soaked overnight in a cleaning solution containing a chlorinated alkaline detergent at the rate of 2 oz per 5 gallons of hard water. The following morning they were scrubbed with a cellulose sponge and detergent and then rinsed in tap water followed by a distilled water rinse. The radioactive count of the clean plates was checked, after which the plates were assembled in the rack and dried prior to dipping in the radioactive milk soil. The radioactive milk solution was prepared by adding approximately 1 ml of phosphoric acid solution containing 10 mc P³² and 7 ml of formaldehyde to 7.8L of skim milk.

The drying and controlled washing procedure were the same as for Group I and II.

Results, washing study

The data from the controlled washings as percent reduction in radioactive counts were tested by an analysis of variance. There appears to be no difference among finishes (P = 0.23) or among plates of the same finish. The radioactive counts were reduced by an average of 97.6% in the one washing.

GROUP IV STUDIES

Experimental

The Group IV studies were identical with Group III studies except that a milder washing procedure was used which permitted more residue to remain on the plates. In this washing procedure the plates were dipped once, no soaking period, in 149°F water containing 1 oz alkaline detergent per five gallons of water, and immediately rinsed by dipping twice in 120°F water. The plates were dried in a hot-air oven (130-140°F) prior to being counted. After each soiling the plates were washed, dried, and counted three times.

Results, washing study

The reduced washing procedure in the Group IV studies left more soil on the plates than the Group III studies. Therefore, it was possible to subject the plates to three washing and counting cycles and still have counts that were measurably above background. The results indicated that there was no difference in the percent reduction in counts after washing; the P of the analysis of variance F was 0.20 for finishes and 0.25 for plates of the same finish. The average percent reduction in radioactive counts of the plates, by finishes, after each of the three washings are listed in Table 2.

Results, Group III and IV soiling study

The soiling procedures for Group III and Group IV were identical; therefore, in addition to analyzing each set of data it was possible to analyze the combined data. Data are summarized in Table 3.

Таві	E	2-Perc	ENT F	EDUCTIO	ONOF	RADIO	ACTIVE	COUNT.
Group	IV	STUDY	AFTEF	FIRST,	Secon	D, AND	Third	WASHING

Washing	Percent reduction of radioactive count of plate surfaces indicated :							
No.	$2\mathbf{B}$	3	4	7				
1	70.8ª	71.9	71.5	72.8				
2	74.2	75.4	74.6	76.1				
3 9	75.2	76.6	76.5	77.5				

"These values represent the average of 10 replications.

The Group III data indicated a difference in the radioactive count among finishes (P less than 0.005) also a difference among days, (P less than 0.005). To determine which plates were different the data for Nos. 2B and 3, also 4 and 7, were analyzed separately. Nos. 2B and 3 are not different, also 4 and 7 were not different (P less than 0.005 in both tests).

The Group IV data showed the same trend toward differences in the four finishes as the Group III data indicated; however, differences were smaller (P = 0.15).

TABLE 3-SUMMARY OF RADIOACTIVE COUNTS IN GROUP III AND IV STUDIES.

	Radioa	ctive counts in t	housands
Stainless steel plate surface	Group III counts 10 dippings	Group IV counts 10 dippings	Groups III & IV combined, 20 dippings
2B		91	171
3	79	87	166
4	65	100	165
7	67	83	150

"Sum of counts on the four plates of that finish.

Analysis of the combined data of the Group III and Group IV studies indicates differences among finishes. However, in the combined analysis a significant interaction, (P = 0.005) appeared. When the mean squire of this interaction was used to calculate F (10) instead of the error mean square, P increased from 0.020 to 0.100.

TABLE 4-RANKING OF FINISHES RELATIVE TO THE RADIO-ACTIVE COUNT, COMBINED GROUP III AND VI DATA.

		No.			
Stainless steel plate surface First ^a		Second	Third	Fourth	Avg. scoreb
2B	4	10	3	3	45
3	7	4	5	4	46
4	6	4	5	5	49
7	3	2	7	e e - 8	60

*First indicates highest count.

^bOn the basis of 1 point for first, 2 points for second, etc.

A summary of the ranking of the plates on the basis of average radioactive count by finish for the 20 tests in Group III and IV are shown in Table 4.

DISCUSSION

The problem under investigation was soil deposition and soil removal from surfaces commonly used on food processing equipment. The results generally indicated that soiling and washing are not simple soil-additive or soil-removal phenomena.

Soiling and soil build-up

In the soil deposition studies a significant difference in radioactive count was found in the Group I studies and after 5, and 10 dippings in the Group II studies. In the Group II studies all five surfaces seemed to pick up soil at approximately the same rate after 10 dippings. The lines in the soiling graph, Figure 5, are approximately parallel over the 15 to 44 dip range. The data from Group I corrected for comparative purposes show similar trends, but for some unexplained reason the plates did not show as great an increase in radioactive count after 14 dippings as did Group II after 10 dippings. (Note: a correction must be made for difference in initial radiation of the radioactive milk soil before comparing Group I and II counts directly.) The fact that the plates were new at the time of the Group I studies may be the explanation for the difference.

These data suggest that build-up in the absence of washing takes place in three stages; initial soiling, concurrent soil removal and deposition with net soil level remaining approximately constant and finally concurrent soil removal and deposition with level of soil increasing uniformly. It seems probable that in the middle phase there is a selective build-up of the soil that clings most tenaciously to the surface. The data in Figure 5 seems to indicate that once the build-up stage is well established all surfaces add soil at the same constant rate.

In all cases where significant differences were observed the same relationship among surfaces existed in that finishes 2B and 3 tended to be similar as did finishes 4 and 7. Where ranking was employed the order of decreasing counts was usually 2B, 3, 4 and 7.

Soil Removal

In all studies there appeared to be no differences in the rate of radioactive soil removal (expressed as the percentage of soil present) either among finishes or among plates of the same finish. These results are in agreement with those reported by Kaufmann *et al.* (8) in their studies on the relative bacteriological cleanability of stainless steel finishes. The rate of soil removal proved to be most interesting as shown in Figures 4 and 6. The first washing removed radioactive soil equivalent to the amount deposited in about the last 10 dippings. The effect of subsequent washing varied from Group I to Group II, in Group I washings 2 through 10 had practically no effect on soil removal.

The washing data was plotted as the logarithm of the radioactive counts vs washings, Figures 4 and 6, since the rate of removal of a soil is usually an exponential function of the number of washings. In Group II, Figure 6, there was an exponential reduction in radioactive counts with number of washings that extended approximately from wash 2 to 9 which were followed by the ineffective washings 10 through 18.

In the Group IV studies a reduced washing technique was used and the plates were washed and counted three times. Even though these plates were soiled only once the three washings did not produce an exponential radioactive count removal rate as was expected, but gave a pattern similar to Figure 4 and 6 where only the first wash was unquestionably effective. The fact that a rigorous cleaning procedure was necessary to reduce the radioactive count of a soiled plate to the background level is another indication that some of the soil adheres tightly to these surfaces.

Relative Behavior of Finishes

The differences in radioactive counts on plates with different finishes when dipped in a radioactive milk soil has been consistent throughout this study and these differences have been consistently small. The trend was for finishes Nos. 2B and 3 to behave similarly and finishes No. 4 and 7 to behave similarly. The overall trend was for the No. 2B finish to have the highest counts. In three out of the four groups of tests there were no measurable differences between the No. 4 and No. 7 finishes.

The differences in the behavior of the glass and steel surfaces evaluated is undoubtedly due to the nature of the surface in respect to chemical and physical characteristics, which determine the rate of residue deposit and build-up. The interaction between micro particles of soil (perhaps at the molecular level) and the surface of the metal (again, at the molecular level) will determine how tightly the soil is bound to the surface.

The differences observed in this study among stainless steel plates are due to the surface rather than the plate material since in the original selection of the 8 x 8 inch panels (8) each panel was cut from a different sheet and panels were obtained from several stainless steel manufacturers. If a basic material or manufacturing technique was a factor there would have undoubtedly been differences among plates with the same finish.

Recent work by Jennings and Bourne (7) substantiates the belief that soil deposition and the adherence of soil to a surface is a very complicated physiochemical relation in which the nature of the surface has a role.

Acknowledgement

The authors wish to acknowledge and thank Dr. R. C. Nicholas of the Food Science Department for his assistance in the preparation of this manuscript.

References

1. American Public Health Association. Standard Methods for Examination of Dairy Products. 10th Ed. 1953.

2. Comar, C. L. Radioisotopes in Biology and Agriculture. McGraw-Hill Pub. Co., New York, N. Y. 1955.

3. Cucci, M. W. Use of radioactive phosphorus to measure the amount of milk stone deposited on rubber, pyrex glass

and tygon tubings. J. Milk and Food Technol. 17: 11, 332, 333. 1954.

4. Harris, J. C. Detergency Evaluation and Testing. Interscience Publishers Inc., New York, N. Y. 1954.

5. Holland, R. F., J. D. Shaul, D. A. Theokas and H. M. Windlaw. Cleaning stainless lines in place. Food Engineering **25**:5, 75. 1953.

6. Jennings, W. G., A. A. McKillop and J. R. Luick. Circulation cleaning. J. of Dairy Science. 40:1471. 1957.

7. Jennings, W. G. and M. C. Bourne. Some physiochemical relationships in cleaning hard surfaces. Paper presented at 21st Annual Meeting of the Institute of Food Technologists, May 8, 1961, New York, N. Y. 1961.

8. Kaufmann, O. W., T. I. Hedrick, I. J. Pflug, C. G. Pheil, and R. A. Keppeler. Relative cleanability of various stainless steel finishes after soiling with inoculated milk solids. J. Dairy Science, 43:28. 1960.

9. Kaufmann, O. W., T. I. Hedrick, I. J. Pflug and C. G. Pheil. Relative cleanability of various finishes of stainless steel in a farm bulk tank. J. Milk and Food Technol. 23:377. 1960.

10. Snedecor, G. W. Statistical Methods, 5th Ed. The Iowa State College Press, Ames, Iowa.

NEWS AND EVENTS

RESUME OF THE SECOND NATIONAL CONGRESS ON ENVIRONMENTAL HEALTH

Three organizations sponsored the Second National Congress on Environmental Health which was held at Ann Arbor, Michigan, June 6, 7, and 8, 1961. These were, The University of Michigan, School of Public Health, The American Public Health Association and the National Sanitation Foundation. Some two hundred persons from the fields of public health, industry, commerce and education assembled to hear nationally known speakers discuss the many and varied aspects of the environment and its influence on man.

Carey P. McCord, M. D., of the University of Michigan, expressed the purpose and goal of the Conference in these words:

"The purpose of the Congress was to explore environmental resources and their value and usefulness for man in terms of needs for industry, government, and conservation - in keeping with health social and economic trends: to consider principles and methods of practice that will encourage a maximum development of our environmental resources for society.

The goal of the Congress was to explore and develop environmental concepts and principles, methodology, and practice that will encourage maximum health and the orderly development of our environment for the needs of society. Team work among those who participated in the Congress, seasoned by intelligent determination and rugged individualism, may contribute materially to the American way of life."

The key note address was given by Dr. Henry F. Vaughn, President of the National Sanitation Foundation. Certain of Dr. Vaughn's remarks pointed very succinctly to the fact that the facilities and the resources of many groups working together are essential for the shaping of man's life to give it the fullest benefits. His remarks, in part were as follows:

"The total environmental health program, to be implemented successfully, must embrace the resources and facilities not only of the official health agencies, but of the voluntary agencies, industry, labor, and the public at large, all utilized on a selfplanned and self-integrated basis. The ultimate purpose in environmental health control is not merely the absence of a depth and breadth of filth; it is the fuller life to be had by all people, one in which all may benefit from the program, one in which we increasingly shape the environment for thriving, not merely surviving.

Man continually manipulates some factors in his environment to favor health conservation; the production of goods and services; and the satisfactions of his recreation, leisure, mental composure, and stability in a positive and fruitful way of life. A discordant environment is not conducive to one's internal peace. No one has an inherent right to do