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PARTICLE SIZE DISTRIBUTION ANALYSIS OF SOILS USING LASER DIFFRACTION

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Summary

The use of laser diffraction for the particle size distribution analysis of the sub-63 μm fraction of soil samples is described. Each sub-63 μm fraction was obtained from the wet sieving of 1500 mg of whole soil. Using similarity probabilities, the data obtained, when combined with other information from wet sieving and/or organic matter content, will enable the correct identity of a given soil sample with that of an unknown to be made. Although the sub-63 μm fraction can account for 450 mg or more of the total soil content, analyses of this fraction can be conducted on sample sizes as small as 100 mg.

Key words: Soils; Particle size analysis; Laser diffraction; Similarity probabilities

Introduction

Particle size analysis is an important aspect in the forensic evaluation of soil. Several authors [1—3] have demonstrated that such analyses can be used successfully to discriminate between soils from different geographical locations. The measurement of particle sizes in excess of 63 μm is routinely carried out using sieves. Although micromesh sieves can extend the lower range of sieving down to 5 μm, they are not easily used in practice. Since a substantial proportion of clay soils contain particles smaller than 63 μm, it is clear that sieving does not offer an efficient means of differentiation.

In this paper, the measurement and discrimination of different soil samples originating from the same locality is reported based on the laser diffraction technique. Unlike sieving, the method is quick and effortless. Very small quantities of soil can be analysed and particles as small as 1 μm measured and classified.

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Materials and Methods

Theory

When a spherical particle is illuminated by a parallel beam of monochromatic, coherent light, a diffraction pattern is formed. This diffraction pattern is large compared with the geometrical image of the particle and is superimposed on the geometrical image. If a lens is palced in the light path after the particle and a detector placed at the focal plane, then undiffracted light is focussed to a point on the axis and the diffracted light forms a pattern of rings around the central spot (Fig. 1). Movement of the particle does not cause movement of the diffraction pattern since light diffracted at an angle, θ, always give the same radical displacement in the focal plane irrespective of the position of the particle in the illuminating beam [4–6]. The intensity distribution is maximum at the centre of the diffraction pattern and oscillates with strongly decreasing amplitude as the radius increases (Fig. 2). The diameter of the diffraction pattern is inversely proportional to the particle diameter. By analysing the light energy contained within any ring in the focal plane, the size distribution can be inferred. In practice, diffraction patterns are summed for several measurements to obtain an integral measurement. A microprocessor then analyses this integral diffraction pattern to determine the size distribution. The analysis only holds when the particle diameter is greater than the wavelength of the illuminating radiation. Hence most instruments employing the He–Ne laser ($\lambda = 0.6328\ \mu$m) can measure size distributions down to 0.7 μm.

There are at least three commercially available particle size analysers which utilise the principle of laser diffraction: the Granulometer 715 (CILAS, France), the Malvern 3600E Type (Malvern Instruments, U.K.) and the Microtrac (Leeds and Northrup, U.S.A.).

In a previous paper [2], a statistical method for distinguishing dry-sieved soil samples was reported. This consisted of calculating a similarity probability between a blind and a control soil sample for each variable. The similarity probability is defined as

$$\Phi(z) = \int_{-\infty}^{z} \frac{1}{\sqrt{2\pi}} e^{-z^2/2}\, dz$$

where

$$z = \left| \frac{\bar{x} - \bar{b}}{\sigma/\sqrt{n}} \right| .$$

For n subsamples from each of the control and blind soil samples, $\bar{x}$ and $\bar{b}$ are the respective means and σ^2 is the variance. A computer algorithm exists for the calculation of the similarity probabilities. (Details are available

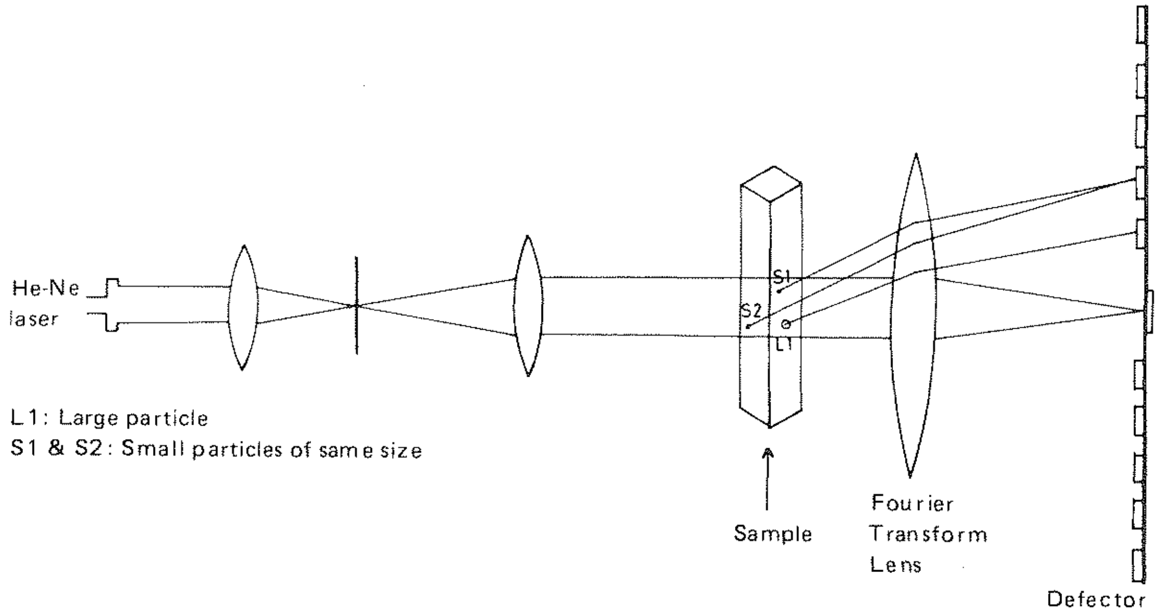

Fig. 1. Schematic optical configuration of the granulometer 715 laser instrument.

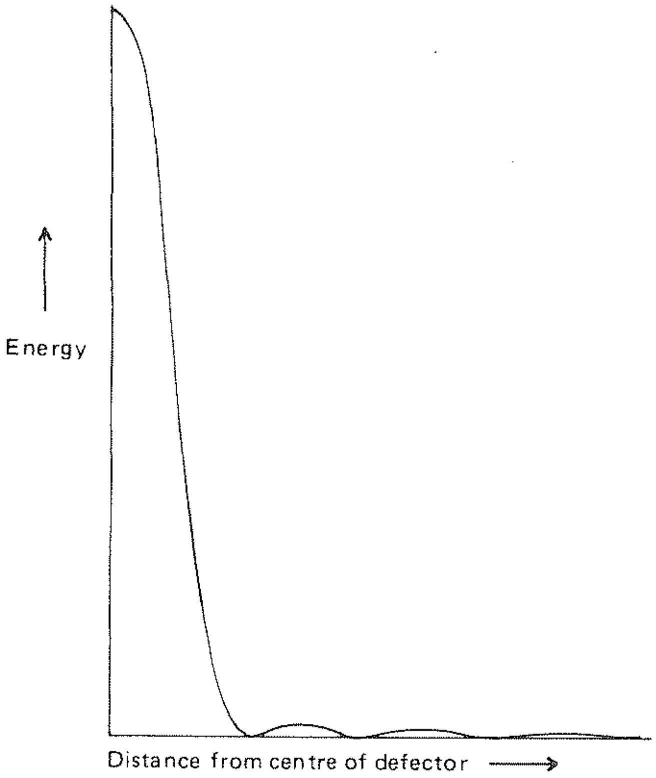

Fig. 2. The energy intensity distribution produced by a particle.

from the authors). The same method was applied to wet-sieved soil samples, with the sub-63 μm fraction being analysed using the Granulometer 715 laser diffraction instrument.

Soil Analysis

Three different studies were carried out using the laser instrument. The first consisted of repeatability investigations using one sub-63 μm fraction soil sample. Three soil sub-samples were prepared by dispersion of 400 mg of soil in 10 ml of distilled water using an ultrasonic bath (Sonicleaner Type 6443AE ultrasonics, Division of Dawe Instruments Ltd.). The sample holder of the granulometer was filled with 380 ml of filtered tap water (Millipore, HAWP 0.45 m). The water was sonicated, scanned and the background number of particles determined. Each soil sub-sample was thoroughly shaken and transferred into the sample holder. All soil particles were transferred by repeated rinsing using distilled water. Finally, the volume of suspension in the sample holder was adjusted to 400 ml using distilled water. The suspension was then stirred, sonicated and measured. Each sub-sample was measured six times giving rise to 18 particle size distributions. In addition, the same soil was analysed using masses of 300, 200, 100 and 50 mg.

In the second study, the accuracy and precision of the instrument and methods employed were assessed. The accuracy of the instrument was assessed using the latex standards supplied by Coulter Electronics (stated "sizes" 2.11, 18.1, 40.1 μm) and the BCR 70 Quartz standard (stated "size" 3 μm) (Coulter Electronics Ltd., Northwell Drive, Luton, Berks., U.K. Community Bureau of Reference, Directorate General XII, Commission of the European Communities, 200 Rue de la Loi, B1049 Brussels, Belgium). The quartz standards were prepared in a similar manner to the soil samples. The latex suspension standards were used as supplied. The concentration of these were unknown. Measurements were performed after vigorous shaking by addition of drops of suspension to the sample holder containing 400 ml of filtered water. In all cases, the first few drops were discarded prior to addition and all suspensions were stirred and sonicated before measurements. After each measurement was completed, the sample outlet was opened and the sample holder thoroughly washed. Background measurements of the filtered water usually indicated whether further washing was necessary before commencing the next measurement.

The precision of the instrument and of methods employed was tested using 400 mg of one sub-63 μm soil fraction on each of five consecutive days. Each day three sub-samples of the soil were taken and six consecutive readings obtained from each sub-sample.

Finally, the sub 63 μm fraction of 100 different soil samples were assessed using the laser instrument. These soil samples had been collected from 100 cells, each of area 3 m × 3 m in a 10 × 10 grid spanning a field and were intended to be representative of the surface soil within the field. Three sub-samples per soil sample were analysed and two measurements per sub-sample were determined thus giving rise to six particle size distributions per soil sample. Each soil sub-sample weighed 400 mg and was prepared as described in the repeatability investigations.

Results and Discussion

The results obtained for repeatability studies are presented in Tables 1 and 2, while the data obtained using different weights are presented in Table 3. These results indicate a high degree of repeatability. In Table 3, as the quantity of soil decreased, the repeatability remained virtually unchanged. However, at 50 mg, the results became inconsistent and repeatability was lost. This would suggest that soil samples could be examined quite confidently down to the 200-mg level (0.5 mg/ml) and possibly down to 100 mg. Some consideration was given to reducing the volume of filtered water below 400 ml. However in many such instruments the design increased the risk of introducing air bubbles into the circulating system when volumes of 250 ml and less were used. One instrument, the MALVERN 3600E, does have a smaller sampling cell (15 ml) and it may be possible using this cell to analyse smaller samples than the ones presently investigated.

A comparison using the latex standards between the size distributions obtained using the Coulter Counter (Model TA II) and the laser granulometer demonstrated little variation. In all cases, the laser granulometer exhibit a slight broadening (a shallow sigmoid curve). The 50% cumulative point, however, is fairly consistent (±2 μm) and thus the distribution is broadened only, not skew. Reasons for this difference could be due to

TABLE 1

RESULTS OF SIX CONSECUTIVE READINGS OF ONE SUB-SAMPLE OF A SOIL

Particle size (μm)	*Readings (%) vol.*					
<1.0	9.6	9.4	9.3	9.6	9.7	9.3
1—1.5	2.5	2.7	2.7	2.8	2.7	2.9
1.5—2.0	6.4	6.5	6.9	7.0	6.7	7.1
2—3.0	9.8	10.4	10.4	11.1	10.9	11.3
3—4.0	8.8	8.3	8.8	9.1	8.7	8.3
4—6.0	12.5	12.0	12.1	12.1	12.1	12.5
6—8.0	9.3	8.2	8.1	8.8	8.3	8.0
8—12.0	10.2	11.3	10.7	11.0	11.2	11.6
12—16.0	9.2	8.2	8.7	8.0	8.1	7.2
16—24.0	9.4	8.4	8.0	9.1	8.0	9.5
24—32.0	4.0	4.5	4.9	4.2	4.7	3.2
32—48.0	3.6	5.4	5.0	2.7	4.4	4.8
48—64.0	2.8	1.6	2.2	2.4	2.2	1.5
64—96.0	1.1	2.5	1.4	1.5	1.5	2.2
96—128.0	0	0	0	0	0	0
128—192.0	0	0	0	0	0	0

TABLE 2

TYPICAL RESULTS FOR REPEATABILITY OF TRIPLICATE ANALYSES OF A SOIL SAMPLE

Particle size (μm)	*Coefficient of variation (%) for six consecutive readings per sub-sample*		
	Sub-sample 1	*Sub-sample 2*	*Sub-sample 3*
<1.0	1.8	1.6	2.0
1—1.5	4.8	4.8	4.6
1.5—2.0	4.1	4.0	4.4
2—3.0	5.2	5.4	5.0
3—4.0	3.6	3.2	3.4
4—6.0	1.8	1.4	1.4
6—8.0	5.9	6.4	6.0
8—12.0	4.5	4.4	4.8
12—16.0	8.3	8.6	8.8
16—24.0	7.9	7.6	6.4
24—32.0	14.4	14.2	14.0
32—48.0	23.2	23.1	23.4
48—64.0	23.1	23.4	22.8
64—96.0	31.2	30.8	30.4
96—128.0	0	0	0
128—192.0	0	0	0

shape effects or methods of preparation and determination of size distributions. A typical result is given in Fig. 3.

Measurements to determine differences due to non-spherical particles were carried out using the BCR 70 standard. The BCR range of certified materials consist of particles of randomly shaped natural quartz certified by sedimentation. Despite the marked non-sphericity of these particles, the results obtained are in reasonable agreement with those obtained using sedimentation. These results are presented in Fig. 4.

TABLE 3

RESULTS SHOWING SECTIONS OF THE PARTICLE SIZE DISTRIBUTION OF THE SAME SOIL USING DIFFERENT WEIGHTS

Particle size (μm)	*400 mg*		*300 mg*		*200 mg*		*100 mg*		*50 mg*	
	Mean vol. %	*CV%*	*Mean vol. %*	*CV%*	*Mean vol. %*	*CV%*	*Mean vol. %*	*CV%*	*Mean vol. %*	*CV%*
1—1.5	2.7	4.8	2.7	4.5	2.9	4.2	2.8	3.9	2.4	14.2
3—4	8.7	3.6	8.9	2.3	8.3	4.6	9.1	9.2	7.8	14.9
2—16	8.2	8.3	7.0	7.4	6.8	14.2	8.4	5.7	4.8	49.0
24—32	4.3	14.4	6.0	19.3	4.3	8.3	5.5	19.2	6.2	54.5
64—96	1.7	31.2	0.4	75.0	1.6	52.9	0.0	0.0	0.6	141.7

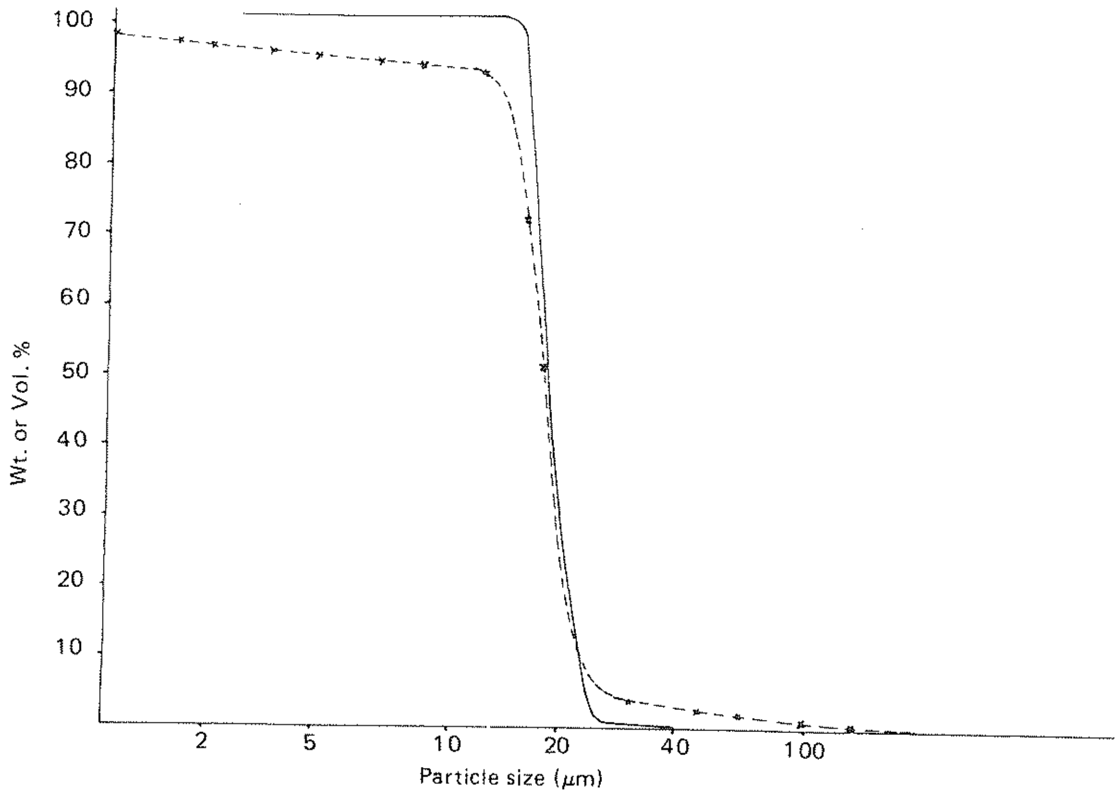

Fig. 3. Accuracy assessment of the Granulometer 715 Laser diffraction instrument using F24 latex standard. ——, Coulter Counter Model TA II (weight peak split = 19.3 μm); — — —, ILAS Granulometer 715 (50% cumulative point = 18.5 μm)

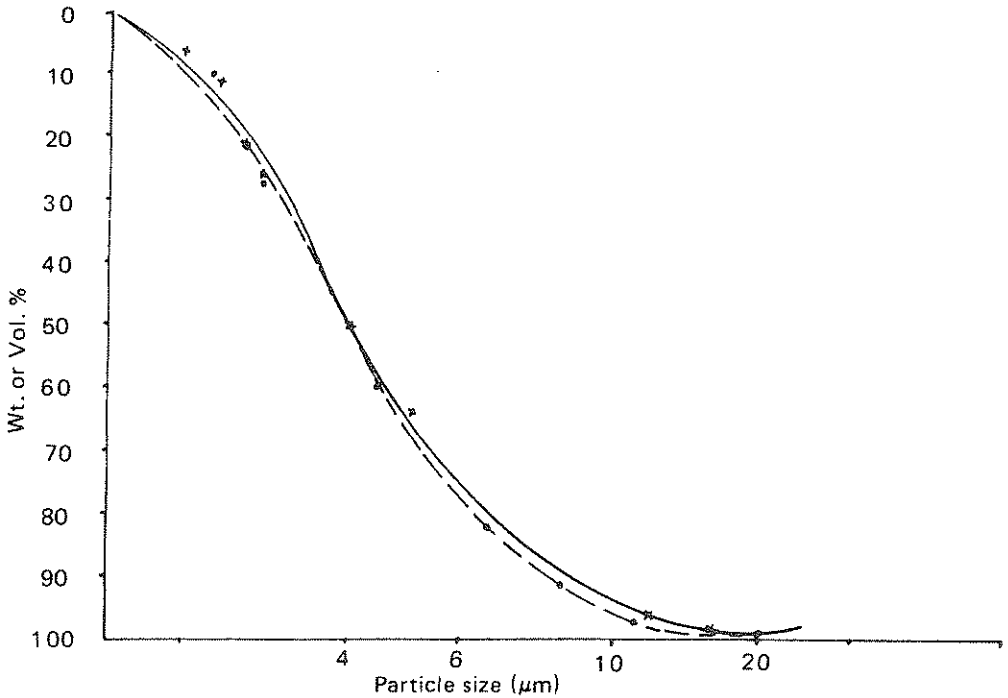

Fig. 4. Accuracy assessment of the Granulometer 715 Laser diffraction instrument using BCR 70 standard. ——, Coulter Granulometer 715; ———, BCR 70 certified (sedimentation).

TABLE 4

RESULTS SHOWING SECTIONS OF THE PARTICLE SIZE DISTRIBUTION OF THE SAME SOIL ANALYSED OVER A 5-DAY PERIOD

Particle size (μm)	*3 Sub-samples analysed on one day*		*3 Sub-samples analysed on five consecutive days (total = 15 sub-samples)*	
	Mean vol. %	*CV%*	*Mean vol. %*	*CV%*
1–1.5	2.7	4.8	2.8	4.6
3–4	8.7	3.6	8.6	3.9
12–16	8.2	8.3	8.2	8.0
24–32	4.3	14.4	4.1	13.8
64–96	1.7	31.2	1.9	32.4

The precision of the instrument and of methods employed was assessed using one soil sample. The results obtained are presented in Table 4 from which it can be seen that the analytical precision is in general very good.

The analysis of the sub-63 μm fraction of soil arising from each grid within the field gave rise to six particle size distributions. In order to maximise differences between samples and minimise differences within samples, the mean percentage of particles in each class was calculated. Furthermore, the number of class intervals were kept small by combining several classes. Thus the number of classes was reduced from 16 to 4. The classes consisted of 0–2 , 2–6, 6–16 and 16–192 μm thus giving classes with upper limits of 2, 6, 16 and 192 μm.

The statistical method previously described [2] was employed. However, on this occasion, the variables median and modal class intervals were replaced by the percentages within each class of the particle size distribution for each soil while the percentage of organic matter remained unchanged.

The data obtained from the 100 cells formed the control data set. Blind samples were similarly examined using the four classes described above and the results compared with those of the control data set. In all, eight types of comparisons were made. These were as follows: (i) The particle size distribution obtained using the laser granulometer only, i.e. comparing the four classes of the blinds with the four classes from each of the 100 cells. This is a direct comparison and the data were not normalised. (ii) The particle size distribution obtained using the laser granulometer in a normalised form. This is similar to (i) except that the data were normalised to take into account the original weight of the sub-63 μm fraction obtained from wet sieving. (iii) The combined particle size distributions of wet sieving and laser diffraction technique. In this case, the laser granulometer data were not normalised. (iv) Same as in (iii) but the laser diffraction data were normalised. (v) The particle size distribution from the laser diffraction technique not normalised and the percentage of organic matter. (vi) Same as in (v) but the laser diffraction data were normalised. (vii) A combination of particle size distributions from both wet sieving and laser diffraction technique and the percentage of organic matter. The laser diffraction data were not normalised. (viii) Same as (vii) but the laser diffraction data were normalised. The normalisation procedure is illustrated for a typical particle size in the Appendix.

It was observed that the repeatability of determinations diminishes as the particle size increased hence the necessity for combining several intervals

TABLE 5

PREDICTION OF ORIGIN OF BLIND SOIL SAMPLES USING UNNORMALISED AND NORMALISED PARTICLE SIZE DISTRIBUTIONS FROM LASER DIFFRACTION ANALYSIS

The similarity probability associated with correct origin of the blind soil sample is underlined.

Blind soil sample	*Unnormalised*					*Normalised*				
	Five highest similarity probabilities to control soil samples					*Five highest similarity probabilities to control soil samples*				
	1st	*2nd*	*3rd*	*4th*	*5th*	*1st*	*2nd*	*3rd*	*4th*	*5th*
1	0.60	0.57	0.33	0.26	0.20	0.43	0.42	0.23	0.20	0.12
2	0.54	0.008	0.005	0.003	0.002	0.78	0.20	0.19	0.17	0.08
3	0.77	0.39	0.16	0.11	0.10	0.53	0.36	0.25	0.23	0.19
4	0.61	0.005	0.004	0.002	0.001	0.65	0.60	0.36	0.33	0.28
5	0.79	0.34	0.28	0.27	0.26	0.57	0.51	0.50	0.33	0.30
6	0.58	0.41	0.38	0.19	0.08	0.55	0.53	0.31	0.30	0.28
7	0.50	0.30	0.26	0.25	0.23	0.66	0.49	0.44	0.41	0.46
8	0.61	0.32	0.30	0.15	0.10	0.78	0.68	0.31	0.30	0.27
9	0.58	0.53	0.23	0.21	0.16	0.78	0.37	0.34	0.19	0.15
10	0.66	0.02	0.009	0.008	0.006	0.88	0.18	0.09	0.07	0.04
11	0.43	0.008	0.007	0.004	0.002	0.63	0.43	0.39	0.38	0.37
12	0.37	0.36	0.31	0.22	0.03	0.70	0.63	0.48	0.34	0.23
13	0.82	0.72	0.52	0.44	0.41	0.78	0.61	0.50	0.45	0.41
14	0.64	0.51	0.35	0.31	0.25	0.75	0.12	0.11	0.06	0.05
15	0.64	0.17	0.13	0.08	0.05	0.53	0.32	0.13	0.12	0.11
16	0.59	0.28	0.12	0.11	0.10	0.82	0.27	0.13	0.09	0.07
17	0.62	0.60	0.55	0.51	0.50	0.78	0.61	0.50	0.46	0.45
18	0.56	0.41	0.29	0.27	0.15	0.55	0.52	0.50	0.49	0.33
19	0.33	0.27	0.14	0.07	0.06	0.78	0.75	0.23	0.22	0.19
20	0.75	0.45	0.32	0.28	0.27	0.55	0.54	0.50	0.48	0.46
Summary										
1st					12					15
2nd					6					2
3rd					1					/
Total					19					17

to form classes. This variation in the upper classes also precluded comparison of entire size distributions without reduction of classes.

Typical prediction results obtained using the particle size distribution from the laser diffraction technique only correctly identified 12 out of 20 blind soil sample origins in the unnormalised form (Table 5). When the data were normalised, the prediction of correct values increased from 12 to 15. This is to be expected since normalising the distribution of the sub-63 μm fraction scales the distribution according to its contribution to the total soil mass. The additional information enhances discrimination. The various combinations increased the correct prediction either to 19 or 20 (Table 6). The correct identification of the sixth blind soil sample was particularly difficult and it was only confidently correctly identified when normalised particle size distributions were considered. In contrast the eighth blind soil sample was incorrectly predicted on two occasions in which normalised particle size distributions were employed. It is interesting to note that a combination of particle size distributions of both laser and wet sieving had the same performance as the percentage of organic matter and laser particle size distribution. However, the latter displayed much higher probabilities of similarity without loss of confidence in prediction and the analytical time was considerably less.

The greater degree of discrimination achieved from the combination of wet sieving and laser diffraction technique indicate that both procedures are fairly compatible. The combinations of percentage organic matter and particle size distributions from both wet sieving and laser diffraction yielded the maximum degree of confident predictions. Both the normalised and unnormalised laser diffraction data performed well and all blind soil sample origins were correctly predicted.

The authors realise that it would be better if the soil could be analysed directly on the granulometer without prior fractionation by sieving. Indeed,

TABLE 6

A SUMMARY OF PREDICTION OF ORIGIN OF BLIND SOIL SAMPLES BASED ON LASER DIFFRACTION ANALYSIS ONLY (I), LASER DIFFRACTION ANALYSIS AND WET SIEVING (II), LASER DIFFRACTION ANALYSIS AND PERCENTAGE ORGANIC MATTER (III), LASER DIFFRACTION ANALYSIS AND WET SIEVING AND PERCENTAGE ORGANIC MATTER (IV)

a, unnormalised laser diffraction data; *b*, normalised laser diffraction data.

Score	*I*		*II*		*III*		*IV*	
	a	*b*	*a*	*b*	*a*	*b*	*a*	*b*
1st (correct prediction)	12	15	19	20	19	19	20	20
2nd	6	2	/	/	/	1	/	/
3rd	1	/	/	/	/	/	/	/
Total out of 20	19	17	19	20	19	20	20	20

in the authors experience, none of the particle sizing instruments available on the market at the present time is capable of achieving this aim. The main limitations are the geometry of the instruments and problems of sedimentation as the particle sizes approach 2 mm.

Conclusion

The results obtained suggest that a combination of organic matter and laser diffraction analysis could be usefully employed in the routine analysis of soils in forensic work. Using such a combination, the analytical time is reduced while prediction success remains high.

Acknowledgement

The authors would like to acknowledge the cooperation of Coulter Electronics Ltd., Malvern Instruments U.K. and DELCITA Ltd.

Appendix

Wet sieving of a typical soil sample provides the following particle size distribution

>1 mm	*1 mm—500 μm*	*500—250 μm*	*250—90 μm*	*90—63 μm*	*<63 μm*	*Total*
1.2%	2.6%	7.8%	47.6%	6.2%	34.6%	100%

Analysis of the sub-63 m fraction using the laser granulometer leads to a particle size distribution with 16 class intervals:

0—1 μm	*1—1.5 μm*	*1.5—2 μm*	*2—3 μm*	*3—4 μm*	*4—6 μm*	*6—8 μm*	*8—12 μm*	*12—16 μm*	*16—24 μm*	*24—32 μm*	*32—48 μm*	*48—64 μm*	*64—96 μm*	*96—128 μm*	*128—196 μm*	*Total*
10.7%	2.7%	7.8%	11.3%	10.2%	12.8%	10.9%	12.2%	7.2%	5.8%	5.9%	0.5%	2.0%	0%	0%	0%	100%

After combining classes the unnormalised particle size distribution will become

0—2 μm	*2—6 μm*	*6—16 μm*	*16—192 μm*	*Total*
21.2%	34.3%	30.3%	14.2%	100%

and scaling these percentages by 0.346 the normalised particle size distribution is derived:

0—2 μm	*2—6 μm*	*6—16 μm*	*16—192 μm*	*Total*
7.3%	11.9%	10.5%	4.9%	34.6%

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