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Grain size analyzers: results of an intercomparison study

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Abstract:	<p>In sciences involving soil and sediment, particle size distribution (PSD) has been one of the parameters given most attention over the past few decades. Formerly measured by sieving and sedimentation techniques, it is nowadays routinely characterized by the laser diffraction method (LDM). Many manufacturers develop particle size analyzers using LDM, but each device is characterized by specific parameters that can lead to different PSD. At the Rhône Sediment Observatory, suspended particulate matter collected along the Rhône River are analyzed for PSD by four different LDM devices. Analyses were conducted on certified materials and sediment samples for each device. The tests highlighted the difficulty of accurately characterizing PSD, even in the case of certified materials. First, differences observed for a specific device were linked to the heterogeneity observed in the subsamples due to the presence of organic materials such as tree leaves. Second, the difference regarding the certified materials was linked to the laser diffraction method which leads in some cases to underestimating clay content and sand. Third, the main difference observed between the devices was linked to sonication. The results demonstrate that its power is rarely investigated and that it has a considerable impact when used. However, despite significant differences, the trend was similar for each device, with accurate characterizations of the main modal class in most cases. Thus, in the absence of exact knowledge of parameters such as sonication power and pump speed, it is recommended to compare only the trends of the results obtained from the different devices.</p>
Keywords:	Particle size distribution; laser diffraction; ultrasound sonication; suspended particulate matter; Sediment
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1 Grain size analyzers: results of an intercomparison study

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8 ABSTRACT

9 In sciences involving soil and sediment, particle size distribution (PSD) has been one of the parameters
10 given most attention over the past few decades. Formerly measured by sieving and sedimentation
11 techniques, it is nowadays routinely characterized by the laser diffraction method (LDM). Many
12 manufacturers develop particle size analyzers using LDM, but each device is characterized by specific
13 parameters that can lead to different PSD. At the Rhône Sediment Observatory, suspended particulate
14 matter collected along the Rhône River are analyzed for PSD by four different LDM devices. Analyses
15 were conducted on certified materials and sediment samples for each device. The tests highlighted the
16 difficulty of accurately characterizing PSD, even in the case of certified materials. First, differences
17 observed for a specific device were linked to the heterogeneity observed in the subsamples due to the
18 presence of organic materials such as tree leaves. Second, the difference regarding the certified
19 materials was linked to the laser diffraction method which leads in some cases to underestimating clay
20 content and sand. Third, the main difference observed between the devices was linked to sonication.
21 The results demonstrate that its power is rarely investigated and that it has a considerable impact
22 when used. However, despite significant differences, the trend was similar for each device, with
23 accurate characterizations of the main modal class in most cases. Thus, in the absence of exact

1 24 knowledge of parameters such as sonication power and pump speed, it is recommended to compare
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4 25 only the trends of the results obtained from the different devices.
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6

7 26 **Key word**

8
9

10 27 Particle size distribution, laser diffraction, ultrasound sonication, suspended particulate matter,
11
12 28 sediment
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14

15 29 **Article Highlights**

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- 18
19 30 • Differences on the particle size distribution were always observed even with the certified
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21 31 samples.
- 22
23 32 • The use of ultrasound sonication increases or triggers differences in the particle size
24
25 33 distribution.
- 26
27 34 • The lower is the proportion of a textural class (clay/silt/sand), the higher is the relative
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29 35 difference.
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36 37 **1. INTRODUCTION**

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39 38 Much research has been conducted over recent decades to understand the dynamics and
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41 39 behavior of sediment and suspended particles in rivers. Understanding the processes that affect the
42
43 40 transport of these particles is crucial for ecological, economic and societal purposes, as the world's
44
45 41 rivers are becoming more anthropized [1,2]. The investigation of their nature and origins has now
46
47 42 become essential [3]. Among the different parameters used to characterize particles when studying
48
49 43 their behavior (e.g., mineralogy, shape, color), particle size remains one of those most investigated
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51 44 [4,5] and it is often an issue in geoscience studies.
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1 46 The characterization of particle size distribution (PSD) can be performed using several
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4 47 techniques such as sieving, by pipette based on Stokes diameters, and laser diffraction [6–8]. The
5
6 48 choice of the optimum grain-size analysis technique depends on the aim of the study and the type of
7
8 49 sediment [8]. For loamy sediments, laser diffraction instruments produced the best results for the
9
10 50 various criteria [8]. The main differences observed between the laser diffraction method (LDM) and
11
12 51 the other methods are related to particle morphology and clay mineralogy [5,9]. Clay content can be
13
14 52 underestimated in suspended sediments [10].
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17
18 53 The LDM is widely used on sediment and suspended particles as it covers a wide range of grain
19
20 54 sizes and requires short analysis time [5,8,11]. For example, particle size analyses are processed on
21
22 55 cores recovered from marine, estuarine and coastal zones for paleoceanographic and sea level
23
24 56 reconstructions [12–15] and paleo-environmental questions associated with human history [16–18].
25
26 57 PSD is now commonly used to improve soil and sediment fingerprinting [19] and to investigate the
27
28 58 concentration of trace metals in sediments. Apart from the geosciences, many domains such as
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30 59 forensic studies [20], pharmaceuticals and industry use this technology and require greater detail on
31
32 60 the data acquired from different lasers.
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36
37 61 Nowadays, many devices from different manufacturers are used to measure PSD by LDM, with
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39 62 different designs and software applications, leading to slightly different distributions [8,21].
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41 63 Bieganski et al. (2018) suggested that about 24 factors can influence the PSD results and lead to
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43 64 uncertainty or error. Differences arise as all these parameters vary between devices and laboratories,
44
45 65 since resolving the issue of the homogeneous resuspension of sediment is a complex [22]. For example,
46
47 66 it has been demonstrated that the use of ultrasonic power can destroy aluminum and silica particles
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49 67 due to the dissolution mechanism [23] and that the addition of oxidizing solutions to remove organic
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51 68 matter can also alter clay particles such as vermiculite [24]. However, intercomparisons of PSD
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53 69 measurements were performed more between the different PSD techniques described above than
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55 70 between different LDM devices. Nevertheless, differences were observed for the PSD and the
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1 71 proportion of clay measured by the devices of different manufacturers [8,10]. Unfortunately, in the
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4 72 study conducted by Goossens (2008) on loamy sediments (sieved at 90 μ m), the impact of sand
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6 73 particles and the use of ultrasound sonication (US) has not been addressed.
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10
11 75 To investigate PSD, most of these studies used statistical parameters such as mean, median
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13 76 and standard deviation to summarize PSD or Kurtosis and the Skewness index to describe the variability
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15 77 of PSD [25,26]. However, such parameters might not be sufficient to describe PSD in a non-lognormal
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17
18 78 or multimodal distribution [27,28]. Suspended sediment PSDs are mainly multimodal and the location
19
20 79 of modes can be related to the transportation processes and the origins of the particles [28]. Also,
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22
23 80 modes can be used to investigate the dynamics of sediment deposition [29]. Several mathematical
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25 81 solutions were developed to summarize and describe raw PSD such as two or five parameters Log-
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27 82 normal equations [30,31], demodulation [32] and surface plots [28]. Unfortunately, these approaches
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29
30 83 can be complex for people unfamiliar with advanced mathematics. This may explain why scientific
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32 84 publications and technical reports are still published with common parameters [33–35]. Also,
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35 85 depending on the study context, it is not necessary to rely on advanced solutions as common
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37 86 parameters may be sufficient for investigating PSD.
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42 88 Since 2009, within the context of the Rhône Sediment Observatory (OSR) program, scientists
43
44 89 have studied the transport of particles and their associated contaminants along the entire course of
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46 90 the Rhône River (from Lake Geneva to the Mediterranean Sea), the main source of sediments in the
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48
49 91 northern Mediterranean [36]. All these investigations are well-documented due to the presence of
50
51 92 several monitoring stations located along the Rhône River and in its main tributaries [37]. Particle
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53 93 samples for chemical analysis are routinely collected. Analyses of PSD are systematically conducted
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56 94 not only to study the transport of particles but also to improve the interpretation of contaminant
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58 95 concentrations and they can also be used for transport modelling. For example, the use of different
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1 96 sampling techniques for suspended particle sampling (particle trap and continuous flow
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4 97 centrifugation) leads to a bias in particle size [38]. In the case of large environmental monitoring
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6 98 networks such as the OSR program, PSD analyses are processed by an array of laboratory lasers.
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11 100 It is therefore crucial to evaluate the potential biases induced by the use of different devices
12
13 101 in the PSD results. The Intercomparison was conducted by studying the differences observed on
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15 102 common parameters, such as the proportion of clay/silt/sand resulting from the analysis - with and
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17
18 103 without US - of reference standards and samples collected in the field. The objective was not to define
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20 104 which device is the most accurate but quantify the relative difference that might be observed and how
21
22 105 the results can be interpreted and compared. Answering this question is crucial for the organizations
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25 106 responsible for river surveys, as we provide tools for comparisons between all the PSDs analyzed within
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27 107 the OSR.
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31 108 **2. MATERIALS AND METHOD**

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34 109 **2.1. Particle size analyzers**

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37 110 In the OSR program, grain size is assessed using the following devices: a Beckman Coulter LS
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40 111 13 320 (Beckman Coulter, Fullerton, CA, USA), a Cilas 1190L (Cilas Company Ltd., Orléans, France), a
41
42 112 Malvern Mastersizer 2000 (Malvern Instruments Ltd., Malvern, UK) and a Sequoia LISST-Portable|XR
43
44
45 113 (Sequoia Scientific, Bellevue, WA, USA). These devices will be referred to as Coulter, Cilas, Malvern and
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47 114 Portable, respectively, for the rest of the article. As expected, the main characteristics of these devices
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49 115 differ (Table 1).
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53 116 **2.2. Protocol of intercomparison**

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56 117 The intercomparison of the devices was conducted using certified reference materials (CRMs) and
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58 118 natural samples from the Rhône River and its tributaries:
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- 1 119 • Measurements were performed on CRMs to investigate the accuracy of the different devices
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4 120 and highlight the differences between them. Three CRMs were selected in order to accurately
5
6 121 represent the ranges of suspended particle size, mostly silt-size [38], observed in the Rhône
7
8 122 River: BCR-066 (0.35-3.5 μm) and BCR-067 (2.4-32 μm) [39,40]; SRM1003c (25-50 μm) [41].
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11 123
12
13 124 • Artificial multimodal distributions were created to assess the capacity of the device to
14
15 125 efficiently characterize the different modes. To do this, three mixtures of the 3 CRMs were
16
17 126 prepared by weighing masses in order to reach the following proportions (BCR-066/BCR-
18
19 127 067/SRM1003c in %): Mix_A: 25/50/25, Mix_B: 50/25/25 and Mix_C: 25/25/50.
20
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23 128
24
25 129 • As CRMs are mostly composed with glass-quartz particles, they are not qualitatively
26
27 130 representative of the suspended particulate matter (SPM) encountered in the Rhône River
28
29 131 [42], especially regarding their aggregation properties. Therefore, an intercomparison was also
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31 132 conducted on the SPM samples collected in the observatory. Four samples were collected
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33 133 using different methods at different stations to characterize the diversity observed in this
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35 134 observatory:
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39 135 ○ Arc River (45°33'44.6"N 6°12'22.8"E): lag deposits that settled during a flood event were
40
41 136 collected manually with a plastic spatula. This sediment was sieved in order to keep only a
42
43 137 sandy fraction between 100 μm and 2 mm.
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45
46 138 ○ Azergues River (45°56'11.5"N 4°43'24.6"E): bottom sediment collected by a sediment
47
48 139 dredge (Eckman),
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50
51 140 ○ Jons monitoring station in the middle Rhône River (45°48'42.3"N 5°05'09.2"E): SPM
52
53 141 collected by particle trap (Masson et al., 2018) during baseflow in March 2017,
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1 142 ○ Barcarin station in the downstream Rhône River (43°25'12.5"N 4°44'50.7"E): SPM collected
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4 143 using a Niskin bottle 1m above the surface during a flood event in November 2017. This
5
6 144 was the only liquid sample comprising SPM in Rhône River water.
7

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11 146 • The last step was conducted on a mixture of the samples collected in the Azergues and Arc
12
13 147 Rivers to control the proportion of modes and the impact of coarse particles. The mixtures
14
15 148 were prepared by weighing dry particles in the following proportions (Azergues/Arc in %):
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17 149 Mix_1: 95/5, Mix_2: 90/10, Mix_3: 85/15, Mix_4: 80/20 and Mix_5: 70/30.
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22 151 Each sample was homogenized then split into 4 subsamples before being sent to the different
23
24 152 laboratories for PSD analysis. For each sample, measurements were repeated on 2 or 3 distinct aliquots
25
26 153 depending on the quantity available. Prior to measurement, the subsamples were dispersed in water
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28 154 according to Arvaniti et al. (2014).
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32 155
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34 156 The Mie model was used when possible as the optimal model, as it is more useful in samples
35
36 157 characterized by fine silt and clay content [43]. The Coulter device was the only device in this
37
38 158 intercomparison that uses both models. Depending on the device, the refraction indexes were chosen
39
40 159 to be similar and close to silica as the variation of this index can result in different PSDs [11]. The other
41
42 160 parameters of the analysis method (Table 1), such as stirrer time and speed, pump speed adapted to
43
44 161 the volume of the bowl, ultrasonic duration, and measurement duration, were determined by the
45
46 162 users to obtain the best possible results (these parameters were optimized for each device with
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48 163 particulate samples collected in the OSR program).
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1 165 After a series of tests on the certified standards (data not presented), measurements were
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4 166 performed by conforming to the best obscuration range given by the manufacturer (Table 1). The
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6 167 results of the tests show differences occurring outside those ranges.

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11 169 Each subsample was analyzed at least three times without US then three times with US to
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13 170 investigate the repeatability of the measurements and the effect of sonication on the natural samples.

16 171 **2.3. Comparison parameters**

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19 172 One of the main difficulties in conducting an intercomparison on PSD is the need to use
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22 173 synthetic parameters, as the raw distributions cannot be directly compared due to channel
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24 174 inhomogeneity. In the following study, intercomparison will be conducted on several of the
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26
27 175 parameters used most frequently in environmental studies and especially in the OSR program:

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29 176 • The median (d50) that synthesizes the distribution by one factor. However, this parameter is
30
31 177 not the most relevant in the case of multimodal distribution, as is mostly encountered in rivers
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33
34 178 [28,31].
- 35
36 179 • The proportions of clay (<1.95 μm), silt (1.95 – 62.5 μm) and sand (62.5 – 2000 μm) are
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38
39 180 commonly used for normalization, as contaminants are mostly fixed on clay and silt. Again, this
40
41 181 does not give information on distribution shape, especially in the case of multimodality [28]
42
43 182 and the range of the classes can vary depending on the scale used [26,44]. This separation into
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45
46 183 3 classes will be referred to as “simple classes” for the rest of the text.
- 47
48 184 • The proportions of detailed classes follow the Doeglas scale [45]: clay (<1.95 μm), very fine silt
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50 185 (V_F_Silt: 1.95 – 3.91 μm), fine silt (F_Silt: 3.91 – 7.81 μm), medium silt (M_Silt: 7.81 –
51
52 186 15.63 μm), coarse silt (C_Silt: 15.63 – 31.25 μm), very coarse silt (V_C_Silt: 31.25 – 62.5 μm),
53
54
55 187 very fine sand (V_F_Sand: 62.5 – 125 μm), fine sand (F_Sand: 125 – 250 μm), medium sand
56
57
58 188 (M_Sand: 250 - 500 μm), coarse sand (C_Sand: 500 – 1000 μm), very coarse sand (V_C_Sand:

189 1000 – 2000 μm). It allows a good and comparable description of the distribution but the higher
190 classes are less-well adapted for statistical analyses than previous parameters. This separation
191 will be referred to as “detailed classes” for the rest of the text.

2.4. Specific case of the LISST Portable|XR device

192 Due to its lower range of PSD than the other devices (Table 1), and as it cannot characterize
193 the particles with a diameter lower than 0.34 μm , this LDM device is sensitive to rising tails [46–48].
194 Rising tails can be linked to the refractive index for small particles [49] or shape effects [12], and most
195 studies recommend removing the lower classes and keeping the larger size classes that are consistent
196 [46]. Thus, as rising tails were observable for almost all the samples, the first 5 classes ($< 0.72\mu\text{m}$) of
197 all the samples measured by the Portable were removed.

198 However, for this study, both raw and corrected PSDs were used to investigate the impact of
199 rising tails on the PSD. Corrected PSD will be referred to as “Portable_C” for the rest of the text. Only
200 the results of Portable_C will be presented in the graph for the sake of clarity and the results without
201 correction are available in the Electronic Supplementary Material. Also, the presence of bubbles during
202 the analysis led to the presence of modes in the higher classes. This was especially relevant for the
203 CRMs that should not be characterized by such coarse particles. Corrections were performed on
204 references (BCR-066 & BCR-067) and a mixture of references (Mix_C), by deleting the coarser classes.

3. RESULTS

3.1. Accuracy of the devices

205 The first part entailed measuring the CRMs. For this step, the use of US had a negligible impact
206 as CRMs are characterized by non-cohesive materials (quartz). Only the results with US will be
207 presented in this chapter.

1 211
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4 212 First, the results obtained by each device from the different aliquots of the CRMs (Fig. 1) were
5
6 213 characterized by low standard deviations (SD) except for mixture C (Mix_C) measured by the Coulter
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8 214 (high SD on clay and coarse silt). For this device with this mixture, the mean d50 ranged from 2.4 ± 0.1
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10
11 215 μm to $6.2 \pm 1.4 \mu\text{m}$ for the three aliquots measured and the mean proportion of clay ranged from 27
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13 216 $\pm 2 \%$ to $42 \pm 1 \%$ (Electronic Supplementary Material).

14 217
15
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18 218 Second, the values measured by the devices were compared to the certified values (Table 2 –
19
20 219 Fig. 1). For the finest CRM (BCR-066), all the devices overestimated the d50 (Table 2) and
21
22 220 underestimated the proportion of clay (Fig. 1). The absolute difference ranged from 13 % to 34% for
23
24
25 221 the clay proportion, which represented 15% to 40% of the relative difference. For BCR-067 and
26
27 222 SRM1003c, the measured d50s were similar to the certified parameters with a relative difference lower
28
29
30 223 than 20% (Table 2). For the simple classes, the relative difference was also lower than 20% but each
31
32 224 device measured particles in classes in which the CRMs should not be observed. Indeed, all the PSDs
33
34
35 225 measured by the devices were more spread out with a low amount (<10%) of particles observed in the
36
37 226 finer and coarser classes than the expected classes of the CRMs. This difference was clearly observed
38
39 227 on the detailed classes (Fig. 1) and the relative difference exceeded 20% for all the devices on the
40
41
42 228 classes certified by the BCR-067 (from 25% to 175%). For the SRM1003c, the difference exceeded 10%
43
44 229 only once with the proportion of very coarse silt measured by the Portable_C device (38%) (Fig. 1).
45
46 230 Thus, all the devices underestimated the proportion of particle in the modal class (Fig. 1).

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51 232 The intercomparison of the devices was conducted on those results. The d50s measured on
52
53 233 the CRMs by the four devices were similar whereas significant differences were observed between the
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56 234 devices for the mixtures (Table 2). For instance, the d50 measured by the Cilas for mixture C ($11.4 \pm$
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58 235 $0.5 \mu\text{m}$) was more than twice as high as the d50 measured by the Coulter ($4.6 \pm 2.3 \mu\text{m}$). Modal classes
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1 236 of the CRMs (unimodal) and the mixtures (bimodal) were accurately characterized by all the devices
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4 237 with the exception of mixture B measured by the Coulter. For this mixture, the PSD measured by the
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6 238 Coulter was unimodal and no particles coarser than medium silt was measured whereas it represented
7
8 239 more than 20% for the other device (Fig. 1). Thus, the proportion of clay measured by the Coulter was
9
10 240 significantly higher than the other devices for this mixture (54% vs 30% to 33% – Fig. 1). For the other
11
12 241 samples, the proportions measured by the devices were similar (<10% absolute difference) except for
13
14 242 the finest CRM (BCR-066). The proportions of clay measured by the Cilas and the Coulter on this CRM
15
16 243 (74% and 70%, respectively) were significantly higher than the two other devices (53% and 58% - Fig.
17
18 244 1).

24 245 **3.2. Natural samples**

25
26 246 The first natural sample, collected directly in the water flow with a Niskin bottle at Barcarin,
27
28 247 was mostly silt sized with a non-negligible proportion of clay (Fig. 2 – Table 3). The PSD measured by
29
30 248 the different devices were similar, so as for the d50 (Table 4). A difference was observed only for the
31
32 249 Coulter which measured a significant and higher proportion of clay (+11-13%), as observed previously
33
34 250 for Mixtures B and C (Fig. 1). The d50 measured by the Coulter was strictly lower than the other devices
35
36 251 (Table 4). Sonication was used only on the Cilas and had a negligible impact on this sample (Electronic
37
38 252 Supplementary Material).

39
40
41 253
42
43 254 The sample collected by particle trap at Jons was coarser than the SPM from Barcarin (Fig. 2),
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45 255 with a significant proportion of sand (up to 27 % - Table 3). The differences between the devices
46
47 256 observed on the measures performed without US remained lower than 10% (Fig. 2) and the d50s were
48
49 257 similar (Table 4). As observed previously, the d50 and the clay content measured by the Coulter were
50
51 258 respectively lower and higher than the results obtained by the two other devices. The use of US led to
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53 259 significant change in the PSD for the Portable_C (from 20% to 3% of very fine sand), while PSD
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1 260 remained similar for the Coulter and Cilas. Thus, the proportion of silt was significantly higher (+20%)
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4 261 for the Portable_C with sonication than for the other devices (Table 3).
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6 262
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8 263 The bottom sediment collected in the Azergues River was characterized by a mix of silt and
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10
11 264 sand particles (Table 3), with the main mode observed between the coarse silt and the very fine sand
12
13 265 (Fig. 2). In absence of US, the trend was similar for each device and only the Portable_C was
14
15 266 characterized by a higher proportion in the silted classes and a lower proportion of sand. The
16
17
18 267 proportion of silt measured by Portable_C ($72.6 \pm 9.9\%$) was significantly higher than the other devices
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20 268 (from 44.2 to 51.3 %) but characterized by a high standard deviation. The use of US on this sample led
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22
23 269 to a significant variation for all the devices and to an increase of silt particles (+ 6% to 37%) for all the
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25 270 devices (Table 3). However, this variation was greater for the Cilas (Fig. 2) (+37% of silt) than for the
26
27 271 three other devices (+6-16%). The proportions of the classes after US were different and separated
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30 272 into two groups: Cilas and Portable_C with almost 90% silt, with coarse silt as the modal class, and
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32 273 Coulter and Malvern characterized by a large proportion of sand (35-43%) and very coarse silt as the
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34 274 modal class. Once again, the differences observed for the d50 were significant whether US was used
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37 275 or not (Table 4). For instance, d50s measured by the Malvern with and without US (respectively $40.0 \pm$
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39 276 $5.4 \mu\text{m}$ and $71.6 \pm 5.8 \mu\text{m}$) were two times higher than on Portable_C ($20.9 \pm 1.8 \mu\text{m}$ and 37.9 ± 9.1
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41 277 μm).
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46 279 The last sample, the lag deposit collected in the Arc River, was characterized by fine sands (Fig.
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49 280 2). For the four devices, the proportion of sand was higher than 80% and reached 100% for the Malvern
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51 281 (Table 3). The Coulter and Portable_C distributions were characterized by higher uncertainty intervals.
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53 282 These devices also measured a higher mean proportion of silt particles compared to the other two
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56 283 devices, resulting in a significantly lower d50 (Table 4). However, despite this difference, the trend was
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58 284 similar between the devices with the maximum proportion observed in the fine sand class (Fig. 2). The
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1 285 PSDs measured by the Malvern were less spread than for the other device while the proportion of sand
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4 286 classes measured by the Cilas was more spread with a lower proportion in the modal class (F_Sand)
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6 287 and a higher proportion in the coarser sand class (M_Sand). Finally, as this sample was mostly sandy,
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8 288 US had a negligible impact on the distribution, as observed for the d50 (Table 4).
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12 289 **3.3. Mixtures of natural samples**

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15 290 Measures were then performed on the mixtures of the Arc and Azergues samples. As expected,
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17 291 the increase of the proportion of the coarser sample (from mixture 1 to 5) led to a higher d50 (Fig. 3)
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19 292 and higher proportions in the coarser classes (Electronic Supplementary Material) whatever sonication
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21 293 was used or not. However, breaks were observed for the Coulter (Mix_1), the Malvern (Mix_4 and
22 294 Mix_5) and the Portable_C (Mix_3 and Mix__5).
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29 296 The differences observed between the devices were similar to the difference observed for the
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31 297 Azergues sample as it represented more than 70% of the mixture. Thus, the parameters were different
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33 298 both with and without US (Fig. 4). Significant differences were observable for d50 and with the
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35 299 proportion of silt and sand, whereas the proportion of clay remained lower than 5% for each device
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38 300 (Fig. 4). The position of the main mode was also different, both without (simple and detailed classes)
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40 301 and with US (detailed classes). Using US led to the strong variation for simple classes measured by the
41 302 Cilas and the Portable_C (+35% silt) whereas it had no effect for the Coulter (Fig. 3-4). US also affected
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43 303 the Malvern device (+20% silt). The trends and the differences between the devices were similar
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45 304 whatever the proportion of sand in the samples.
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51 305 **4. DISCUSSION**

52 306 The measures performed by the 4 devices on the different samples (CRMs, sediments and
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54 307 associated mixtures) allowed determining their respective capacities in terms of measurement quality.
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1 308 The results demonstrated that several differences were observed despite the good and reproducible
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4 309 characterization of the main/modal classes and their distribution by the devices. These differences can
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6 310 be separated into three groups: intra-variation, variation with the certified materials, and variation
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8 311 between the devices.
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12 312 **4.1. Intra-variation**

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15 313 The intra-variation is the difference observed for the aliquots of the same subsample measured
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17 314 by the same device. For the CRMs, this difference was observed only in mixture C measured by the
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19 315 Coulter (Fig. 1) and is linked to the operator subsampling operation. Subsampling appears to be highly
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22 316 operator-dependent and can infer variations on the results of the same sample that are even higher
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24 317 than the analytical differences observed [5]. The intra-variation of the natural samples was observed
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26 318 mostly for coarse classes (very coarse silt to fine sand) measured by the Coulter and the Portable_C
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28 319 devices (Fig. 2). This was observed both without and with US and was mainly related to the
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30 320 heterogeneity of the sample particles. Indeed, during the subsampling some samples contained coarse
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34 321 organic particles such as tree leaves that modified the proportions between modes. For the
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36 322 Portable_C, it was also related to the presence of bubbles as explained in the chapter 2.4. To improve
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38 323 the characterization of sand particles, the pump speed of this device was increased for coarse samples
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41 324 and triggered the formation of bubbles. It is therefore relevant to perform measures on multiple sub-
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43 325 samplings of a sample (at least three) to characterize a more precise mean PSD and to be careful with
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45 326 the presence of coarse organic materials and bubbles. Also, the sieving of coarse particles could be a
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47 327 solution for decreasing the pump speed.
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51 328 **4.2. Variation with the CRMs**

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54 329 The measures performed on the CRMs for all the devices presented differences with the
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57 330 certified values. First, they underestimated the proportion of clay in the finest reference (BCR-066) and
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1 331 the different associated mixtures (Fig. 1). This underestimation was linked to the particle morphology
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4 332 and the clay mineralogy [9] affecting the PSD measured by LDM. Similar observations had already
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6 333 performed by [10,21]. Second, the PSDs measured by all the devices were more spread out than the
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8 334 associated references or mixtures. Also, particles were observed in classes where they should not have
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11 335 been present. For example, all the devices measured clay particles with the BCR-067 and SRM1003c
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13 336 (Fig. 1). However, the proportions in these classes were lower than 10%. This difference might be
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15 337 related to the presence of particles not washed during cleaning. Despite the measure of the
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18 338 background PSD with tap or dionized water, some particles might have been trapped in the system
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20 339 and resuspended during other measurements. This difference might also be linked to the different
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23 340 methods used to characterize the particle size for their certification. BCR-066 and BCR-067 were
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25 341 characterized by the Pipette method [39] which might have triggered differences with the LDM, as
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27 342 observed with the proportion of clay.

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32 344 To summarize, all 4 devices exhibit the same trends in accuracy from small to coarser particles.
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34 345 However, deviations on the d50 remain roughly lower than 10% for the standard ranging from 4 μ m to
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36 346 35 μ m. As explained previously, the devices do not accurately characterize clay, whatever the
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39 347 proportion observed, with the exception of the total absence of clay. Higher relative differences are
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41 348 observed when the proportion of clay is low (approx. 10%). For the silt, a small difference is observed
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44 349 in the CRMs characterized by almost only silt particles. Below 30% silt, the difference can be significant
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46 350 (from 20% up to 300%), especially for CRMs mainly characterized by clay and silt (BCR-066). Finally,
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49 351 the absence of particles in any simple class is accurately characterized by the devices.
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51 52 352 **4.3. Intercomparison of the devices**

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55 353 The variation between the devices was observed on almost all the samples including the CRMs
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57 354 and their mixtures and was directly related to the specificities of the devices. As explained previously,
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1 355 each device is manufactured differently (water volume, laser cells, etc.). One of the main source of
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4 356 differences is linked to the use of US and its power. Strong (Cilas, Portable_C) and weak (Coulter)
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6 357 variations were therefore observed after using US for certain samples. The absence of variation for the
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8 358 Coulter demonstrated that the US power might have been insufficient to separate the aggregates.
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10 359 However, it is difficult to investigate US power on CRMs as they are mostly composed of non-cohesive
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12 360 materials and, unfortunately, this parameter was not expressed in Joules by mL [50], so the power
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14 361 used by the different devices was different (Table 1). This observation demonstrates the need to
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16 362 describe the power and time of US used in future studies to ensure reproducibility, as expressed by
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18 363 Bieganowski et al. (2018). Sonication is used to estimate the aggregation index of soil and sediment
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20 364 samples [51] and the results obtained may not be comparable in the absence of such description.
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25 365 Another strong difference was observed when the main mode was located close to an interval
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27 366 between two classes, a phenomenon also observed for the CRM SRM1003c. The proportion of these
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29 367 two classes could be significantly different between the devices which distribute the mode between
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31 368 them (Fig. 1). Moreover, this distribution can be problematic when the interval is located between clay
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33 369 and silt or silt and sand such as the Azergues sample (Fig. 2 – Table 3). Vigilance is crucial when
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35 370 comparing such results.
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39 371 Results on raw PSD and corrected PSD of the Portable were compared to investigate the impact
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41 372 of the rising tails observed for the finest classes (Electronic Supplementary Material). The results
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43 373 demonstrated that rising tails had a negligible impact on the proportion of the classes. Highest
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45 374 difference was observed on the clay content of the Barcarin sample with 18% on the raw PSD and 11%
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47 375 on the corrected PSD. Moreover, the potential characterization of a lower proportion of clay by
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49 376 deleting the finest classes remained non-significant and the values were mainly in the range of the
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51 377 values of the other devices. For this specific study, it was not relevant to remove the finest classes.
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53 378 However, correction remained necessary for the coarse modes induced by the presence of bubbles
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55 379 caused by the pump speed as explained above.
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4 381 To evaluate the differences and allow users to determine those they can obtain with their
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6 382 devices, figures 5, 6 and 7 summarize the observations by grouping all the measures performed on all
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8 383 the natural samples and mixtures for all the parameters studied. Relative differences were then
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11 384 estimated for each device.

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13 385 For d50 (Fig. 5), the larger differences (up to 80% of relative difference with the mean d50)
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15 386 were observed when the sample was a mix of silt and sand particles with a d50 ranging from 25 μm to
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17 387 50 μm or from 50 μm to 80 μm , with and without US, respectively. For samples with a high d50 (higher
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19 388 than 150 μm), the difference remained lower than 20% while it could exceed 20% for samples with a
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21 389 low d50.
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25 390 For the simple classes (Fig. 6), the relative difference was significant (>20%) when the
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27 391 proportion was lower than 70% and 80% with and without US, respectively, and this difference
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29 392 increased when the proportion decreased and was lower than 25%.
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32 393 Similar observations were performed for the detailed class (Fig. 7), the relative difference
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34 394 decreased as the proportion increased, but remained mostly higher than 20% whatever the proportion
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36 395 and the US. Therefore, since multimodality was characterized in most cases, detailed classes should be
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38 396 used to describe the trend of the PSD rather than comparing results from different devices on a
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40 397 qualitative basis.
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46 399 Finally, the Specific Surface Area (SSA) was estimated [52] based on the PSDs of the different
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48 400 references and samples. However, as the proportion of clay was different between the devices, and
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50 401 even non-significant, the SSAs estimated by them were all substantially different from each other. It
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52 402 appeared that this parameter is difficult to reproduce on different devices and cannot be compared
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5. CONCLUSION

The tests performed during this study demonstrated that the comparison of particle size measured using different devices can be complicated since differences were observed on certified materials and natural samples. Despite using similar protocols and samples, the results obtained for the selected parameters (d50, simple and detailed classes) were characterized by differences that could be significant. Those differences were directly related to the specificities of the devices, as each device is manufactured differently (water volume, number of laser cells, etc.). In the absence of information on measurement parameters such as US power in J.ml^{-1} and pump speed, the comparison of PSDs from different studies can be erroneous, even for a similar device. It is therefore better to only compare the trend (such as the modal classes or the presence of these types of particle) rather than the measured proportion. Quantitative comparisons should only be performed if the parameters of the different analyses are known.

Differences in d50 were observed even with CRMs. Deviation from the expected d50 observed on all the lasers presented similar trends, with increases in particle size. Although there were differences in the amplitude of uncertainty, they all estimated the d50 for particle-sizes above $10\mu\text{m}$ within 10% uncertainty. However, for samples mostly characterized by clay and fine silt particles, the d50 was overestimated. This is important, especially for natural samples where these fractions are often present. Indeed, this information is required in numerous studies. Moreover, on natural sediments mixed with clay silts and sands, the problem of sand is not trivial. Sand is often not seen in these cases when clay size particles are present in quantity. Sieving sediments to remove the sand might be a solution to eliminate the impact of coarse particles.

For the different classes, the main problem observed concerned the position of the modes. Displaying the distribution of the classes greatly depends on the classification chosen relative to the sediment grain size distribution. It is therefore crucial to define the localization of the modes before

1 428 conducting a comparison with results from different devices. Significant differences can be observed
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4 429 in samples characterized by a PSD with modes between two classes (clay/silt, silt/sand or detailed
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6 430 classes).

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28 438 **Compliance with ethical standards**

30 439 **Conflict of interest**

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36 440 On behalf of all authors, the corresponding author states that there is no conflict of interest.
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Table 1: Main parameters of the grain size analyzers used within the OSR. n.a. not available.

	Main characteristics of the devices			
	Cilas	Beckman Coulter	Malvern	Sequoia
Brand	Cilas	Beckman Coulter	Malvern	Sequoia
Model	1190L	LS 13 320	Mastersizer 2000	LISST-Portable XR
Range of measured size (μm)	0.04 - 2500	0.04 - 2000	0.02 - 2000	0.34 - 500
Number of channels	100	132	100	33
Max. volume of the vessel (mL)	450	1000	1000	117
	Main parameters set for analysis			
Ultrasonic use	30s before	during analysis	during analysis	30s before analysis
	and during analysis			
Type of liquid and dispersant	deionized water	tap water	tap water	deionized water
	without dispersant without dispersant without dispersant without dispersant			
Duration of a single measurement	60 s	60 s	10 s	60 s
Optical model	Mie	Mie+Fraunhöfer*	Mie	Mie
Refractive index	1,55	1,5	1,57	1,54
Absorption coefficient	0,1	0,0	0,1	0,1
Ultrasound sonication power	50%	6 on scale	100%	30%
		of 8 levels		
Stirrer speed	150 RPM	n.a.	1000 RPM	n.a.
Pump speed	120 RPM	80-90%	2500 RPM	20-25%
Obscuration (Obs)	Obs: 5 – 25%	Obs: 8-16%	Obs: 5 – 25%	OT: 0.75 - 0.95
or Optical Transmission (OT)				

* Fraunhöfer used when particles are coarse (LS-13320 Manual)

Table 2 – Mean D50 (μm) and standard deviation determined by the devices without US. n.a. not available.

	BCR66	BCR67	SRM1003c	Mix_A	Mix_B	Mix_C
Certified range (μm)	0.35 – 3.5	2.4 - 32	25 - 50	n.a.	n.a.	n.a.
Certified D50 (μm)	1.13	10.41	32.1 \pm 1.0	n.a.	n.a.	n.a.
Cilas	1.2 \pm 0.1	12.4 \pm 0.1	32.6 \pm 0.8	11.5 \pm 0.1	4.2 \pm 0.4	11.4 \pm 0.5
Coulter	1.5 \pm 0.1	11.3 \pm 0.1	31.6 \pm 0.1	8.9 \pm 0.5	1.8 \pm 0.1	4.6 \pm 2.3
Malvern	1.9 \pm 0.1	12.3 \pm 0.1	31.9 \pm 0.1	10.1 \pm 0.1	3.0 \pm 0.1	8.9 \pm 0.1
Portable_C	1.9 \pm 0.1	11.8 \pm 0.4	32.6 \pm 0.6	9.3 \pm 0.1	2.8 \pm 0.1	5.9 \pm 0.7

Table 3 – Mean proportion (%) and standard deviation of the simple classes measured by the four devices on the natural samples with and without US. n.d. not detected; n.a. not analyzed.

Devices	US	Barcarin			Jons			Azergues			Arc		
		Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand
Cilas	without	8.8 ± 1.3	90.3 ± 0.8	0.5 ± 0.5	3.2 ± 0.4	69.2 ± 0.4	27.4 ± 0.9	2.0 ± 0.1	51.3 ± 0.7	47 ± 0.9	0.7 ± 0.1	1.0 ± 0.1	98.3 ± 0.5
Cilas	with	9.8 ± 2.1	90.0 ± 1.9	0.2 ± 0.4	4.4 ± 0.5	78.3 ± 1.6	17.1 ± 1.5	3.0 ± 0.1	87.6 ± 0.7	9.4 ± 1	1.0 ± 0.1	6.4 ± 2.0	92.8 ± 1.9
Coulter	without	21.6 ± 0.9	78.0 ± 0.7	0.4 ± 0.5	8.0 ± 0.1	64.8 ± 1.0	27.3 ± 1.0	2.0 ± 0.1	49.0 ± 1.5	49.4 ± 1.4	0.7 ± 0.5	13.0 ± 8.1	86.3 ± 8.6
Coulter	with	n.a.	n.a.	n.a.	10.7 ± 1.2	67.3 ± 3.8	21.7 ± 5.0	2.0 ± 0.1	55.3 ± 1.1	42.7 ± 1.1	0.7 ± 0.5	14.2 ± 8.4	84.9 ± 8.8
Malvern	without	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	1.7 ± 0.5	44.2 ± 2.1	54 ± 2.2	n.d.	n.d.	100.0 ± 0.1
Malvern	with	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	2.8 ± 0.4	62.3 ± 3.9	34.9 ± 4.3	n.d.	0.4 ± 0.9	99.6 ± 0.9
Portable_C	without	11.0 ± 1.4	87.5 ± 0.7	1.5 ± 2.1	3.0 ± 0.1	74.0 ± 2.8	23.0 ± 2.8	2.1 ± 0.6	72.6 ± 9.9	25.1 ± 10.7	0.3 ± 0.1	9.6 ± 1.0	90.0 ± 1.0
Portable_C	with	n.a.	n.a.	n.a.	7.0 ± 0.1	89.3 ± 2.1	3.3 ± 3.2	4.2 ± 0.4	89.4 ± 1.7	6.4 ± 2.1	0.7 ± 0.8	15.4 ± 11.8	83.9 ± 12.3

Table 4 – Mean D50 (μm) and standard deviation measured by the four devices on the natural samples with and without US. n.a. not analyzed.

Devices	US	Barcarin	Jons	Azergues	Arc
Cilas	without	9.9 \pm 0.5	34.6 \pm 1.0	57.0 \pm 1.3	194.1 \pm 3.6
Cilas	with	9.2 \pm 0.7	25.0 \pm 1.3	23.1 \pm 1.1	179.3 \pm 5.3
Coulter	without	6.2 \pm 0.4	28.2 \pm 0.7	61.1 \pm 2.8	146.6 \pm 20.0
Coulter	with	n.a.	21.9 \pm 3.7	49.7 \pm 1.3	145.9 \pm 20.5
Malvern	without	n.a.	n.a.	71.6 \pm 5.8	171.9 \pm 1.6
Malvern	with	n.a.	n.a.	40.0 \pm 5.4	171.5 \pm 2.1
Portable_C	without	7.9 \pm 1.0	35.4 \pm 0.2	37.9 \pm 9.1	143.9 \pm 20.1
Portable_C	with	n.a.	15.5 \pm 3.4	20.9 \pm 1.8	132.9 \pm 25.3

Fig.1 Proportion of the detailed classes measured with US by the different devices (Cilas, Coulter, Malvern and Portable corrected) on the CRMs (BCR-066, BCR-067 and SRM1003c) and the different mixtures (A, B and C). Certified proportions are also presented (Reference). Values over the bar represents the percentage of the proportion

Fig.2 Detailed classes measured by the different devices on the natural samples with and without US. Values over the bar represent the percentage of the proportion. No measures were conducted on the Malvern for Barcarin and Jons samples

Fig.3 d50 measured by the different devices on the mixtures of Arc and Azergues samples

Fig.4 Detailed classes measured by the different devices on mixture 2 with and without US. Values over the bar represent the percentage of the proportion

Fig.5 Relative difference between the mean d50 measured by all the devices and the mean d50 measured by each device with and without US. Grey areas represent 10% and 20% difference

Fig.6 Relative differences between the mean proportion of each simple class measured by all the devices and the mean proportion measured by each device with and without US. Grey areas represent 10% and 20% difference. Abscise represent the mean proportion of the associated simple class for all devices

Fig.7 Relative differences between the mean proportion of each detailed class measured by all the devices and the mean proportion measured by each device with and without US. Grey areas represent

10% and 20% difference. The abscissa represents the mean proportion of the associated detailed class for all devices

Figure 1

[Click here to access/download;Figure;Fig1.pdf](#)
Devices and Reference: Cilas Coulter Malvern Portable_C Reference

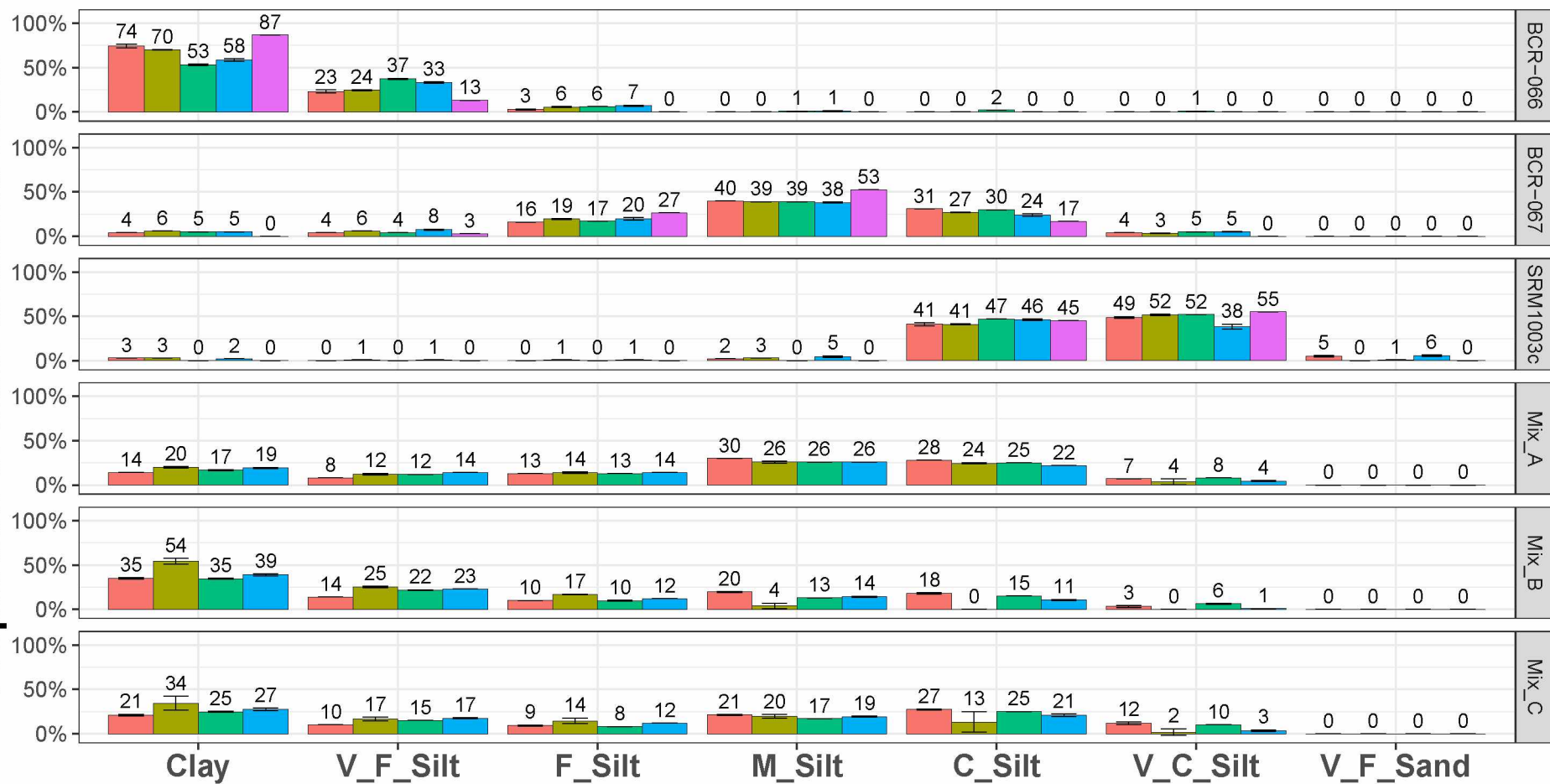
Proportion of each detailed class


Figure2

[Click here to access/download;Figure;Fig2.pdf](#)
Devices: Cilas Coulter Malvern Portable_C

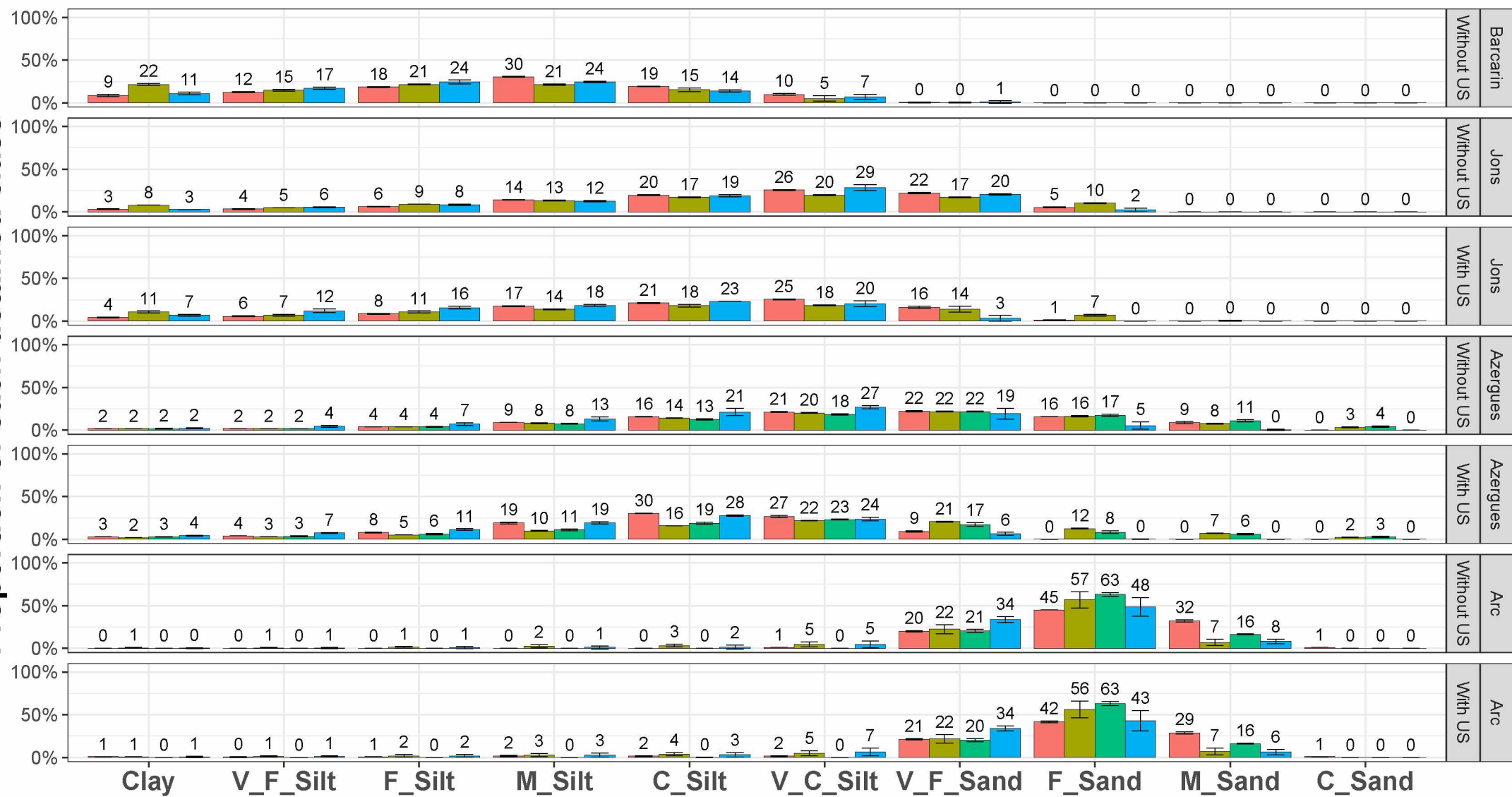
Proportion of each detailed class


Figure3

[Click here to access/download;Figure;Fig3.pdf](#)

US ○ without □ with

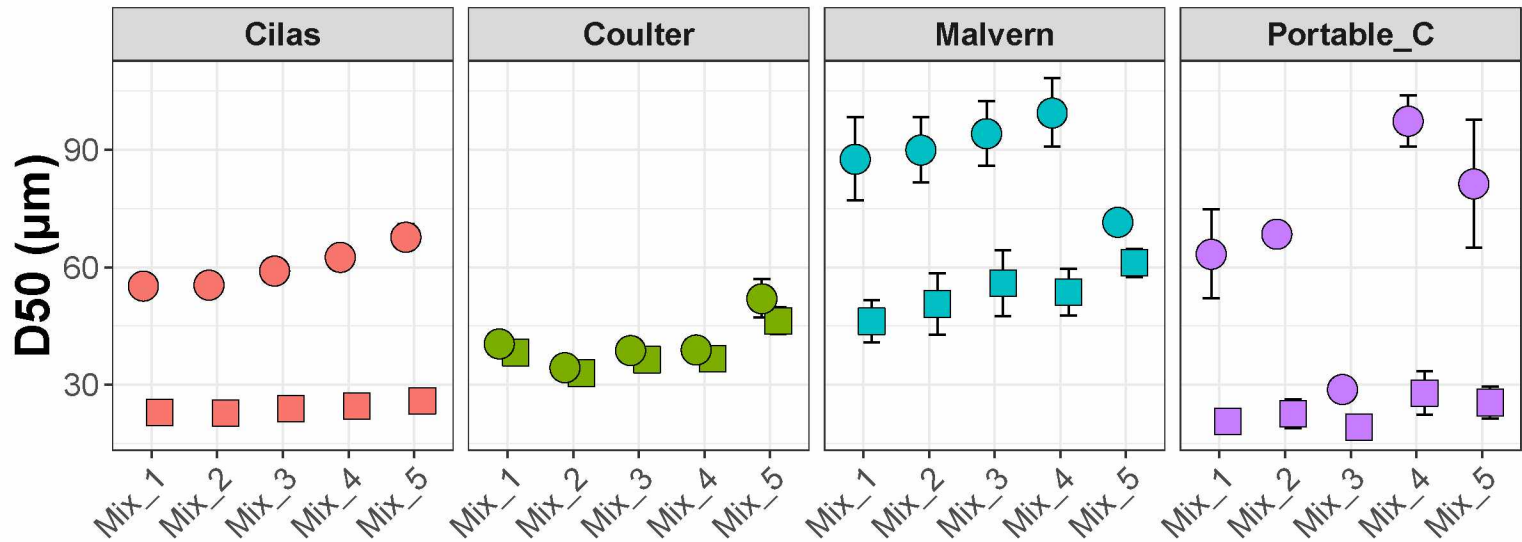


Figure4

Devices: ■ Cilas ■ Coulter ■ Malvern ■ Portable_C

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Proportion of each detailed class

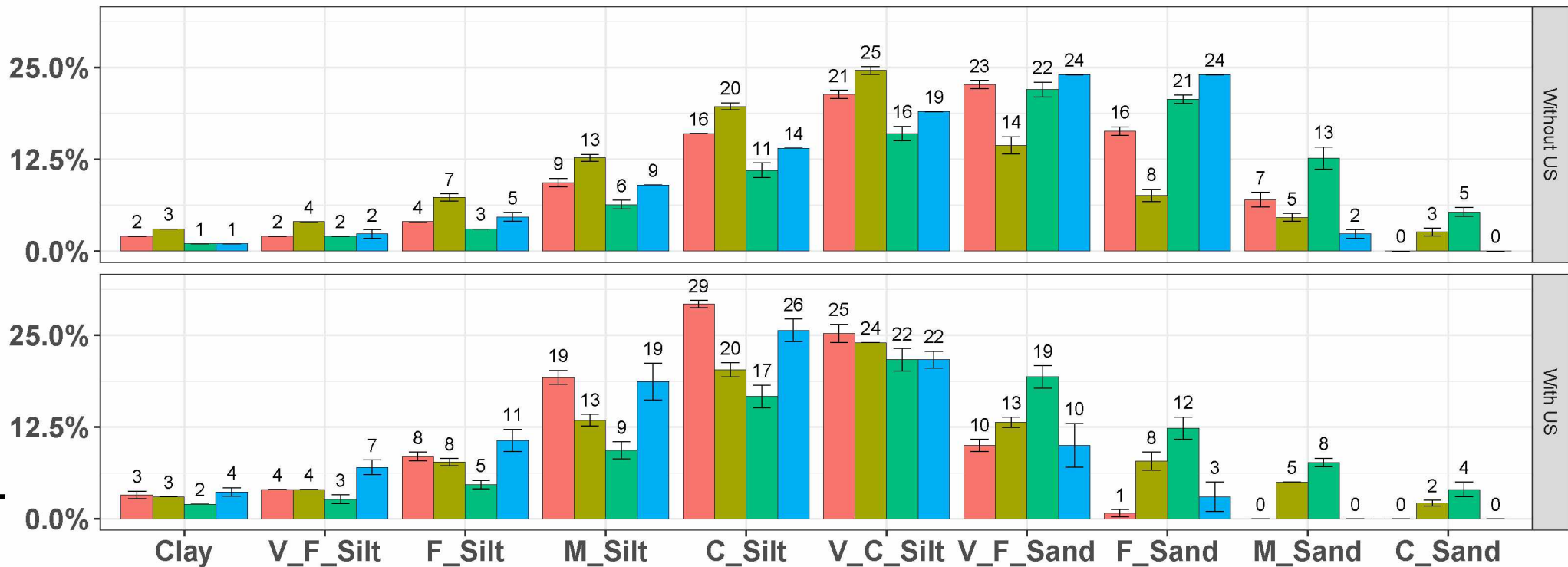


Figure6

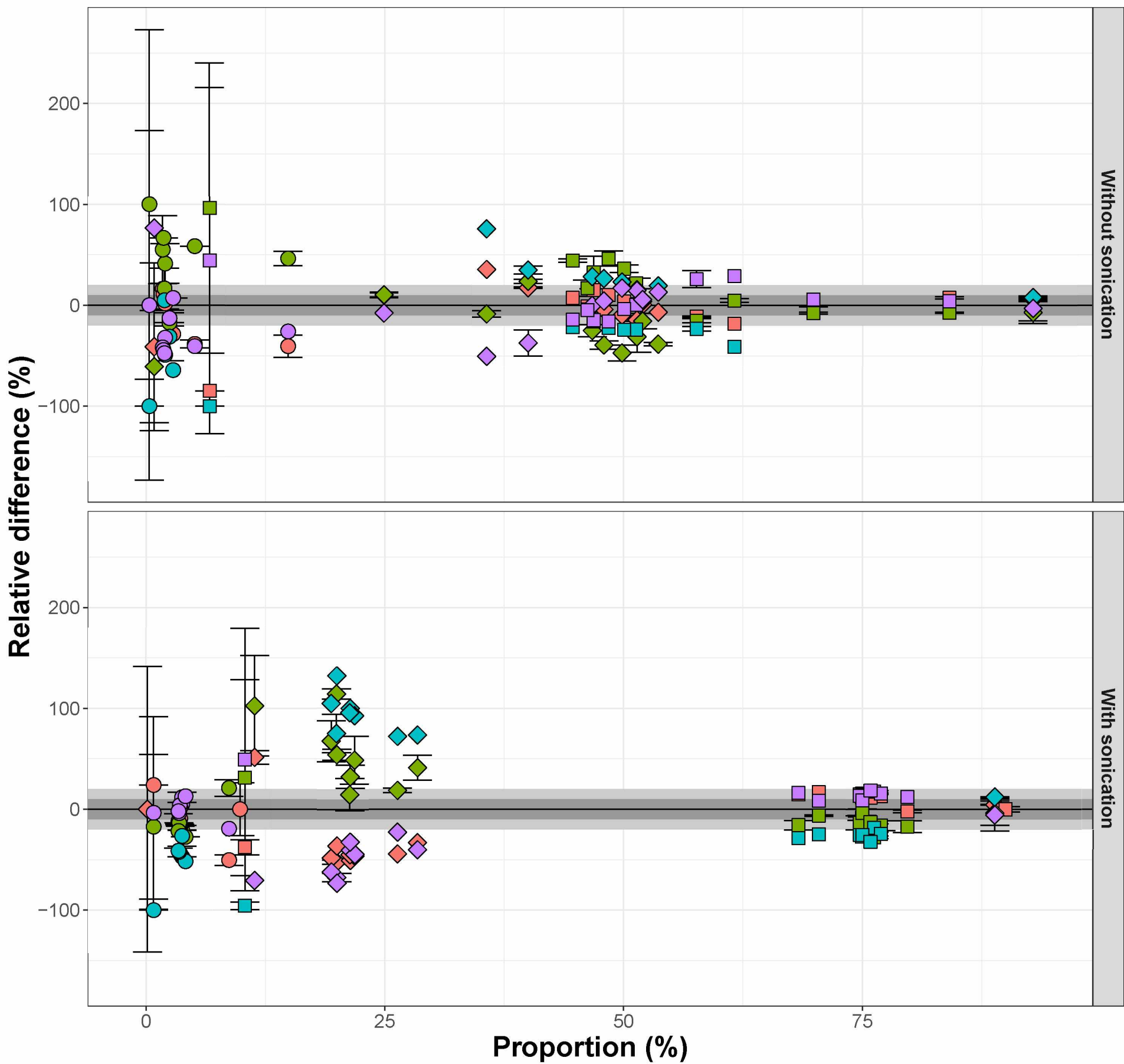
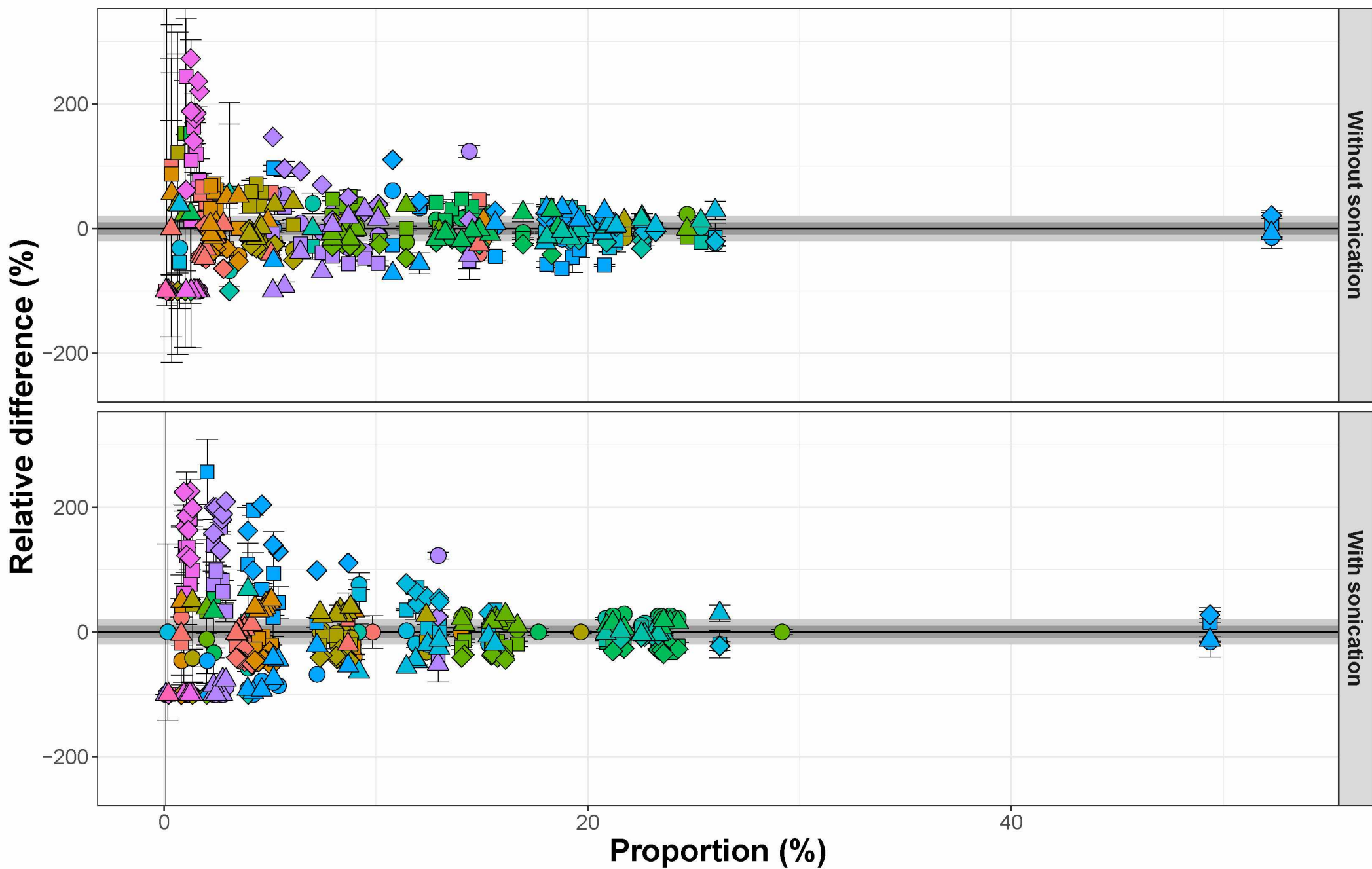
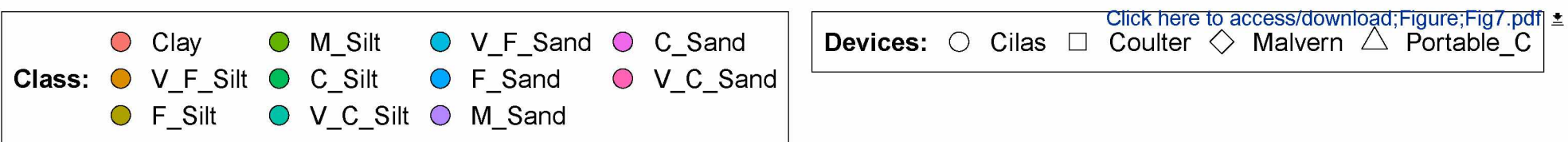
classes: ○ Clay □ Silt ◇ Sand**Devices:** ● Cilas ● Coulter ● Malvern ● Portable_C

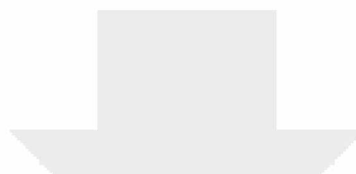
Figure7





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