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Supplemental document.

Table S1: Demographics of all volunteer groups who provided saliva samples for this study.

|	Healthy	Testing clinic COVID-19 negative	Testing clinic COVID-19 positive	Hospitalized COVID-19 positive
Total number of volunteers	152	250	36	19
Age range, n (%)				
0–20	2 (1)	26 (10)	6 (17)	0 (0)
21–40	94 (62)	113 (45)	15 (42)	5 (26)
41–60	48 (32)	77 (31)	12 (33)	7 (37)
61–80	8 (5)	33 (13)	3 (8)	6 (32)
81+	—	1 (0)	—	1 (5)
Sex at birth, n (%)				
Female	54 (36)	134 (54)	20 (56)	4 (21)
Male	48 (32)	115 (46)	16 (44)	15 (79)
Prefer not to say	50 (33)	—	—	—
Symptoms, n (%)				
Respiratory symptoms				
Non-respiratory symptoms				
None				
Not reported				
Disease, n (%)	48 (32)	112 (45)	6 (17)	10 (53)
Other disease	104 (68)	138 (55)	30 (83)	9 (47)
None				
Nicotine consumption, n (%)				
Smoking	9 (6)	92 (37)	4 (11)	2 (11)
Vaping	4(3)	32 (13)	—	1 (5)
Alcohol consumption, n (%)	102 (67)	174 (70)	20 (56)	3 (16)
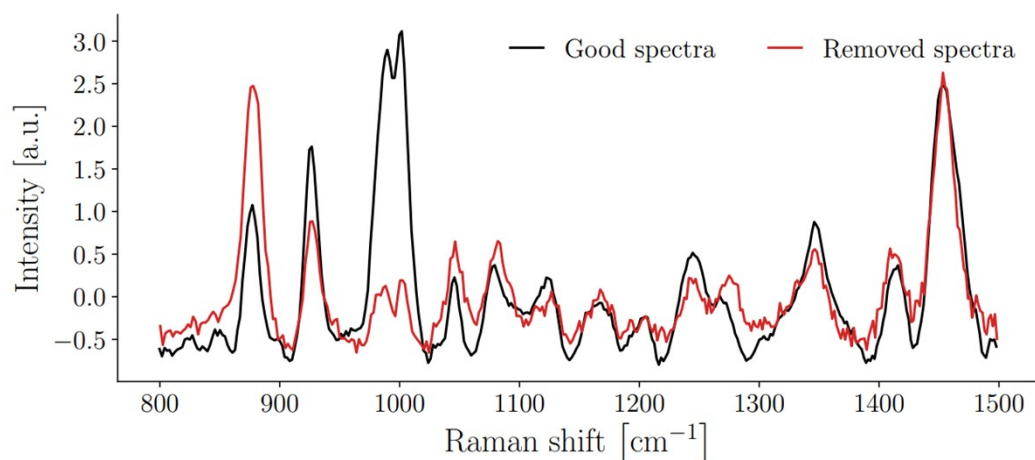
Supplementary Figure S1: Included and excluded Raman spectra from liquid saliva supernatant samples including COVID-19 negative volunteers (PCR negatives from a testing clinic and healthy volunteers), PCR positives from a COVID-19 testing clinic, and hospitalized COVID patients. The black line represents "good spectra" i.e. ones that met the inclusion criteria for spectral quality and the red line represents "removed spectra" i.e. spectra that were excluded from the analysis. Spectra were removed from all four groups of volunteers.
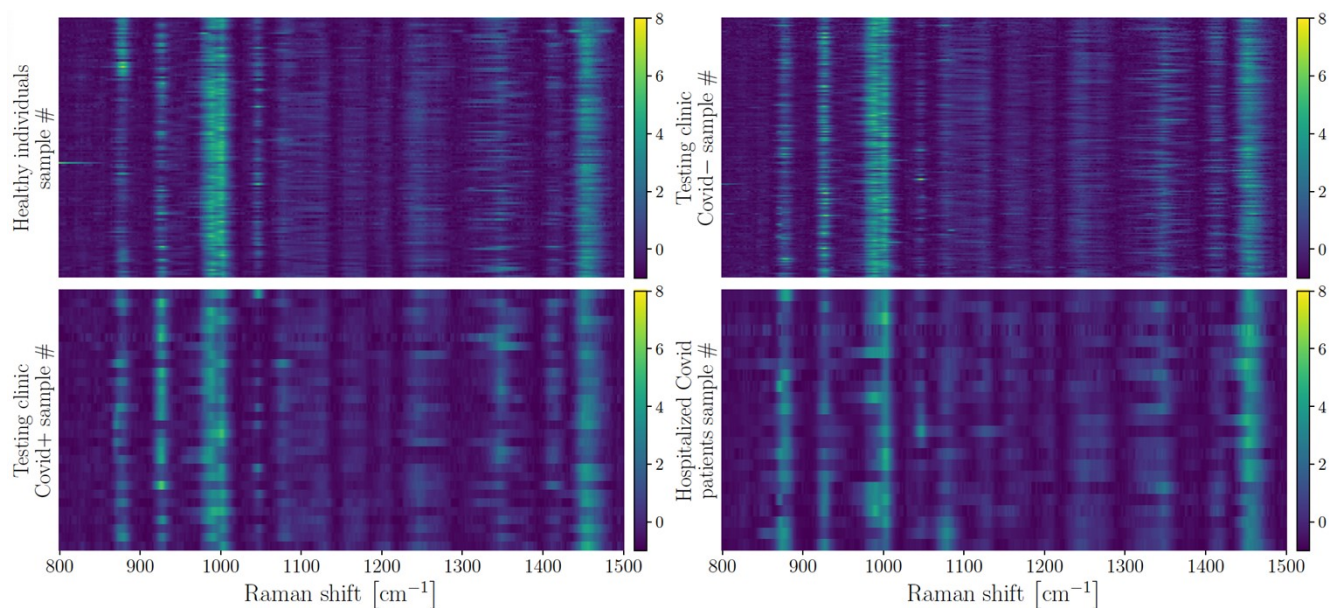
Supplementary Figure S2: Spectrograms of Raman spectra from saliva supernatant samples. The top left panel shows the spectrogram from 124 healthy volunteers who were presumed COVID-19 negative. The top right panel shows the spectrogram from 174 COVID-19 negative volunteers (PCR negatives from a testing clinic and healthy volunteers). The bottom left panel shows the spectrogram from 30 positive samples from the same. The bottom right panel shows spectrogram from 23 patients hospitalized with COVID-19.
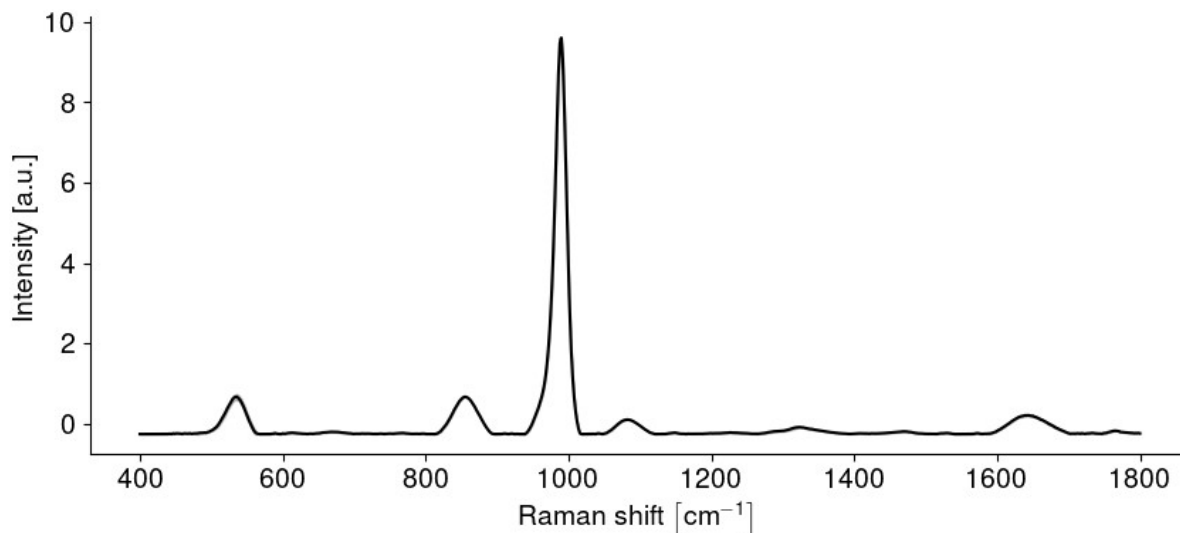
Supplementary Figure S3: Raman spectrum of potassium phosphate (dibasic) dissolved in water. The mean Raman spectrum of three acquisitions taken using the single-point Raman spectrometer. The variance is shown but not visible. Measurements were taken with 1.2W of laser power, 50 accumulations of 1100 ms.
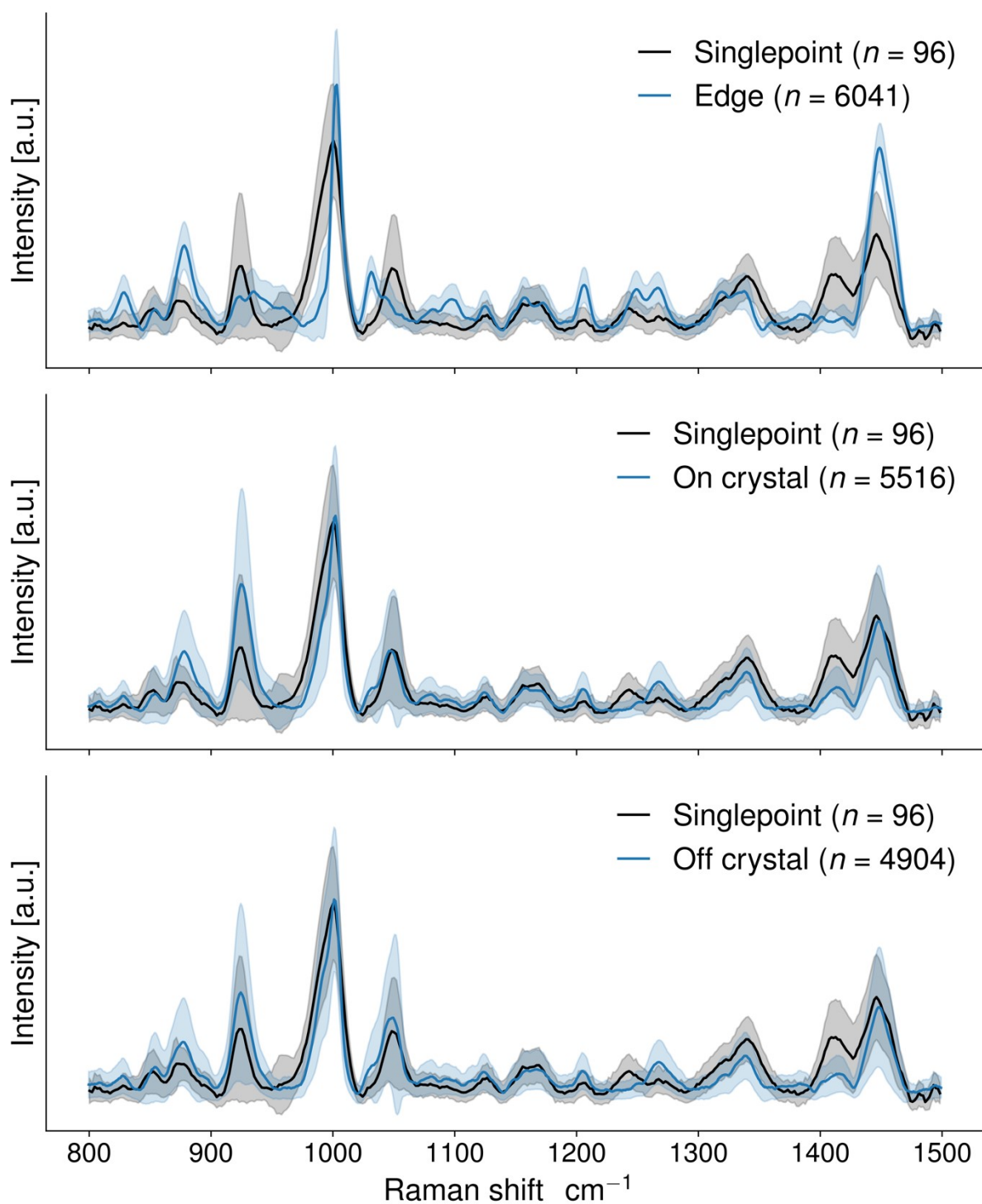
Supplementary Figure S4: Mean measurement computed from Raman spectra from dried saliva supernatant samples taken using the Renishaw microscope (blue line) and the single point system (n= 95 samples, black line). With the precise optics of the microscope, spectra were taken from the edge region (n = 72 samples, top), central crystalline region from directly on the crystals (n = 59 samples, centre), central crystalline region from off the crystals (n = 58 samples, bottom). With the single system, spectra were taken from the whole dried droplet. The standard deviation is shown as translucent shading.