GSA DATA REPOSITORY 2017291

Supplementary Information for "*Composition of Asian dust from cathodoluminescence spectral analysis of single quartz grains*" by Kana Nagashima, Hirotsugu Nishido, Masahiro Kayama, Yasunori Kurosaki, Syuhei Ohgo, and Hitoshi Hasegawa

Measurement of cathodoluminescence spectra of single quartz grains

The quartz grains were embedded in a small dimple (φ 2 mm) on a brass disk with non-luminescent epoxy resin, and were polished and mirror-finished using 1 μm diamond powder. The prepared samples were coated with ~2 nm carbon film to prevent charge build-up on the surface during electron irradiation.

A scanning electron microscopy-cathodoluminescence (SEM-CL) analysis was conducted using a SEM (JEOL: JSM-5410) combined with a grating monochromator (Oxford: Mono CL2) at the Okayama University of Science to measure CL spectra ranging from 300 to 800 nm in 1 nm steps. It takes eight minutes for each measurement. The CL spectra were excited using a continuous electron beam at normal incidence, with accelerating voltage of 15 keV, slit size of 4 mm square, probe current of 2.0 nA, and irradiated area of 2.5 μm^2 with room temperature, and collected using a retractable diamond machined paraboloidal mirror collector. The CL was dispersed by a grating monochromator, which has 1200 grooves/mm, a focal length of 0.3 m, F of 4.2, a limit of 0.5 nm resolution, and a slit width of 4 nm at the inlet and outlet. The dispersed CL was recorded by a photon-counting method using a photomultiplier tube (Hamamatsu: R2228) and converted to digital data.

All CL spectra were corrected for total instrumental response, which was determined using a calibrated standard lamp (Eppley Laboratory: Quartz Halogen Lamp, Kayama et al., 2010). This correlation prevents errors in the peak position of emission bands and allows a quantitative evaluation of CL intensity (Kayama et al., 2010). All results were presented in energy unit (eV) to identify the type and intensity of emission components shown as Gaussian curve in energy unit (e.g., Stevens-Kalceff, 2009). The total measurement procedure is easy and inexpensive.

CL-spectral change during grain-refining process

We investigated the change in CL spectral patterns of the quartz grains during the grain refining process to examine whether the CL of fine quartz grains preserves information on the host rock types. For this purpose, we selected the samples of quartz grains collected from Suna River in Okayama Prefecture, southwestern Japan, where the drainage area is distributed throughout a region of homogeneous granitic rocks of Mesozoic age (Fig. DR1, Geological Survey of Japan-AIST, 2015). As the quartz grains along the river originated from the same type of host rocks (granite), we could isolate the effects of the grain refining process by comparing the CL features of quartz grains with different sizes. The quartz grains were separated into six fractions; < 5, 5–10, 10–16, 16–75, 75–100, and 100–250 μm by sieving and pipetting. CL analysis was conducted for the quartz grains of the six fractions at the Okayama University of Science in the same way as employed for the samples from Taklimakan and Gobi deserts. The spectral deconvolution of measured CL discriminates five emission components corresponding to those shown in Table 1, and gives a relative abundance (fractional area) of each component.

Quartz grains in each size fraction of 5–10, 10–16, 16–75, 75–100, and 100–250 μm showed similar averages of the abundances of the emission components (Fig. DR2; highest abundance for EC5 following by EC1). However, the finest quartz grains in the <5-μm fraction showed lower contributions of EC5 and EC4 compared to the average values of quartz grains in the other size fractions, resulting in the highest abundance of EC1. This trend could be explained by the heat effect of electron irradiation on the quartz grain during the measurement (eight minutes irradiation), which vary the emission efficiencies among the components, especially pronounced in the grains with a smaller fraction size (< 5 μm) due to smaller heat capacity. Otherwise, it suggest that the numbers of some specific impurities or imperfections change when the grains reduces to a size smaller than 5 μm. In other words, quartz grains larger than 5 μm preserve the CL features of the host rocks, although the variations in the abundances of the five emission components increase with decreasing quartz grain sizes.

CL-spectral change due to the irradiation exposure during the measurement

We examined the changes in CL spectral of the quartz grains (5–16 μm) from the Taklimakan and the Gobi deserts in response to the irradiation exposure during the measurement (total 480 seconds). As is shown in Fig. DR3, CL intensities in red region (630 nm, 1.97 eV) show decreasing trend (6–8% decrease per 100 seconds) during the continuous irradiation of 480 seconds, except for the first 100 seconds of a quartz grain from the Taklimakan desert (#002) probably representing the relatively higher contribution of emission component 2 (energy center of 1.9 eV; decreases in response to the electron irradiation is “not” observed in contrast to emission component 3 (Stevens-Kalceff, 2009)). The CL intensities in purple-blue region (380 nm, 3.3 eV) also show a decreasing trend (approximately 22% decrease per 100 seconds) for a quartz grain from the Gobi desert, while the quartz grains from the Taklimakan desert show background values. Although the decreasing rate in purple-blue region is high, total effect of irradiation exposure on CL intensity in purple-blue regions (~16%) is similar or smaller compared to that in red region (< 26%) because of the shorter irradiation exposure time in blue region (80–190 seconds) compared to that in red region (300–430 seconds). Consequently, the effects of irradiation exposure on CL intensities not significantly change the CL-color, and the effect of electron irradiation was not concerned here.

References

Fang, X., Lu, L., Yang, S., Li, J., An, Z., Jiang, P., and Chen, X., 2002, Loess in Kunlun Mountains and its implications on desert development and Tibetan Plateau uplift in west China: Science in China (Series D), v. 45, p. 289–299.

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Stevens-Kalceff, M.A., 2009, Cathodoluminescence microcharacterization of point defects in α-quartz: Mineralogical Magazine, v. 73(4), p. 585–605.

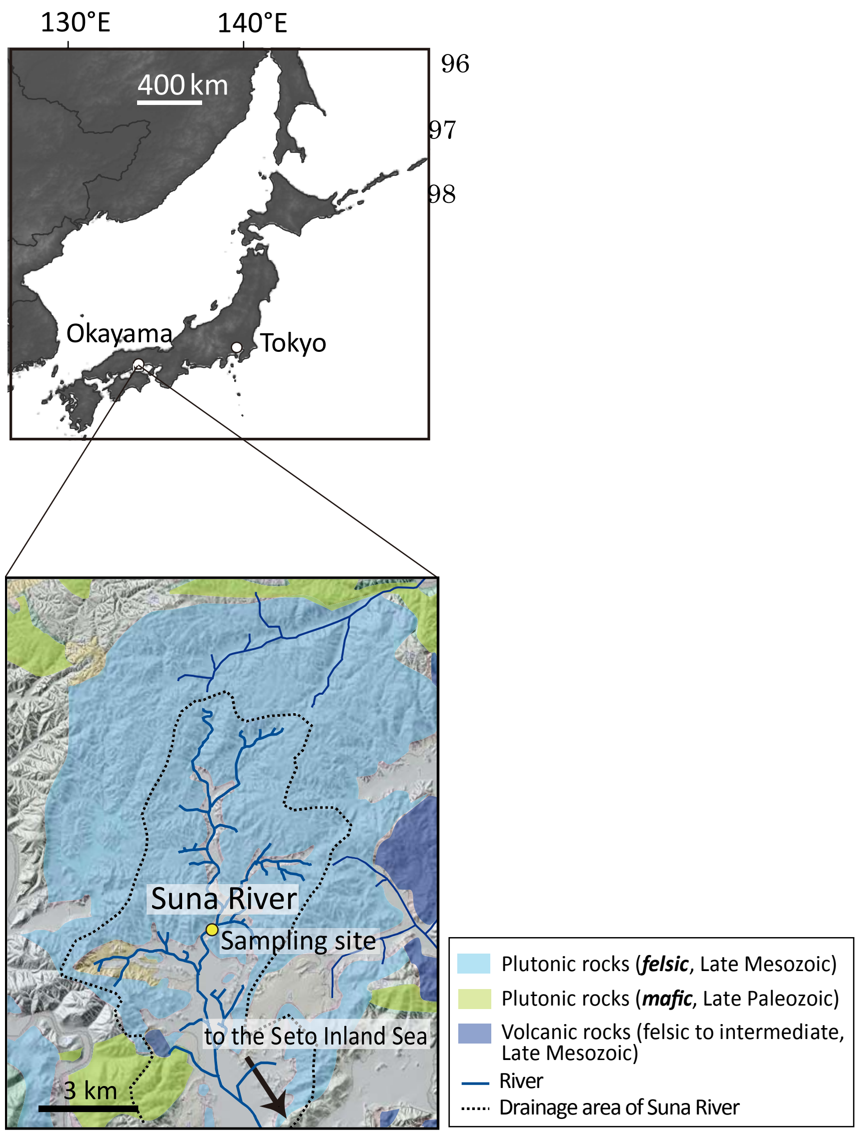


Figure DR1. Location of the sampling site and geological map surrounding the site. (modified after Geological Survey of Japan-AIST, 2015)

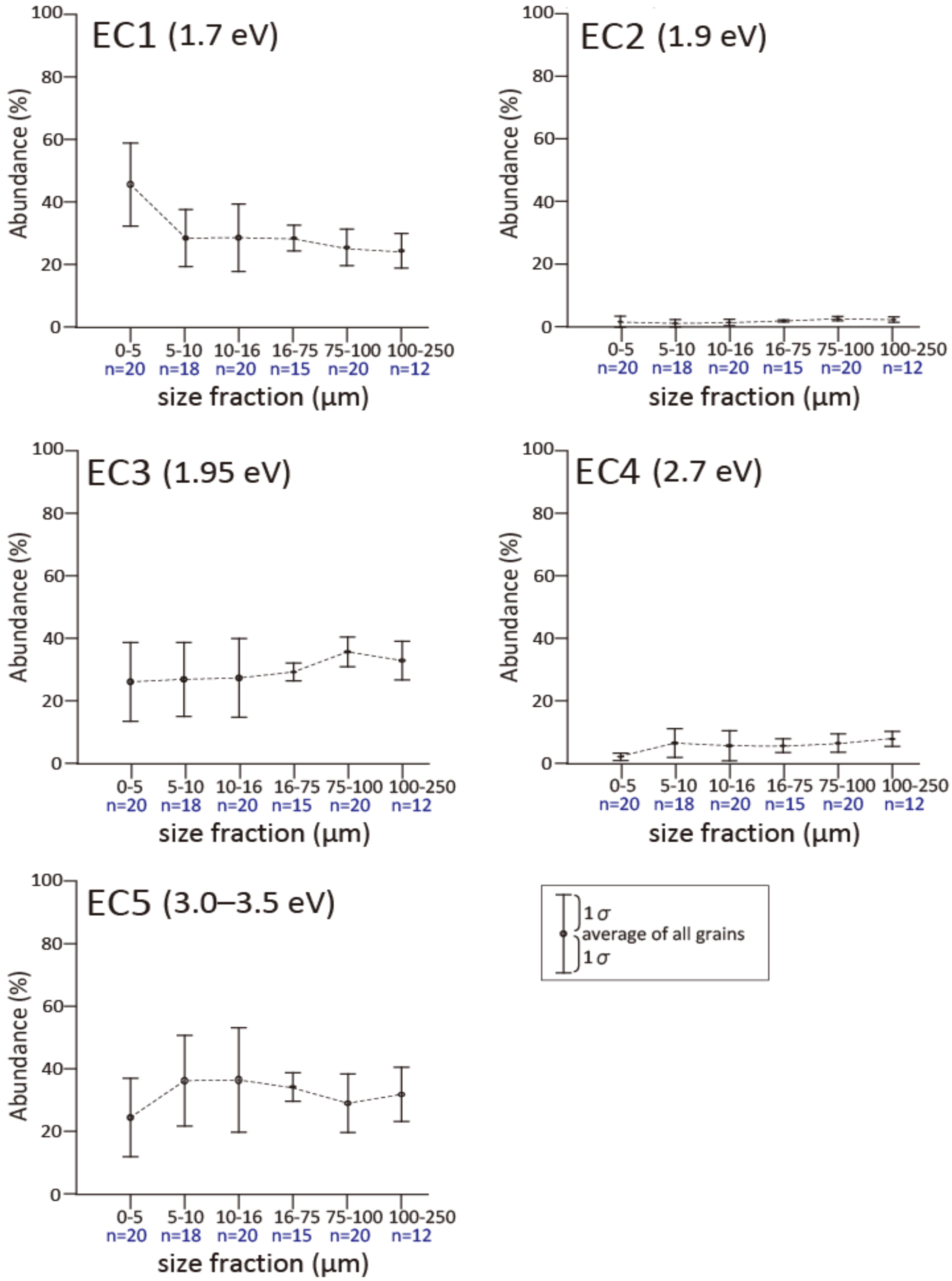


Figure DR2. Abundances of five emission components (EC) for quartz grains in various size fractions.

The bar represents average value and standard deviation for all quartz grains in each size fraction.

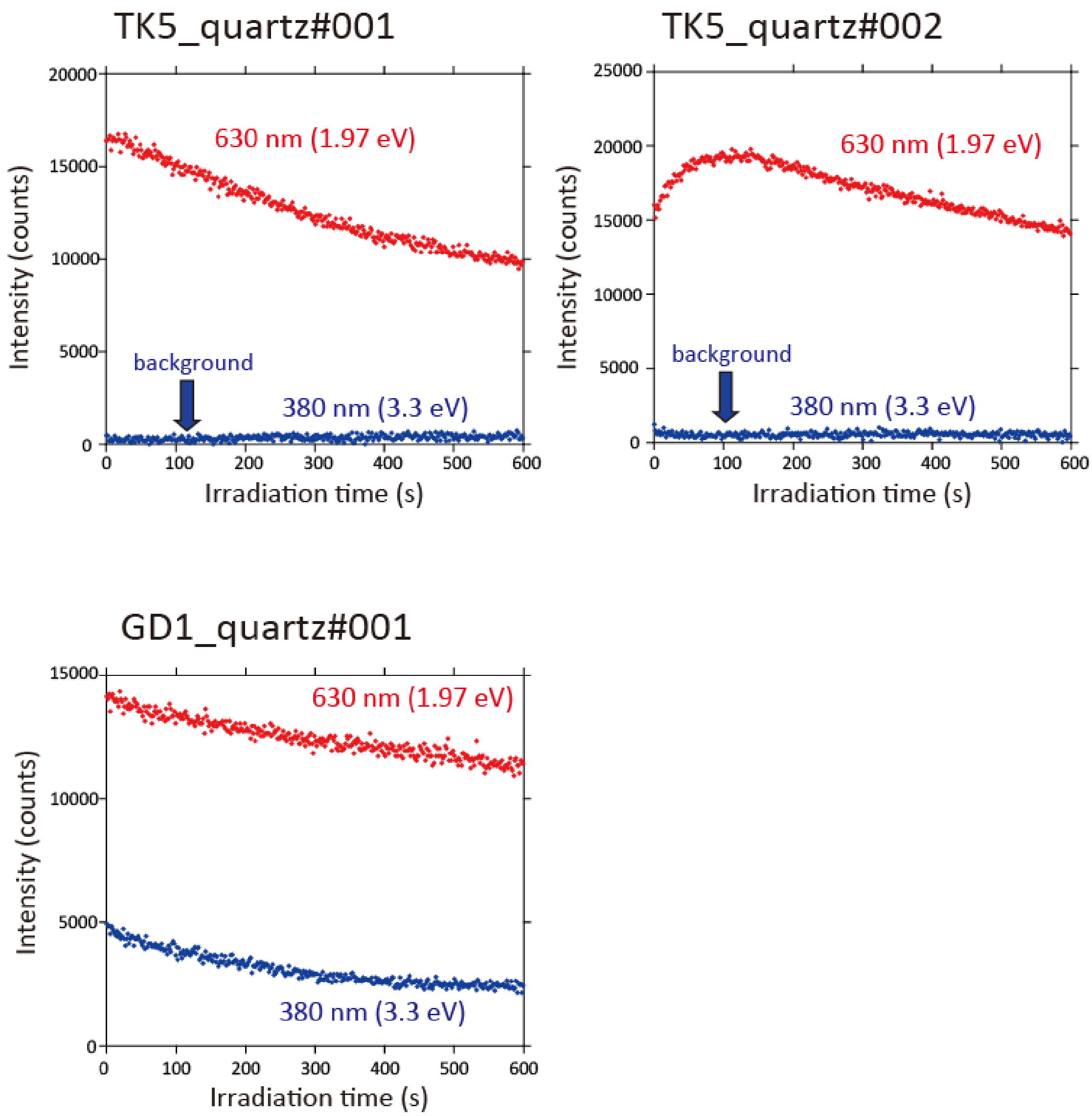
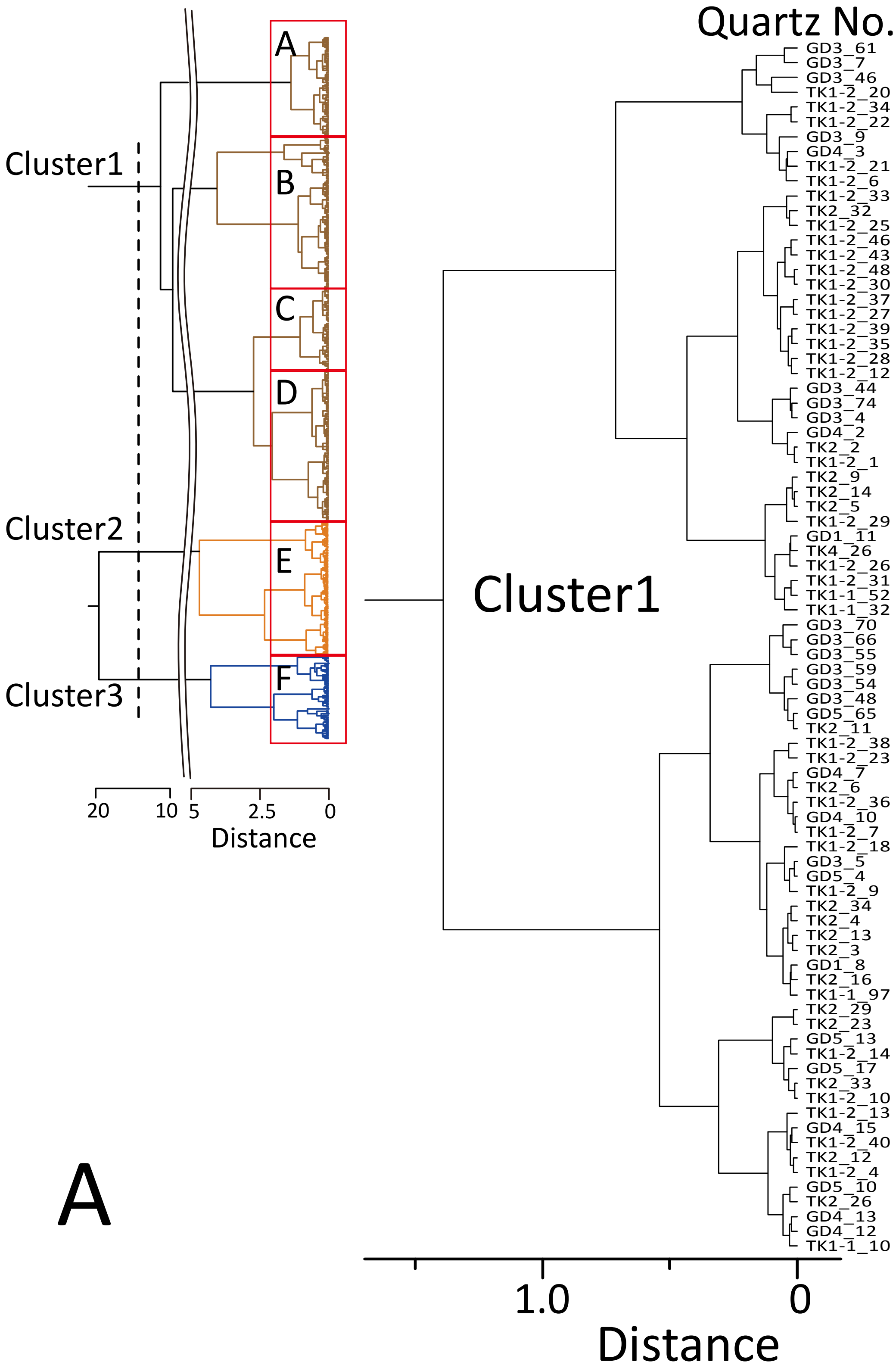


Figure DR3. Examples of CL intensity changes in red (630 nm) and blue (380 nm) regions for quartz grains from the Taklimakan (TK5) and Gobi (GD1) with the irradiation exposure time (15 keV, 2.0 nA, irradiated area = ~2.5 μm^2).


Cluster1
Cluster2
Cluster3
A
B
C
D
E
F
20
10
5
2.5
0
Distance
A
Quartz No.
Cluster1
GD3_61
GD3_7
GD3_46
TK1-2_20
TK1-2_34
TK1-2_22
GD3_9
GD4_3
TK1-2_21
TK1-2_6
TK1-2_33
TK2_32
TK1-2_25
TK1-2_46
TK1-2_43
TK1-2_48
TK1-2_30
TK1-2_37
TK1-2_27
TK1-2_39
TK1-2_35
TK1-2_28
TK1-2_12
GD3_44
GD3_74
GD3_4
GD4_2
TK2_2
TK1-2_1
TK2_9
TK2_14
TK2_5
TK1-2_29
GD1_11
TK4_26
TK1-2_26
TK1-2_31
TK1-1_52
TK1-1_32
GD3_70
GD3_66
GD3_55
GD3_59
GD3_54
GD3_48
GD5_65
TK2_11
TK1-2_38
TK1-2_23
GD4_7
TK2_6
TK1-2_36
GD4_10
TK1-2_7
TK1-2_18
GD3_5
GD5_4
TK1-2_9
TK2_34
TK2_4
TK2_13
TK2_3
GD1_8
TK2_16
TK1-1_97
TK2_29
TK2_23
GD5_13
TK1-2_14
GD5_17
TK2_33
TK1-2_10
TK1-2_13
GD4_15
TK1-2_40
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GD4_13
GD4_12
TK1-1_10
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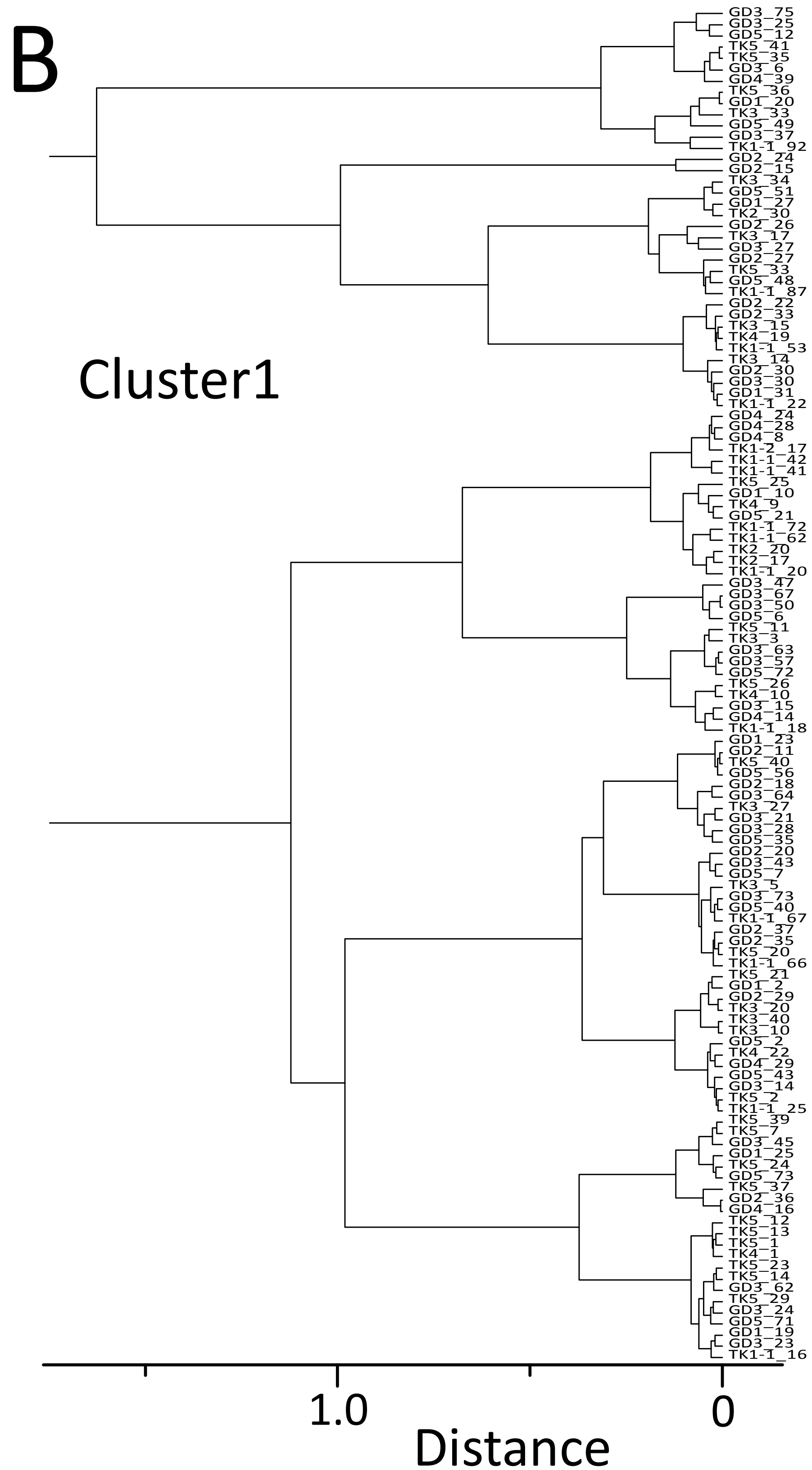
Distance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GD3_25
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TK5_41
TK5_35
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TK3_33
GD5_49
GD3_37
TK1-1_92
GD2_24
GD2_15
TK3_34
GD5_51
GD1_27
TK2_30
GD2_26
TK3_17
GD3_27
GD2_27
TK5_33
GD5_48
TK1-1_87
GD2_22
GD2_33
TK3_15
TK4_19
TK1-1_53
TK3_14
GD2_30
GD3_30
GD1_31
TK1-1_22
GD4_24
GD4_28
GD4_8
TK1-2_17
TK1-1_42
TK1-1_41
TK5_25
GD1_10
TK4_9
GD5_21
TK1-1_72
TK1-1_62
TK2_20
TK2_17
TK1-1_20
GD3_47
GD3_67
GD3_50
GD5_6
TK5_11
TK3_3
GD3_63
GD3_57
GD5_72
TK5_26
TK4_10
GD3_15
GD4_14
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GD1_23
GD2_11
TK5_40
GD5_56
GD2_18
GD3_64
TK3_27
GD3_21
GD3_28
GD5_35
GD2_20
GD3_43
GD5_7
TK3_5
GD3_73
GD5_40
TK1-1_67
GD2_37
GD2_35
TK5_20
TK1-1_66
TK5_21
GD1_2
GD2_29
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TK3_40
TK3_10
GD5_2
TK4_22
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GD5_43
GD3_14
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TK5_7
GD3_45
GD1_25
TK5_24
GD5_73
TK5_37
GD2_36
GD4_16
TK5_12
TK5_13
TK5_1
TK4_1
TK5_23
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GD1_19
GD3_23
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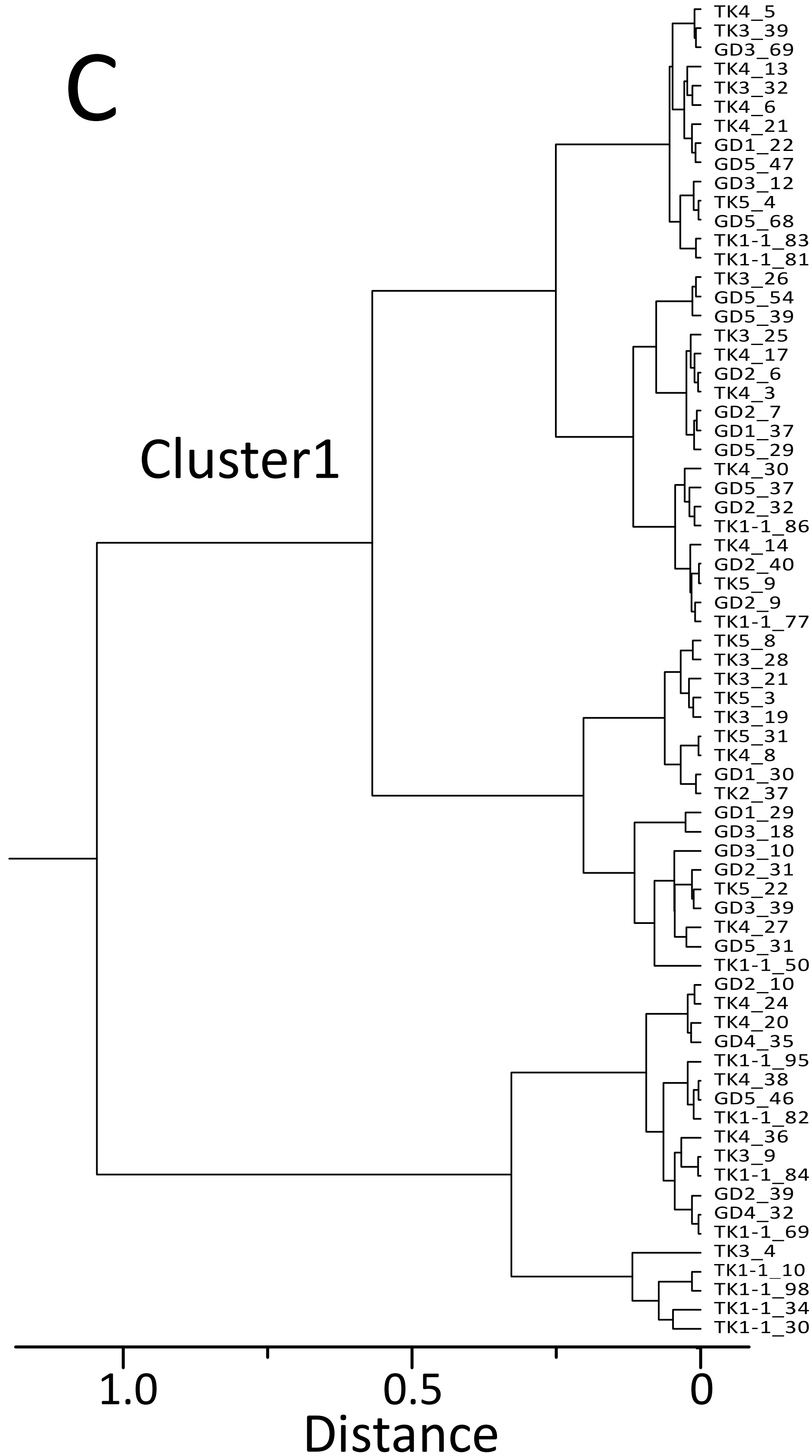
Distance


C
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GD3_69
TK4_13
TK3_32
TK4_6
TK4_21
GD1_22
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GD3_12
TK5_4
GD5_68
TK1-1_83
TK1-1_81
TK3_26
GD5_54
GD5_39
TK3_25
TK4_17
GD2_6
TK4_3
GD2_7
GD1_37
GD5_29
TK4_30
GD5_37
GD2_32
TK1-1_86
TK4_14
GD2_40
TK5_9
GD2_9
TK1-1_77
TK5_8
TK3_28
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GD4_32
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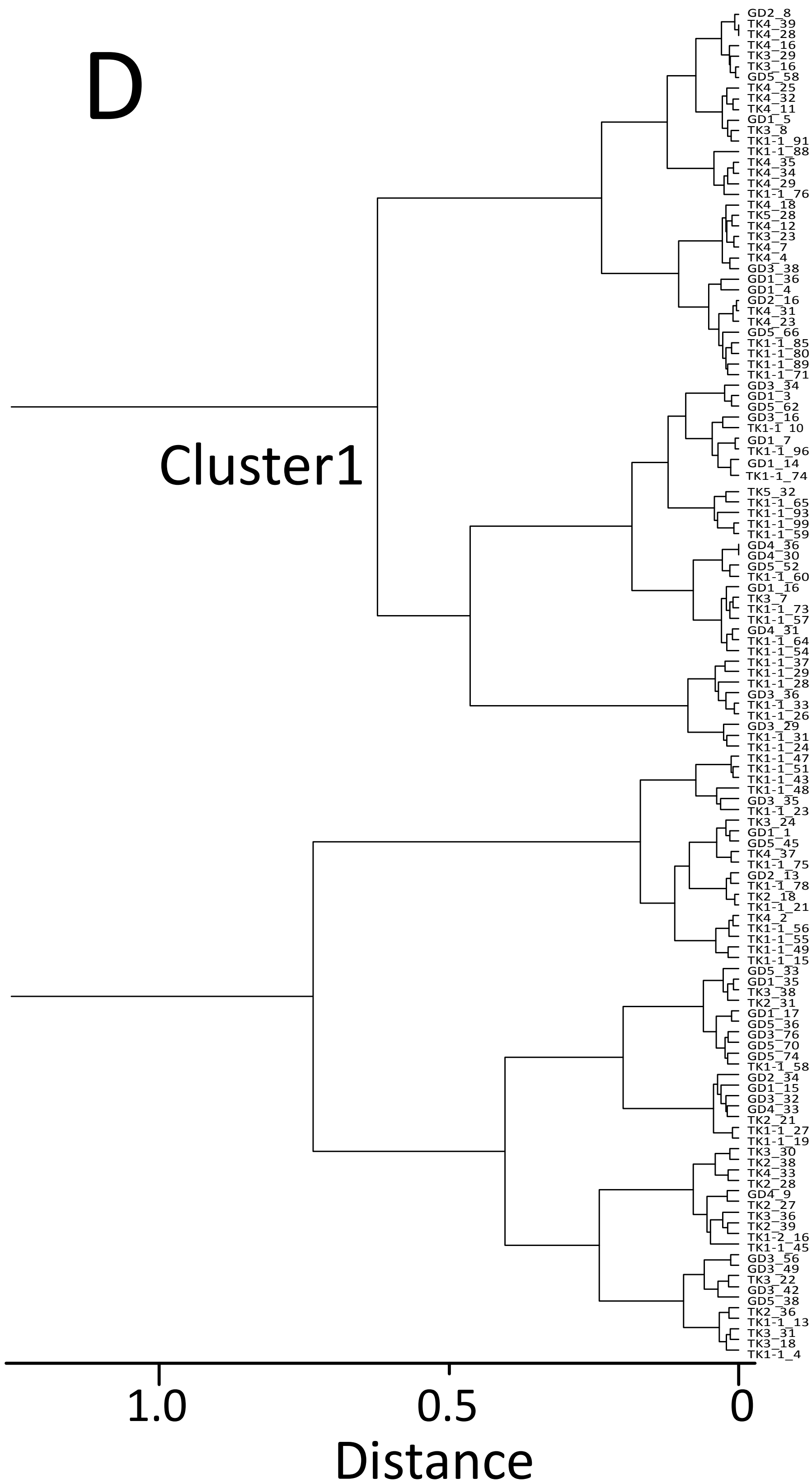
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GD5_58
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TK4_11
GD1_5
TK3_8
TK1-1_91
TK1-1_88
TK4_35
TK4_34
TK4_29
TK1-1_76
TK4_18
TK5_28
TK4_12
TK3_23
TK4_7
TK4_4
GD3_38
GD1_36
GD1_4
GD2_16
TK4_31
TK4_23
GD5_66
TK1-1_85
TK1-1_80
TK1-1_89
TK1-1_71
GD3_34
GD1_3
GD5_62
GD3_16
TK1-1_10
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TK1-1_96
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TK1-1_31
TK1-1_24
TK1-1_47
TK1-1_51
TK1-1_43
TK1-1_48
GD3_35
TK1-1_23
TK3_24
GD1_1
GD5_45
TK4_37
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GD2_13
TK1-1_78
TK2_18
TK1-1_21
TK4_2
TK1-1_56
TK1-1_55
TK1-1_49
TK1-1_15
GD5_33
GD1_35
TK3_38
TK2_31
GD1_17
GD5_36
GD3_76
GD5_70
GD5_74
TK1-1_58
GD2_34
GD1_15
GD3_32
GD4_33
TK2_21
TK1-1_27
TK1-1_19
TK3_30
TK2_38
TK4_33
TK2_28
GD4_9
TK2_27
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TK1-2_16
TK1-1_45
GD3_56
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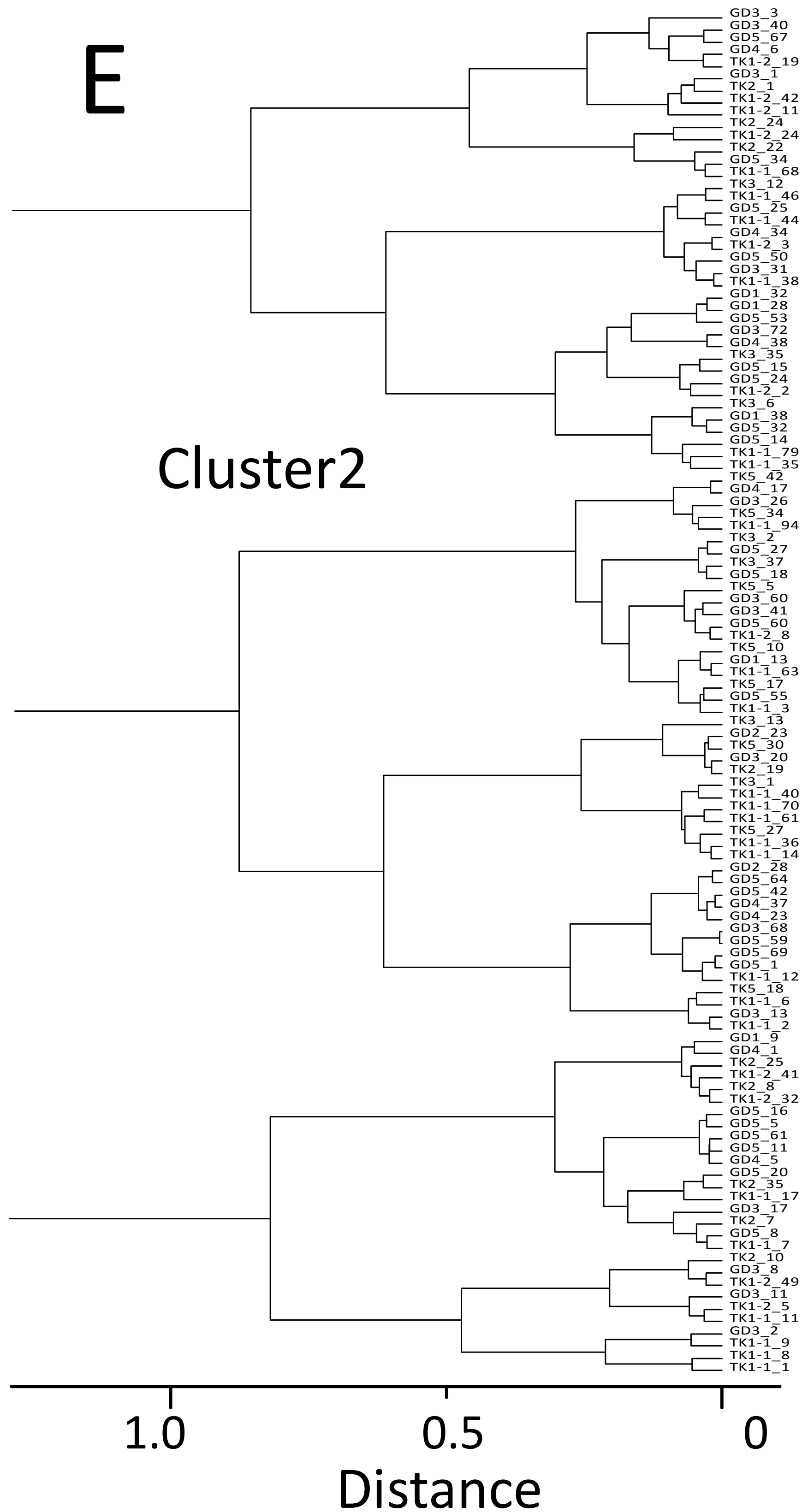
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GD1_38
GD5_32
GD5_14
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TK1-1_35
TK5_42
GD4_17
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TK3_2
GD5_27
TK3_37
GD5_18
TK5_5
GD3_60
GD3_41
GD5_60
TK1-2_8
TK5_10
GD1_13
TK1-1_63
TK5_17
GD5_55
TK1-1_3
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TK5_27
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Distance

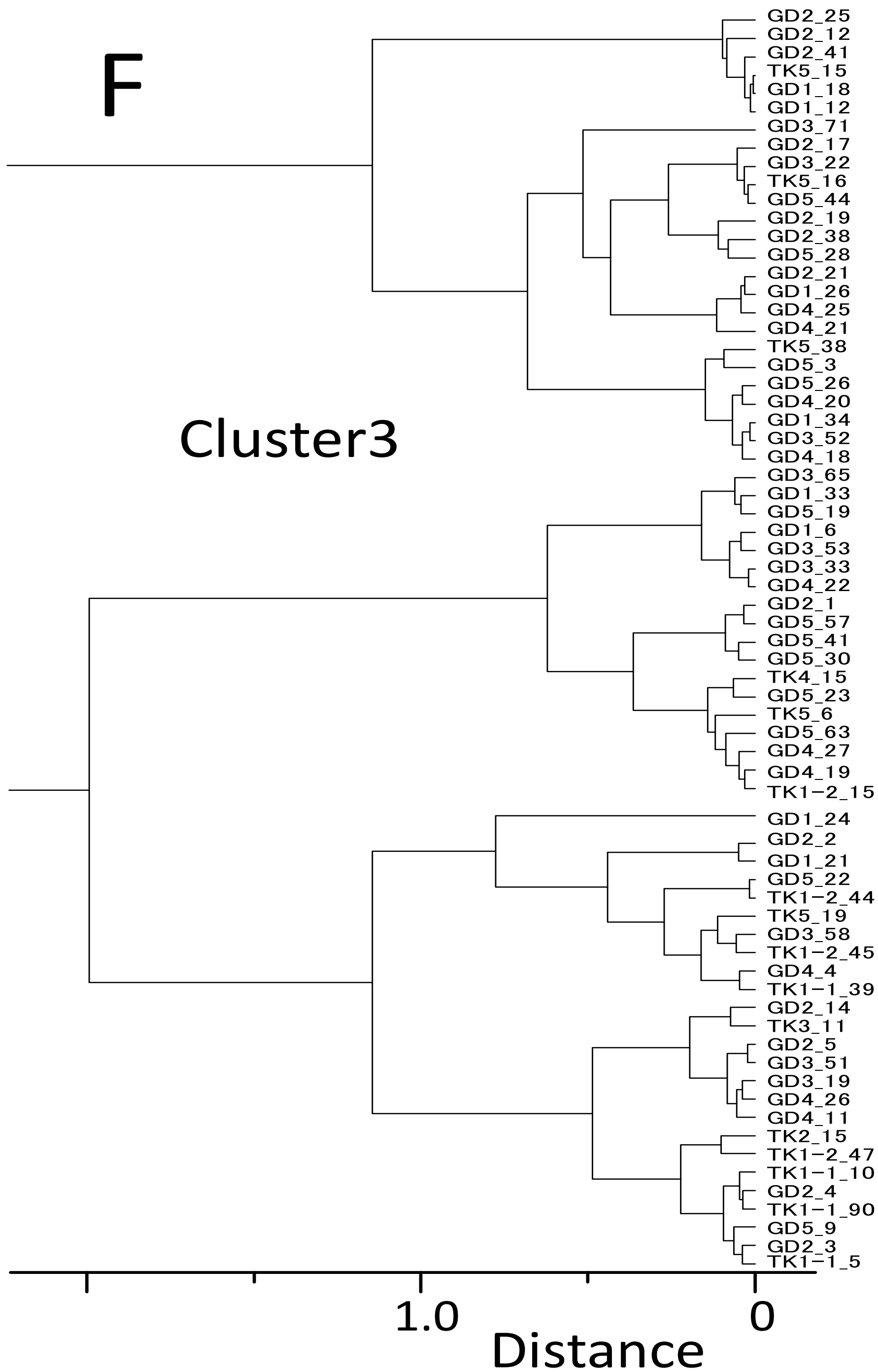

Figure DR4. (A) –(C) Tree diagram of cluster analysis using the fractional areas of five CL emission components (EC1–5) for quartz grains from Kunlun Mountains and the Gobi desert. The numbers of quartz grains are same to Table DR2.

TABLE DR1. LIST OF THE SAMPLING SITES

	Sample type	Area	Sample No.	Lat (deg North)	Long (deg East)	Elevation (m)
Taklimakan	Mountain Loess	Aqqan (Kunlun Mts.)	TK1-1(from the top) TK1-2 (from 9 m below the top)*	36.422	81.973	2556
	Mountain Loess	Near Karakashi River (Kunlun Mts.)	TK2 (from the top)	37.016	79.746	1511
	Riverbed sediment	Karakashi River (Kunlun Mts.)	TK3	37.254	79.761	1320
	Riverbed sediment	Gez river (Pamir plateau)	TK4	38.991	75.543	1803
	Riverbed sediment	Weigan-Kuqa river (Tianshan Mts.)	TK5	41.760	83.115	1098
Gobi	Sand dune	Khongoryn Els (Northern slope of Gobi-Altai Mts.)	GD1	43.765	102.254	1352
	Gobi	Near Mandalgobi (Northern part of Gobi Basin)	GD2	45.925	106.342	1420
	Gobi	Baruun Bayn Ulaan (between Hangayn and Gobi-Altai Mts.)	GD3	45.155	101.523	1235
	Gobi (dry up pool)	Tsogt-Ovoo (Central part of Gobi Basin)	GD4	44.346	105.257	1209
	Gobi (dry up pool)	Tsogt-Ovoo (Central part of Gobi Basin)	GD5	44.388	105.276	1218

* Based on total thick (45 m) of the loess sediment, bottom age (880 ka B.P) and sedimentation rate changes of the Aqqan loess (Fang et al., 2002), the age is estimated to be ca. 100–200 ka B.P.

TABLE DR2. RELATIVE INTENSITIES OF EACH EMISSION CENTERS AND DETERMINED CLUSTERS FOR QUARTZ GRAINS FROM KUNLUN MOUNTAINS AND THE GOBI DESERT

Sample No.	Quartz No.	Fractional area of each Gaussian curve (%)					Cluster
		EC1 (1.7 eV)	EC2 (1.9 eV)	EC3 (1.95 eV)	EC4 (2.7 eV)	EC5 (3.0-3.5 eV)	
TK1-1	1	18	4	50	0	27	2
TK1-1	2	22	3	44	0	30	2
TK1-1	3	26	0	37	0	36	2
TK1-1	4	32	2	51	2	13	1
TK1-1	5	9	2	15	5	69	3
TK1-1	6	20	3	42	2	33	2
TK1-1	7	20	2	45	7	26	2
TK1-1	8	15	4	55	0	26	2
TK1-1	9	8	6	47	3	35	2
TK1-1	10	24	6	46	8	17	1
TK1-1	11	20	4	33	7	35	2
TK1-1	12	25	5	41	0	29	2
TK1-1	13	31	4	51	2	12	1
TK1-1	14	25	4	35	8	28	2
TK1-1	15	34	4	44	6	12	1
TK1-1	16	33	5	39	0	24	1
TK1-1	17	24	5	40	10	22	2
TK1-1	18	30	6	44	0	20	1
TK1-1	19	35	5	46	5	10	1
TK1-1	20	31	6	38	8	18	1
TK1-1	21	35	4	42	4	15	1
TK1-1	22	44	3	34	0	18	1
TK1-1	23	36	5	43	6	10	1
TK1-1	24	39	4	45	5	8	1
TK1-1	25	35	4	42	1	18	1
TK1-1	26	38	5	47	4	7	1
TK1-1	27	35	5	46	4	9	1
TK1-1	28	38	4	48	4	5	1
TK1-1	29	38	5	45	6	6	1
TK1-1	30	45	3	41	5	6	1
TK1-1	31	40	4	43	5	7	1
TK1-1	32	34	5	49	7	5	1
TK1-1	33	38	4	46	5	7	1
TK1-1	34	47	4	38	3	8	1
TK1-1	35	24	2	23	5	46	2
TK1-1	36	25	4	35	9	27	2
TK1-1	37	37	5	44	6	8	1
TK1-1	38	25	3	26	8	38	2

Sample No.	Quartz No.	Fractional area of each Gaussian curve (%)					Cluster
		EC1 (1.7 eV)	EC2 (1.9 eV)	EC3 (1.95 eV)	EC4 (2.7 eV)	EC5 (3.0-3.5 eV)	
TK1-1	39	0	0	1	15	83	3
TK1-1	40	29	3	34	7	27	2
TK1-1	41	33	3	41	7	16	1
TK1-1	42	33	4	40	5	18	1
TK1-1	43	34	4	42	6	13	1
TK1-1	44	26	4	29	7	36	2
TK1-1	45	32	5	46	6	11	1
TK1-1	46	22	3	27	10	39	2
TK1-1	47	34	4	43	6	13	1
TK1-1	48	38	4	41	6	11	1
TK1-1	49	35	4	44	4	13	1
TK1-1	50	40	3	38	6	14	1
TK1-1	51	34	4	42	6	14	1
TK1-1	52	32	5	51	5	7	1
TK1-1	53	43	3	36	1	18	1
TK1-1	54	37	3	49	3	8	1
TK1-1	55	34	2	45	5	14	1
TK1-1	56	35	3	46	4	13	1
TK1-1	57	37	3	49	2	10	1
TK1-1	58	37	3	47	1	12	1
TK1-1	59	38	3	52	2	6	1
TK1-1	60	38	3	49	3	6	1
TK1-1	61	26	3	36	4	31	2
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GD4	39	35	3	30	2	31	1
GD5	1	27	6	41	0	26	2
GD5	2	36	6	41	1	16	1
GD5	3	25	1	16	2	56	3
GD5	4	27	4	52	2	16	1
GD5	5	22	6	49	4	19	2
GD5	6	29	4	43	0	24	1
GD5	7	40	0	42	0	17	1
GD5	8	21	4	43	6	26	2
GD5	9	4	2	13	7	74	3
GD5	10	23	4	49	7	16	1
GD5	11	23	5	47	4	21	2
GD5	12	37	3	26	0	34	1
GD5	13	28	4	47	5	16	1
GD5	14	26	1	25	0	48	2
GD5	15	17	3	29	7	43	2
GD5	16	22	5	47	6	20	2
GD5	17	28	6	45	4	17	1
GD5	18	28	6	35	1	30	2
GD5	19	9	3	28	4	55	3
GD5	20	26	5	44	4	22	2
GD5	21	31	5	39	4	21	1
GD5	22	4	1	9	16	70	3
GD5	23	18	2	14	8	58	3
GD5	24	17	4	33	5	41	2
GD5	25	24	5	30	6	37	2
GD5	26	33	1	11	0	55	3
GD5	27	28	3	36	1	32	2
GD5	28	33	12	0	0	56	3
GD5	29	40	4	44	0	12	1
GD5	30	17	2	16	0	64	3
GD5	31	39	4	40	2	16	1
GD5	32	19	3	22	6	50	2
GD5	33	34	4	50	0	12	1
GD5	34	18	2	26	11	44	2
GD5	35	41	3	38	0	18	1
GD5	36	35	4	47	0	14	1
GD5	37	40	3	42	1	13	1
GD5	38	31	6	52	1	10	1
GD5	39	43	3	44	0	10	1
GD5	40	39	3	41	0	17	1
GD5	41	13	3	17	0	66	3
GD5	42	27	3	39	4	26	2
GD5	43	36	4	42	1	17	1
GD5	44	38	0	0	0	62	3
GD5	45	35	4	45	1	15	1
GD5	46	43	4	46	0	7	1
GD5	47	37	4	45	1	13	1
GD5	48	44	1	29	0	25	1
GD5	49	33	0	24	0	42	1
GD5	50	23	2	25	9	41	2
GD5	51	39	4	29	0	27	1
GD5	52	39	3	50	2	7	1
GD5	53	24	5	27	1	43	2
GD5	54	42	3	44	0	11	1
GD5	55	25	3	36	1	35	2
GD5	56	37	2	41	0	20	1
GD5	57	18	3	19	0	60	3
GD5	58	41	3	48	0	8	1
GD5	59	29	3	41	0	27	2
GD5	60	26	3	34	5	33	2
GD5	61	23	4	48	3	22	2
GD5	62	35	4	50	2	9	1
GD5	63	8	2	19	13	58	3
GD5	64	27	3	39	2	29	2
GD5	65	25	4	54	0	17	1
GD5	66	41	2	46	0	10	1
GD5	67	14	3	32	1	50	2
GD5	68	39	3	45	0	13	1
GD5	69	27	5	41	0	27	2
GD5	70	35	4	49	0	13	1
GD5	71	37	4	38	1	21	1
GD5	72	30	4	49	1	17	1
GD5	73	37	3	37	0	23	1
GD5	74	36	4	47	0	13	1

TABLE DR3. RELATIVE INTENSITY OF EMISSION CENTERS FOR THREE CLUSTERS

Cluster No.	Fractional area of each Gaussian curve (%)				
	EC1 (1.7 eV)	EC2 (1.9 eV)	EC3 (1.95 eV)	EC4 (2.7 eV)	EC5 (3.0–3.5 eV)
Cluster 1	35 ± 7	4 ± 1	45 ± 8	2 ± 2	14 ± 6
Cluster 2	23 ± 6	3 ± 1	35 ± 8	5 ± 4	34 ± 8
Cluster 3	19 ± 12	2 ± 2	10 ± 8	4 ± 7	65 ± 10

TABLE DR4. CLUSTER COMPOSITIONS OF DESERTS' SAMPLES

	Sample No.	Measured grain number (n)	Cluster1 (%)	Cluster 2 (%)	Cluster 3 (%)	Average Cluster 1 (%)	Average Cluster 2 (%)	Average Cluster 3 (%)
Taklimakan	TK1-1	102	74 (64–81)*	22 (15–31)	4 (1–9)	72 [65–78]†	23 [17–30]	5 [3–10]
	TK1-2	49	69 (55–80)	23 (13–35)	8 (3–19)			
	TK2	39	74 (58–85)	23 (12–38)	3 (1–13)			
	TK3	40	80 (65–89)	18 (8–32)	2 (1–12)	82 [74–87]	12 [8–19]	6 [3–12]
	TK4	39	97 (86–99)	0 (0–8)	3 (1–13)			
	TK5	42	69 (53–80)	19 (10–33)	12 (5–25)			
Gobi	GD1	38	66 (49–78)	13 (5–27)	21 (11–36)	60 [54–66]	18 [14–24]	22 [17–27]
	GD2	41	63 (48–76)	5 (1–16)	32 (19–47)			
	GD3	76	68 (57–77)	20 (12–30)	12 (6–21)			
	GD4	39	54 (38–68)	20 (10–35)	26 (14–41)			
	GD5	74	51 (40–62)	33 (22–43)	16 (9–26)			

*In parentheses: 95% confidential interval

†In brackets: propagation error