Supplementary file 1 | Procedures for bulk-XRD analyses

In this study, we determined the bulk mineral composition of core samples by X-ray diffraction (XRD) analyses. Preliminary XRD and smear slide observations conducted onboard suggested that the samples contained clay minerals (smectite, illite, kaolinite, and chlorite), quartz, and plagioclase, but also included considerable amounts of amorphous material such as volcanic glass and biogenic opal (Chester et al., 2012). To quantify the mineral composition, including the amorphous fraction, we used calcite as an internal standard and processed the experimental patterns using a newly prepared calibration curve. The original sediment did not contain calcite, because the sample was recovered from the seafloor below the carbonate compensation depth.

Methods

Powder samples, preheated in an oven at 60 °C for 3 hours, were mixed with the internal standard (calcite) at a ratio of approximately 1:3 in an agate mortar. XRD patterns for these mixtures were recorded using a Rigaku Rint-Ultima 2100 with a one-dimensional X-ray detector (D/teX Ultra, Rigaku) at 40 kV and 30 mA, with 0.5° and 10 mm divergence slits and a 8 mm anti-scattering slit, at a scan rate of 1° per minute at 0.02° steps in 2θ. Diagnostic peaks for individual mineral species used for quantitative analyses were as follows: clay minerals at ~19.8°2θ, quartz at 26.65°2θ, plagioclase at 27.77°–28.02°2θ,; and calcite at 29.42°2θ. These peaks were processed for each experimental pattern (linear background subtraction and counting of integrated peak intensity) using standard Rigaku peak analysis software. From the obtained patterns, we first determined the relative abundance of each crystalline phase in the original sample (i.e., clay minerals, quartz, and plagioclase) with respect to calcite. Because we knew the

absolute amount of calcite in the powder, the relative wt.% could be recalculated to obtain the absolute value. The amorphous content was then estimated by subtracting the total abundance of crystalline phases from 100%.

In this study we calibrated the experimental conditions by measuring 12 standard mixtures composed of clay composite (montmorillonite + illite + chlorite + kaolinite), quartz (Kanto Chemical Co., Inc.), plagioclase (Ca-albite; Nichica Inc.), and calcite (Kanto Chemical Co., Inc.) with variable wt.% combinations (Table 1). The clay composite was prepared by mixing 45 wt.% montmorillonite (SWy-2; The Clay Minerals Society (CMS)), 40 wt.% illite (IMt-2; CMS), 10 wt.% chlorite (chlorite schist from the Besshi area, Sambagawa metamorphic belt, Japan), and 5 wt.% kaolinite (KGa-2; CMS). XRD patterns for the standard mixtures were recorded using the same measurement settings as for bulk sample measurement. The correlation between the integrated peak intensity and the weight fraction of each mineral is represented by a straight line (Fig. 1).

References

1. Chester, F.M., Mori, J.J., Toczko, S., Eguchi, N., and the Expedition 343/343T Scientists, 2012. Japan Trench Fast Drilling Project (JFAST). *IODP Prel. Rept.,* 343/343T. doi:10.2204/iodp.pr.343343T.2012

Sample Name	Total Clay		Quartz		Plagioclase		Calcite	
	Fraction (wt.%)	Integrated intensity	Fraction (wt.%)	Integrated intensity	Fraction (wt.%)	Integrated intensity	Fraction (wt.%)	Integrated intensity
1	5	140	10	7177	15	12062	70	41871
2	10	624	15	20281	70	88337	5	4581
3	15	781	70	84155	5	6163	10	7228
4	70	3300	5	7382	10	7521	15	12111
5	25	1157	25	28141	25	30165	25	18791
6	40	1694	40	43666	10	12370	10	7664
7	10	445	10	10107	40	48970	40	33682
8	50	2194	35	39261	5	7069	10	8605
9	10	269	30	32018	10	13259	50	34665
10	80	3783	10	14296	5	4357	5	4270
11	10	424	50	62151	30	40258	10	8657
12	5	201	10	9986	50	54783	35	24869

Table 1. Standard mixtures and integrated peak intensities for each mineral species

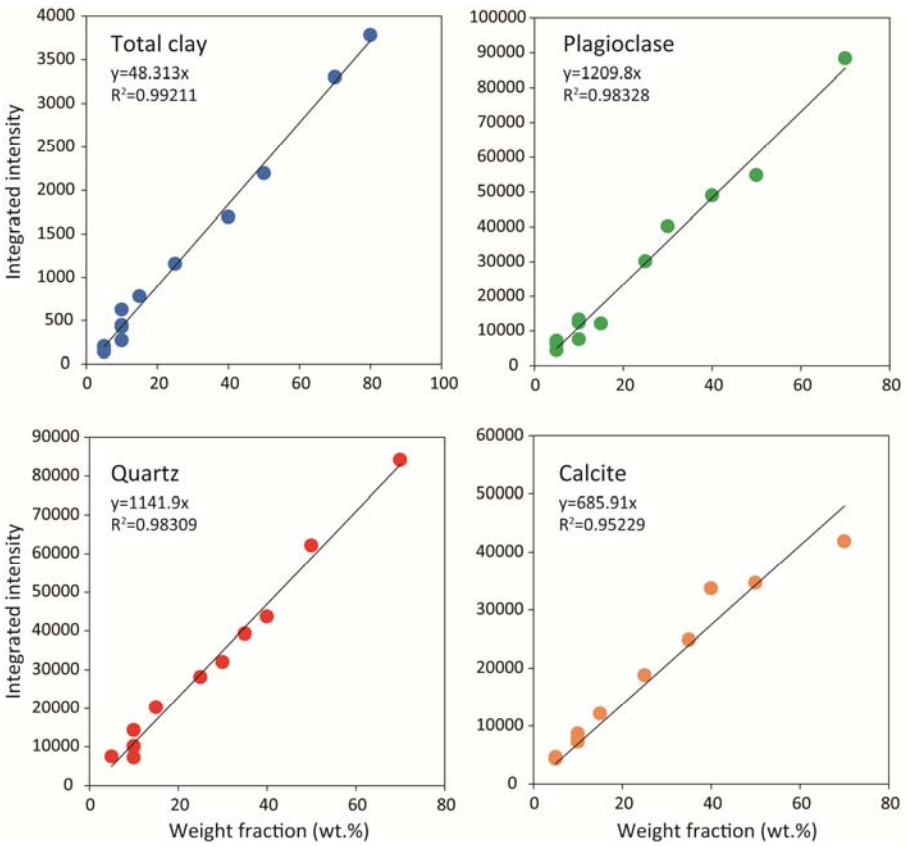

Figure 1. Calibration curves from measurements of standard mixtures of total clay, quartz, plagioclase, and calcite.

Supplementary Data Table 1 | Results of X-ray diffraction for JFAST samples

Unit	Core	Section	Interval (cm)	Depth (mbsf)	Wt.% in clay fraction			Wt.% in bulk sample				Smectite content in bulk sample (wt.%)
					Smectite	Illite	Kaolinite	Total clay	Quartz	Plagioclase	Amorphous	
1	1R	3w	114_117	180.45	81.0	13.9	5.1	35.0	7.1	3.6	54.4	28.3
1	1R	5w	9_12	181.875	82.2	12.6	5.2	24.7	7.1	4.2	64.1	20.3
2	2R	1w	53_56	648.53	70.7	23.0	6.3	24.3	6.9	4.6	64.2	17.2
2	2R	1w	99_102	648.99	73.4	22.8	3.8	38.0	6.2	1.8	53.9	27.9
2	2R	2w	114_117	650.55	79.6	17.5	3.0	39.9	7.5	2.6	49.9	31.8
2	2R	2w	44_47	650.55	66.1	27.3	6.6	39.9	7.5	2.6	49.9	26.4
2	3R	2w	59_62	654.84	60.0	28.0	12.1	24.6	9.0	3.5	63.0	14.7
3	3R	2w	73_76	654.99	79.1	17.5	3.3	42.4	8.9	3.0	45.7	33.5
3	4R	1w	117.5_121	689.23	47.2	36.3	16.5	19.2	8.7	3.5	68.7	9.1
3	5R	1w	77_80	697.175	63.1	25.5	11.3	24.1	10.4	4.4	61.2	15.2
3	5R	2w	1_4	698.17	56.6	29.4	14.0	21.9	8.4	4.5	65.2	12.4
3	6R	2w	3_6	705.33	69.2	20.3	10.5	15.7	6.0	2.9	75.3	10.9
3	7R	2w	111_115	715.25	65.8	23.7	10.5	29.8	9.1	4.1	57.1	19.6
3	8R	2w	113_117	721.545	64.1	22.1	13.8	24.0	10.9	4.9	60.2	15.4
3	8R	3w	27_32	722.085	55.2	29.5	15.3	23.8	9.3	5.0	61.9	13.1
3	8R	3w	93_103	722.745	55.4	30.5	14.1	31.2	11.2	4.9	52.7	17.3
3	9R	1w	5_11	724.55	63.5	24.7	11.8	26.9	10.5	4.4	58.2	17.1
3	10R	1w	61_64	770.61	53.3	31.5	15.2	24.2	8.5	4.1	63.3	12.9
3	10R	1w	88_97.5	770.88	48.9	34.1	17.0	23.7	8.4	4.9	63.0	11.6
3	10R	cc	29_32	772.285	52.6	32.3	15.0	24.4	8.7	5.2	61.7	12.8
3	12R	2w	48_52	787.35	61.3	22.5	16.1	15.5	10.5	5.1	68.8	9.5
3	14R	1w	3_6	810.0	51.3	33.3	15.4	27.0	9.6	3.8	59.6	13.8
3	15R	1w	28_34	816.8	74.2	20.3	5.5	23.9	5.7	3.3	67.1	17.7
3	16R	1w	5_10	818.6	56.5	29.4	14.1	25.3	8.9	5.1	60.6	14.3
4	17R	1w	3_5	821.5	88.8	9.5	1.7	90.2	7.3	2.5	0.0	80.1
4	17R	1w	7_10	821.6	90.9	7.3	1.8	90.6	6.9	2.6	0.0	82.3
4	17R	1w	17_19	821.7	84.1	12.6	3.3	75.8	7.9	2.1	14.3	63.7
4	17R	1w	21_23	821.7	84.2	13.1	2.7	78.5	7.7	2.3	11.5	66.0
4	17R	1w	24_26	821.7	89.4	9.0	1.6	67.9	5.8	2.5	23.7	60.7
4	17R	1w	40_42	821.9	68.5	25.6	5.9	36.6	8.4	2.6	52.3	25.1
4	17R	1w	53_55	822.0	77.9	19.0	3.1	73.7	6.9	2.4	17.0	57.4
4	17R	1w	58_63	822.1	74.2	22.2	3.5	90.6	7.8	1.6	0.0	67.2
4	17R	1w	58_70	822.1	85.6	12.3	2.1	90.5	7.7	1.8	0.0	77.4
4	17R	1w	67.5_69	822.2	81.2	16.0	2.9	90.4	7.6	2.0	0.0	73.4
4	17R	1w	70_75	822.2	91.9	5.3	2.7	91.2	6.7	2.1	0.0	83.9
4	17R	1w	79_79	822.3	87.2	10.4	2.4	92.4	5.5	2.1	0.0	80.5
4	17R	1w	84_85	822.3	93.9	3.6	2.6	79.9	5.4	1.7	13.0	75.0
4	17R	1w	96_98	822.5	77.8	19.2	3.0	89.5	8.0	2.5	0.0	69.7

5	18R	1w	29_32	824.3	75.3	20.6	4.0	52.7	7.8	1.8	37.7	39.7
5	18R	1w	58_64	824.6	72.3	23.6	4.1	52.1	8.7	2.2	37.0	37.7
5	18R	1w	98_101	825.0	69.2	26.0	4.8	49.4	7.9	2.8	39.9	34.2
5	19R	1w	14_17	826.6	74.3	21.8	3.9	29.6	6.2	2.5	61.6	22.0
5	19R	1w	69_72	827.2	68.8	26.1	5.1	27.7	7.0	2.9	62.4	19.1
5	19R	2w	39_42	828.1	58.7	34.7	6.6	36.4	8.8	2.1	52.7	21.4
5	20R	1w	71_74	831.7	65.2	29.3	5.6	24.2	5.8	2.6	67.3	15.8
5	20R	1w	89_102.5	831.9	64.8	29.0	6.2	30.8	5.8	2.1	61.3	20.0
5	20R	1w	122_130	832.2	68.6	26.1	5.3	39.4	7.9	3.3	49.4	27.0
6	20R	2w	56_59	832.9	95.0	5.0	0.0	5.0	18.3	0.0	76.7	4.7
6	20R	2w	64_64.5	833.0	95.1	4.9	0.0	65.1	4.5	1.4	29.1	61.9
6	20R	2w	80_81	833.1	96.2	3.8	0.0	30.3	9.2	0.0	60.5	29.1
6	20R	2w	94_97	833.3	97.2	2.8	0.0	18.8	21.8	0.0	59.4	18.3
6	20R	2w	94_97	833.3	95.0	5.0	0.0	8.3	18.6	0.0	73.1	7.9
6	20R	2w	104_107	833.4	99.2	0.8	0.0	65.6	4.3	1.1	29.0	65.1

Supplementary Data Table 2| Crystallographic features of smectite in Core 17

Unit	Core	Section	Interval (cm)	Depth (mbsf)	FWHM of 001	%S in smectite-illite packet[a]	*b* length (Å)[b]
4	17R	1w	3_5	821.5	0.873	89.1	1.5026
4	17R	1w	7_10	821.6	0.847	89.7	1.5021
4	17R	1w	17_19	821.7	0.952	85.9	1.5025
4	17R	1w	21_23	821.7	0.947	84.0	1.5018
4	17R	1w	24_26	821.7	0.972	87.1	1.5029
4	17R	1w	40_42	821.9	1.354	71.5	1.5031
4	17R	1w	53_55	822.0	1.087	80.2	1.5022
4	17R	1w	58_63	822.1	1.025	79.0	1.5027
4	17R	1w	58_70	822.1	0.991	77.7	n.d.
4	17R	1w	67.5_69	822.2	1.020	81.9	1.5022
4	17R	1w	70_75	822.2	1.157	84.1	1.5024
4	17R	1w	79_79	822.3	0.991	n.d.	1.503
4	17R	1w	84_85	822.3	1.063	80.3	1.5021
4	17R	1w	96_98	822.5	1.070	79.9	1.5025

[a]%S in smectite-illite packet was determined by saddle/peak ratio of smectite 001 and by using a working curve of

Inoue et al. (1989)

[b]*b* cell dimension was determined from smectite 060 reflections

Reference

1. Inoue, A., A., Bouchet, B., Velde, & A., Meunier, 1989. Convenient technique for estimating smectite layer percentage in randomly interstratified illite/smectite minerals. *Clays and Clay Minerals* 37, 227–234 (1989).

Supplementary Data Table 3| Results of X-ray diffraction for DSDP samples

Core	Section	Interval (cm)	Depth (mbsf)	Wt.% in clay fraction			Wt.% in bulk sample				Smectite content in bulk sample (wt.%)
				Smectite	Illite	Kaolinite	Total clay	Quartz	Plagioclase	Amorphous	
33R	1w	90.0-94.0	303.4	77.1	19.6	3.4	16.8	3.8	1.4	77.9	12.9
34R	1w	121.0-125.0	313.21	70.3	26.2	3.5	39.9	7.8	1.7	50.6	28.0
35R	1w	77.0-81.0	322.27	69.3	25.4	5.2	53.4	10.0	2.7	33.9	37.0
36R	1w	91.0-95.0	331.91	78.9	17.6	3.5	58.8	7.9	1.9	31.5	46.4
37R	1w	16.0-19.0	340.66	73.3	21.8	4.9	59.7	8.3	1.6	30.4	43.7
38R	1w	101.0-105.0	351.01	86.1	11.8	2.2	36.8	4.1	1.2	57.9	31.7
38R	6w	31.0-35.0	357.8	82.2	14.7	3.2	39.8	5.0	1.6	53.7	32.7
38R	6w	44.5-48.5	357.9	86.7	11.7	1.6	26.1	3.3	2.0	68.6	22.6
39R	1w	92.0-96.0	360.42	94.1	4.7	1.2	69.3	5.9	2.1	22.7	65.2
39R	2w	22.0-26.0	361.22	93.6	5.6	0.8	82.1	4.6	3.2	10.1	76.9
39R	3w	19.0-23.0	362.69	91.1	7.9	1.1	86.2	5.5	2.1	6.2	78.5
39R	4w	74.5-78.5	364.745	93.5	5.6	0.8	81.8	4.7	2.0	11.5	76.5
39R	5w	40.5-45	365.8	88.1	10.3	1.6	64.1	7.5	2.0	26.4	56.5
39R	5w	40.5-45	365.8	83.8	13.6	2.6	76.4	9.2	2.1	12.2	64.0
40R	1w	110.0-114.0	370.1	77.7	16.9	5.3	62.1	7.8	1.6	28.4	48.3
40R	3w	55.0-59.0	372.55	69.8	24.6	5.6	66.0	7.2	1.1	25.8	46.1
40R	4w	26.0-30.3	373.76	65.4	28.3	6.3	64.8	7.0	1.4	26.8	42.3
40R	5w	31.5-35.5	375.315	77.4	18.8	3.8	57.6	5.2	1.1	36.1	44.6
40R	6w	28.0-35.0	376.18	76.2	19.6	4.2	84.9	8.0	1.7	5.4	64.7
40R	7w	17.0-21.0	377.47	85.1	11.9	3.0	66.3	4.9	2.5	26.2	56.4
41R	1w	15.0-19.0	378.65	95.8	4.2	0.0	37.1	33.8	0.0	29.1	35.6