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Supporting Information

Improved Thermoelectric Performance of Co-Doped β-$FeSi_2$ by Ni Substitution

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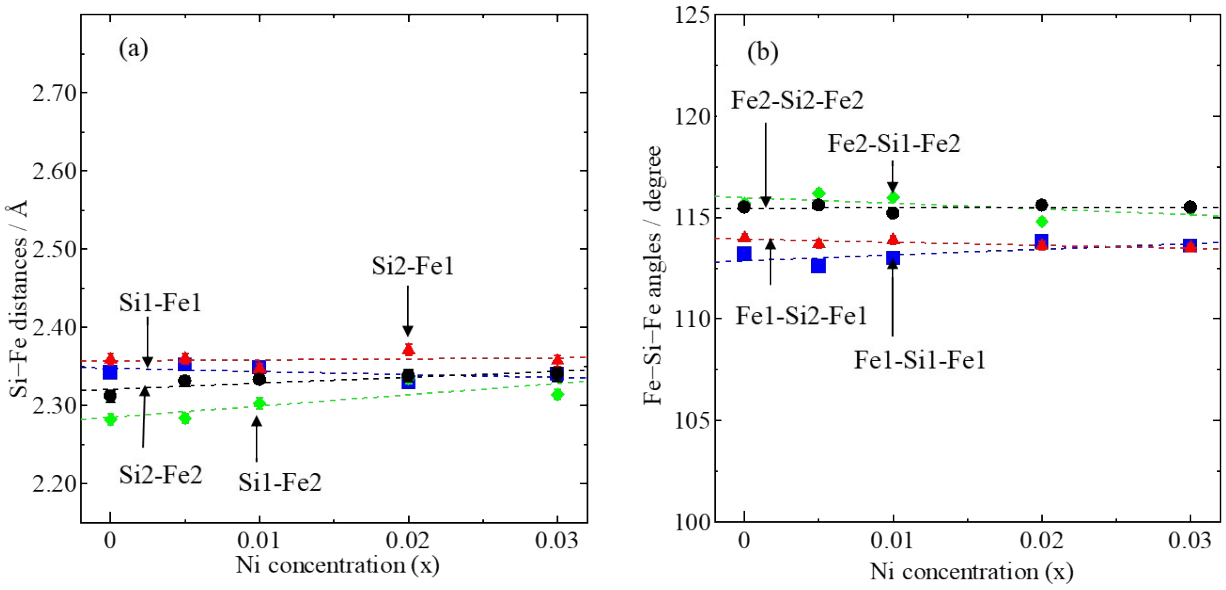

Figure S1. (a) Interact atomic Fe-Si distance and **(b)** interact atomic Fe-Si-Fe angle with x dependence.

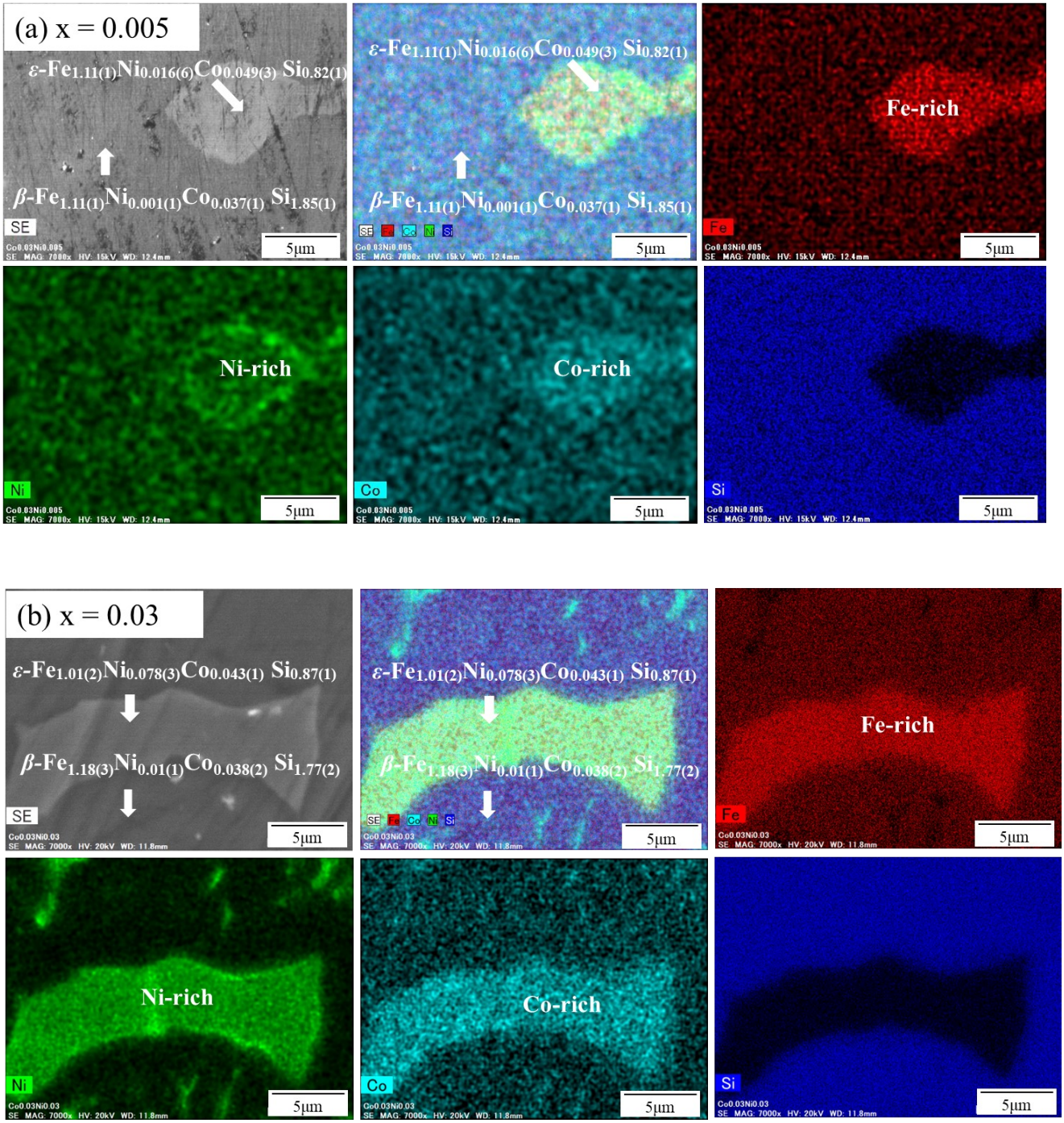

Figure S2. SEM-EDS mapping of β-$Fe_{1-x}Ni_xCo_{0.03}Si_2$, (a) x = 0.005, (b) x = 0.03, after heat treatment with separated mapping of each element, where Fe is mapped in red, Ni in green, Co in cyan, and Si in blue color.

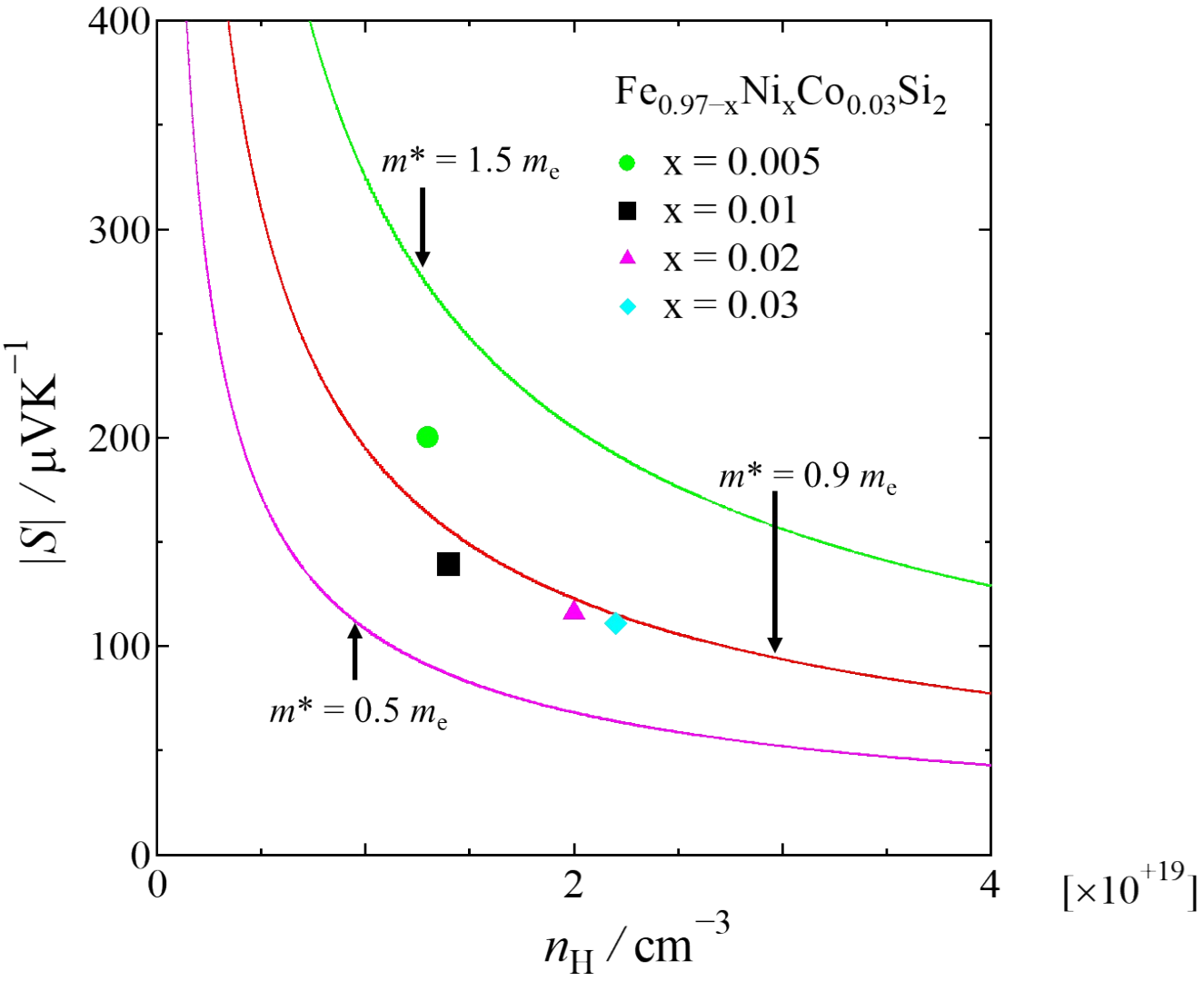

Figure S3. Absolute Seebeck coefficient $|S|$ versus carrier concentration (n_H) at room temperature, where the solid curves are the calculated data estimated by using Mott's formula at various effective masses (m^* = x m_e, where x is variable and m_e is the static mass of the electron, i.e., 9.10938×10^{-31} kg).

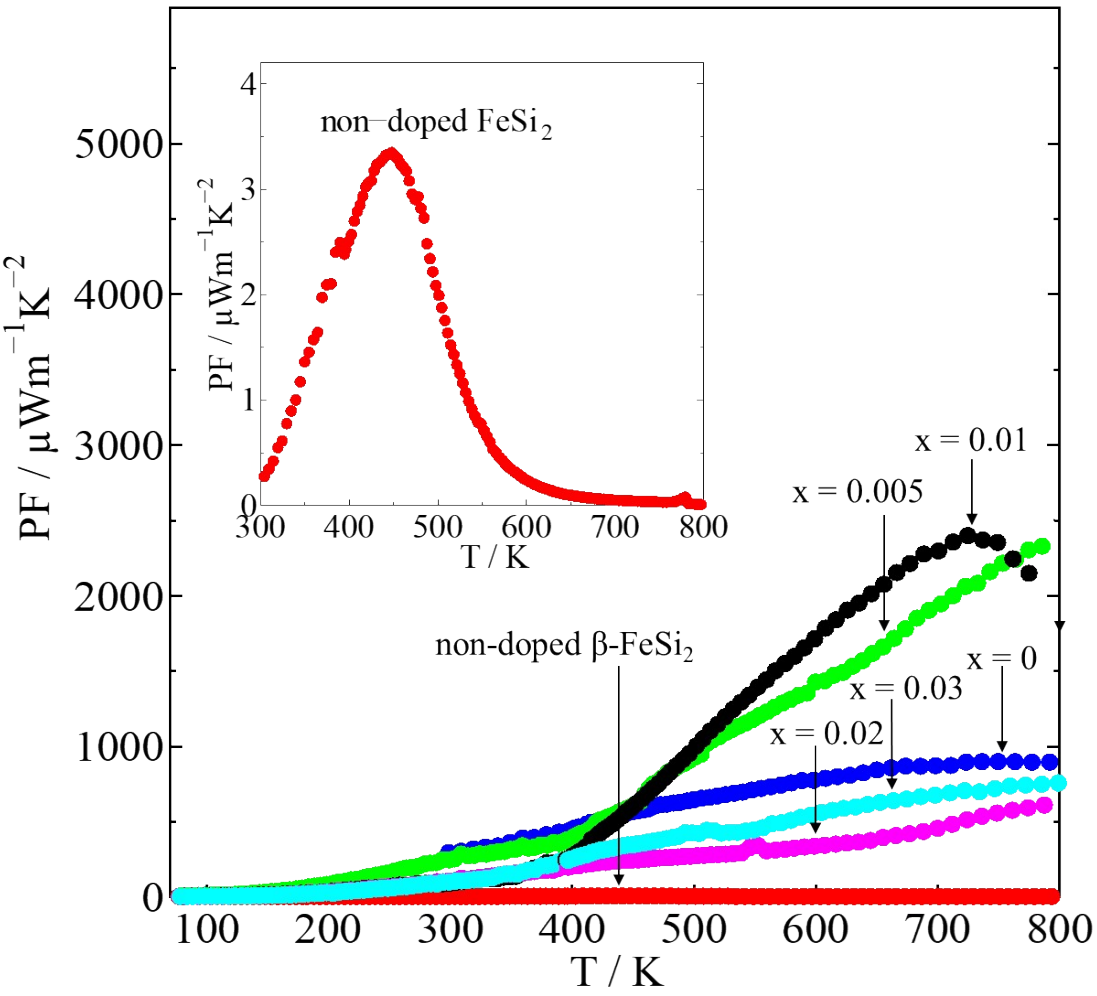

Figure S4. Power factor (PF = S^2/ρ) of non-doped β-$FeSi_2$ and β-$Fe_{0.97-x}Ni_xCo_{0.03}Si_2$ ($0 \leq x \leq 0.03$) with temperature dependence, where the inset magnifies the data of non-doped β-$FeSi_2$.

Table S1 Crystal structure parameters of non-doped β-$FeSi_2$ and β-$Fe_{0.97-x}Ni_xCo_{0.03}Si_2$ ($0 \leq x \leq 0.03$) at room temperature.

Sample		non-doped $FeSi_2$	$Fe_{0.97-x}Ni_xCo_{0.03}Si_2$				
			0	0.005	0.01	0.02	0.03
Space group		*Cmce*	*Cmce*	*Cmce*	*Cmce*	*Cmce*	*Cmce*
a (Å)		9.8788(5)	9.8864(6)	9.8915(5)	9.8929(5)	9.8866(9)	9.8851(4)
b (Å)		7.8008(4)	7.8003(5)	7.8015(4)	7.8020(4)	7.7988(4)	7.7965(3)
c (Å)		7.8372(4)	7.8344(5)	7.8390(4)	7.8412(5)	7.8348(7)	7.8347(4)
V (Å^3)		603.96(5)	604.16(7)	604.93(5)	605.22(6)	604.1(1)	603.82(5)
Fe1	*x*	0.2160(2)	0.2168(3)	0.2167(3)	0.2178(3)	0.2171(3)	0.2178(4)
	y	0	0	0	0	0	0
	z	0	0	0	0	0	0
	B (Å^2)	0.1	0.1	0.1	0.1	0.1	0.1
	g	1.000	0.970	0.965	0.960	0.950	0.940
Co1	*x*	-	0.2168(3)	0.2167(3)	0.2178(3)	0.2171(3)	0.2178(4)
	y	-	0	0	0	0	0
	z	-	0	0	0	0	0
	B (Å^2)	-	0.1	0.1	0.1	0.1	0.1
	g	-	0.030	0.030	0.030	0.030	0.030
Ni1	*x*	-	-	0.2167(3)	0.2178(3)	0.2171(3)	0.2178(4)
	y	-	-	0	0	0	0.000
	z	-	-	0	0	0	0.000
	B (Å^2)	-	-	0.1	0.1	0.1	0.100
	g	-	-	0.005	0.010	0.020	0.030
Fe2	*x*	1/2	1/2	1/2	1/2	1/2	1/2
	y	0.3014(4)	0.3017(5)	0.3006(4)	0.3028(5)	0.3033(4)	0.3034(5)
	z	0.1940(4)	0.1949(4)	0.1949(4)	0.1948(5)	0.1941(4)	0.1942(5)
	B (Å^2)	0.1	0.1	0.1	0.1	0.1	0.1
	g	1.000	0.970	0.965	0.960	0.950	0.940
Co2	*x*	-	1/2	1/2	1/2	1/2	1/2
	y	-	0.3017(5)	0.3006(4)	0.3028(5)	0.3033(4)	0.3034(5)
	z	-	0.1949(4)	0.1949(4)	0.1948(5)	0.1941(4)	0.1942(5)
	B (Å^2)	-	0.1	0.1	0.1	0.1	0.1
	g	-	0.030	0.030	0.030	0.030	0.030
Ni2	*x*	-	-	1/2	1/2	1/2	1/2
	y	-	-	0.3006(4)	0.3028(5)	0.3033(4)	0.3034(5)
	z	-	-	0.1949(4)	0.1948(5)	0.1941(4)	0.1942(5)
	B (Å^2)	-	-	0.1	0.1	0.1	0.100
	g	-	-	0.005	0.010	0.020	0.030

Sample		non-doped $FeSi_2$	$Fe_{0.97-x}Ni_xCo_{0.03}Si_2$				
			0	0.005	0.01	0.02	0.03
Space group		*Cmce*	*Cmce*	*Cmce*	*Cmce*	*Cmce*	*Cmce*
Si1	*x*	0.1217(5)	0.1239(7)	0.1226(6)	0.1238(7)	0.1267(7)	0.1255(7)
	y	0.2811(7)	0.2807(8)	0.2809(7)	0.2792(8)	0.2804(8)	0.2788(9)
	z	0.0394(4)	0.0367(5)	0.0382(4)	0.0401(5)	0.0432(5)	0.0412(6)
	B (Å^2)	0.3	0.3	0.3	0.3	0.3	0.3
	g	1.0	1.0	1.0	1.0	1.0	1.0
Si2	*x*	0.3761(5)	0.3749(7)	0.3752(6)	0.3742(7)	0.3748(7)	0.3748(7)
	y	0.0399(5)	0.0380(6)	0.0400(5)	0.0413(6)	0.0440(6)	0.0440(7)
	z	0.2220(6)	0.2223(8)	0.2216(6)	0.2213(8)	0.2237(7)	0.2221(5)
	B (Å^2)	0.3	0.3	0.3	0.3	0.3	0.3
	g	1.0	1.0	1.0	1.0	1.0	1.0
R_{wp} (%)		3.316	3.474	2.907	3.243	3.194	3.408
R_P (%)		2.108	2.309	1.937	2.016	1.998	2.144
R_R (%)		29.041	34.348	30.814	32.950	31.802	34.86
R_e (%)		0.792	0.784	1.453	1.475	0.803	0.799
R_B (%)		8.543	10.058	8.408	10.119	9.56	11.498
R_F (%)		8.603	8.977	7.901	7.361	7.609	9.43
$S = R_{wp} / R_e$		4.187	4.431	2.001	2.199	3.978	4.265
Si1 - Fe1 / Co1/Ni1 (Å)		2.361(5)	2.342(6)	2.352(6)	2.349(7)	2.329(6)	2.340(7)
Si1 - Fe1 / Co1/Ni1 (Å)		2.402(6)	2.392(7)	2.399(6)	2.389(7)	2.386(7)	2.379(8)
Si1 - Fe2 / Co2/Ni2 (Å)		2.282(5)	2.282(7)	2.284(6)	2.303(7)	2.335(7)	2.314(7)
Si1 - Fe2 / Co2/Ni2 (Å)		2.415(4)	2.438(5)	2.422(5)	2.419(6)	2.416(6)	2.423(6)
Fe1 / Co1/Ni1 - Si1 - Fe1 / Co1/Ni1 (deg.)		112.3(2)	113.2(3)	112.6(2)	113.0(2)	113.8(3)	113.6(3)
Fe2 / Co2/Ni2 - Si1 - Fe2 / Co2/Ni2 (deg.)		116.6(2)	115.7(2)	116.2(2)	116.0(2)	114.8(2)	115.4(2)
Si2 - Fe1 / Co1/Ni1 (Å)		2.372(5)	2.359(7)	2.360(6)	2.347(7)	2.371(7)	2.357(7)
Si2 - Fe1 / Co1/Ni1 (Å)		2.381(5)	2.375(6)	2.384(5)	2.389(6)	2.372(6)	2.387(7)
Si2 - Fe2 / Co2/Ni2 (Å)		2.322(6)	2.312(8)	2.331(7)	2.333(3)	2.338(8)	2.340(9)
Si2 - Fe2 / Co2/Ni2 (Å)		2.388(5)	2.409(6)	2.387(5)	2.398(6)	2.381(6)	2.380(7)
Fe1 / Co1/Ni1 - Si2 - Fe1 / Co1/Ni1 (deg.)		113.5(2)	114.0(2)	113.7(2)	113.9(2)	113.6(2)	113.5(2)
Fe2 / Co2/Ni2 - Si2 - Fe2 / Co2/Ni2 (deg.)		116.1(2)	115.5(2)	115.6(2)	115.2(2)	115.6(2)	115.5(2)

Table S1 Continued.