Supporting Information for

Parameter Visualization of Benchtop Nuclear Magnetic Resonance Spectra Toward Food Process Monitoring

Koki Hara [1,2], Shunji Yamada [2], Eisuke Chikayama [2,3] and Jun Kikuchi [1,2,4,*]

[1] Graduate School of Medical Life Science, Yokohama City University, 1-7-29 Suehiro-cho, Tsurumi-ku, Yokohama, Kanagawa 230-0045, Japan; koki.hara@a.riken.jp
[2] RIKEN Center for Sustainable Resource Science, 1-7-22 Suehiro-cho, Tsurumi-ku, Yokohama, Kanagawa 230-0045, Japan; shunji.yamada@riken.jp (S.Y.); chikaya@nuis.ac.jp (E.C.)
[3] Department of Information Systems, Niigata University of International and Information Studies, 3-1-1 Mizukino, Nishi-ku, Niigata-shi, Niigata 950-2292, Japan
[4] Graduate School of Bioagricultural Sciences, Nagoya University, 1 Furo-cho, Chikusa-ku, Nagoya, Aichi 464-0810, Japan
* Correspondence: jun.kikuchi@riken.jp; Tel.: +81-45-508-7220

Analytical tools are available at https://github.com/riken-emar/TransferSimulationNMRspectra.

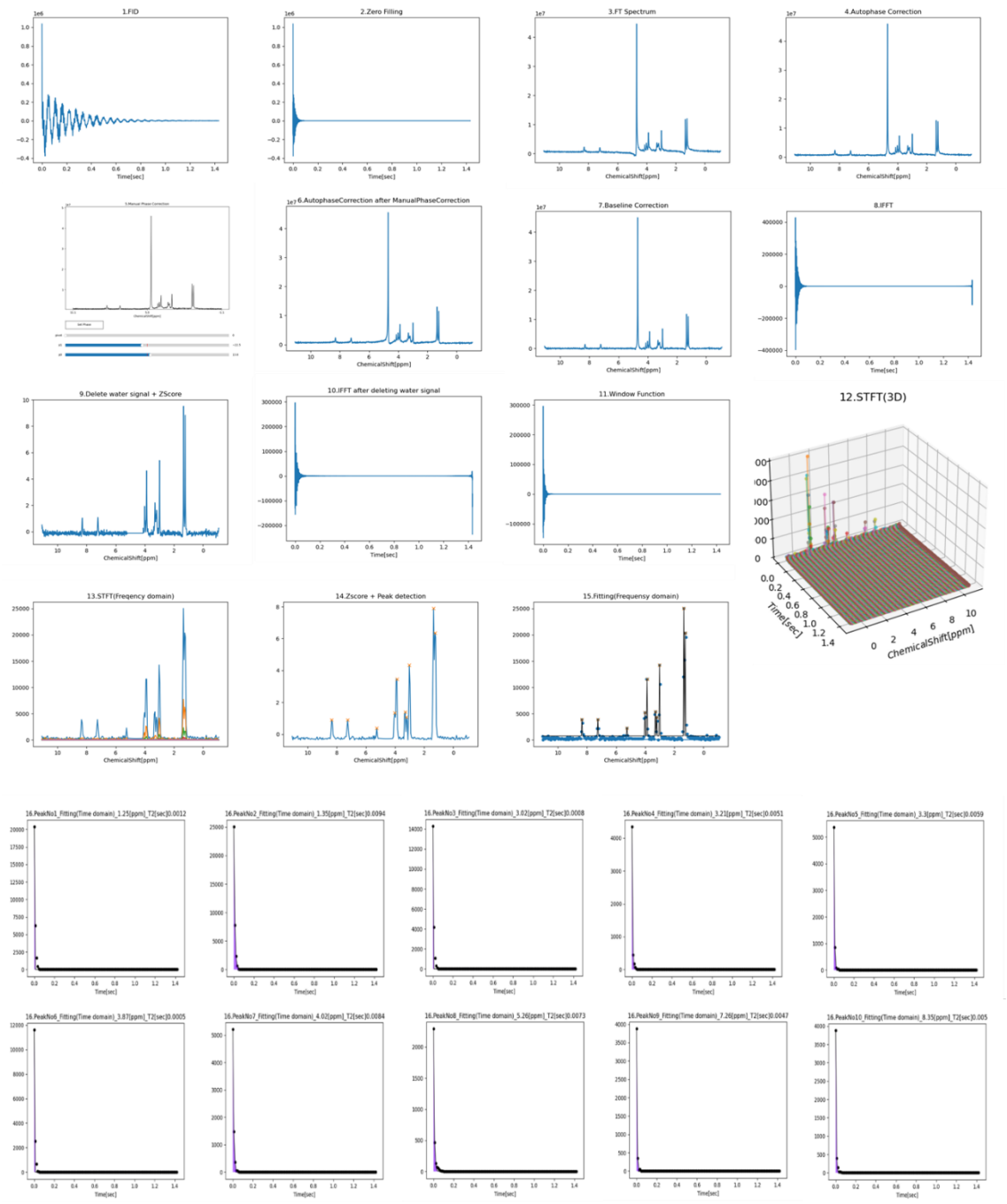

Figure S1. Output of the proposed data processing in tuna's ^{1}H NMR (nuclear magnetic resonance) data. NMR data are processed as follows. 1) original free-induced decay, 2) zero filling, 3) Fourier transform, 4) autophase correction, 5) manual phase correction, 6) autophase correction after manual phase correction, 7) baseline correction, 8) inverse Fourier transform, 9) water signal deletion and z-score calculation, 10) inverse Fourier transform after water signal deletion, 11) window function processing, 12) short-time Fourier transform (STFT), 13) visualization of frequency domain of STFT processed data, 14) z-score calculation, and peak detection, 15) fitting of frequency domain, and 16) visualization of peaks in time domain.

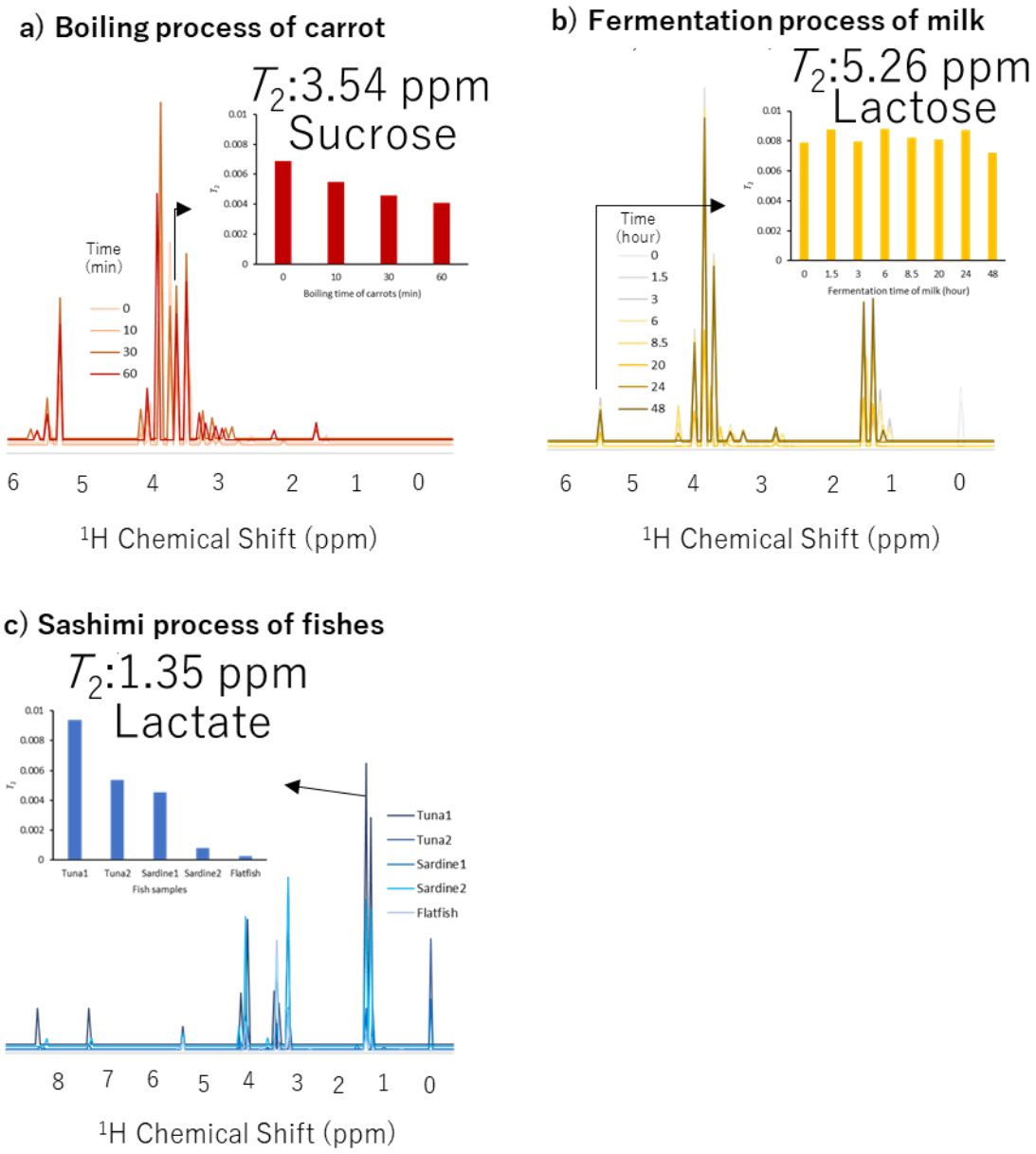

Figure S2. Parameter visualization of ^{1}H NMR spectra in primary industry processes. An example of parameter visualization of chemical shifts and T_2 for NMR datasets for fishery, livestock, and agriculture is shown. a) NMR dataset of sashimi process of fishes (tuna, flatfish, and sardine) in the fishery industry. b) NMR dataset of the fermentation process of milk in the livestock industry. c) NMR dataset of the boiling process of carrots in agriculture.

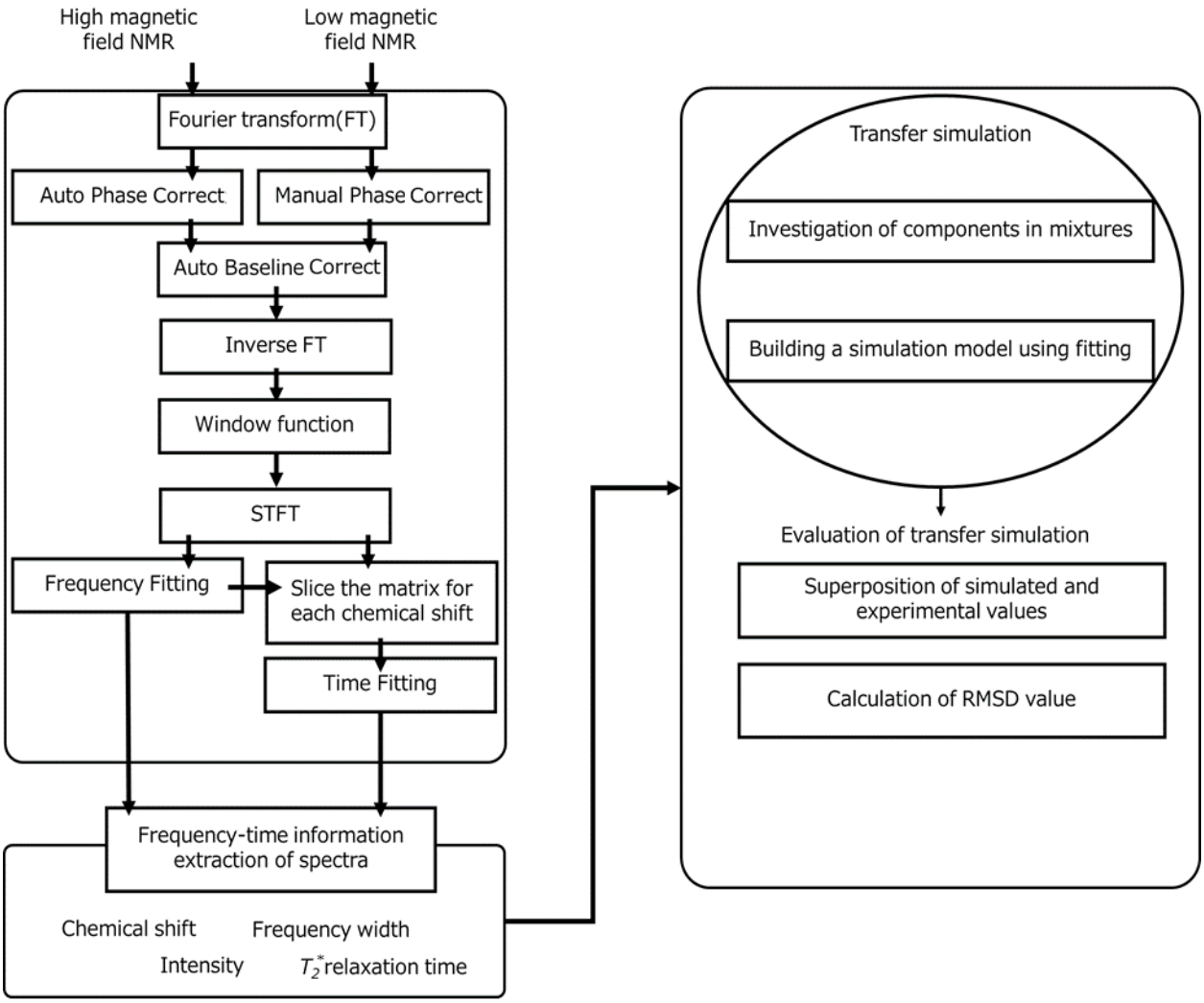

Figure S3. Diagram of simulation based on time–frequency analysis and line broadening. NMR spectra are transferred from high-field to low-field NMR.

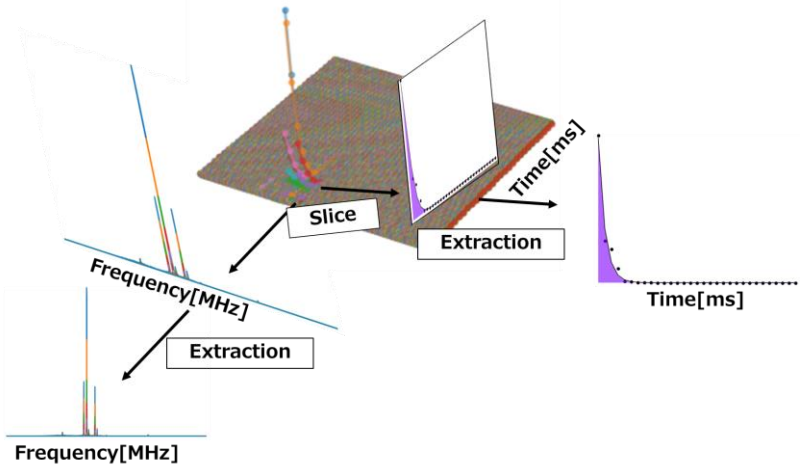

Figure S4. Flow of frequency and time information extraction by STFT.

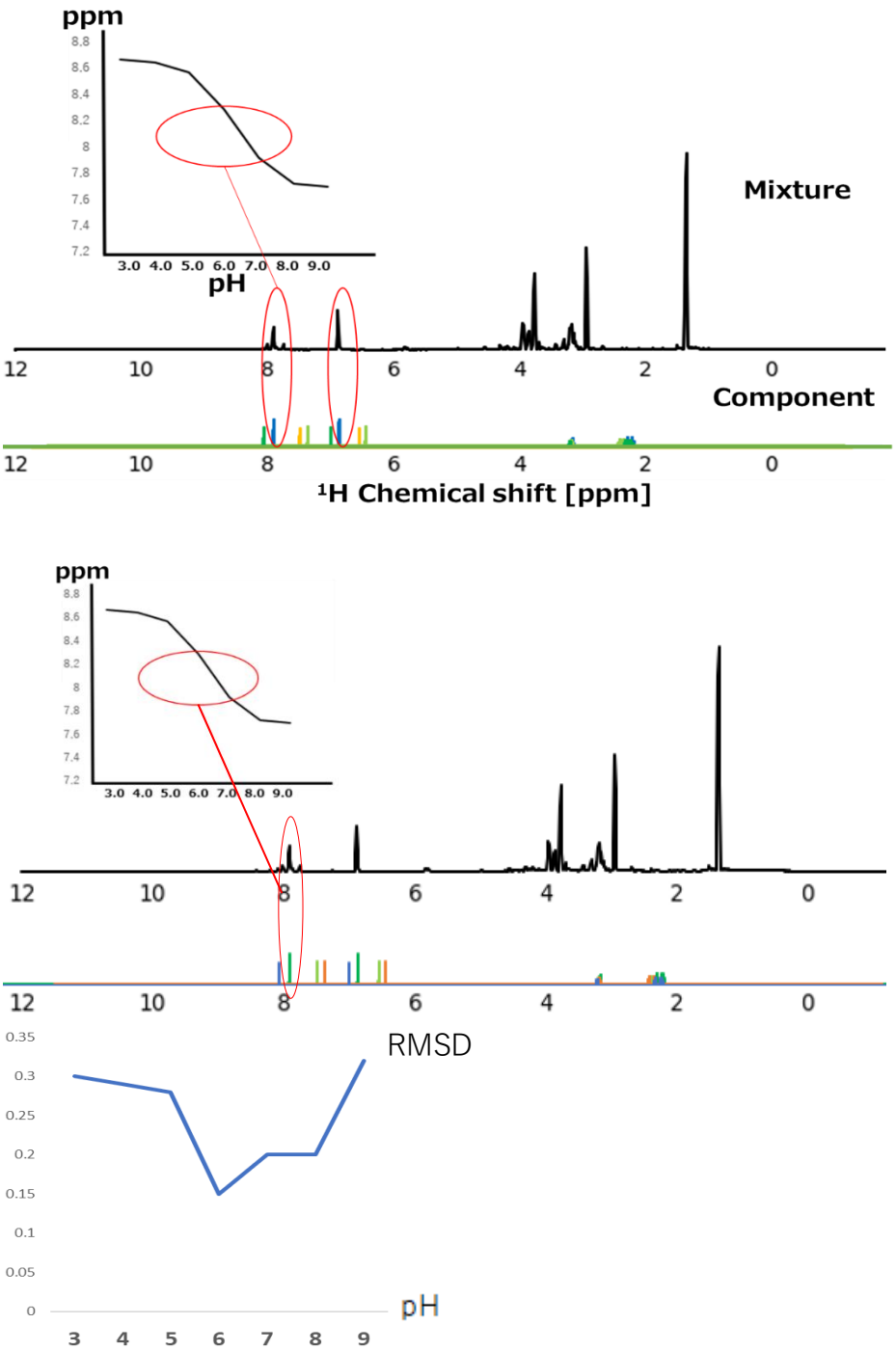

Figure S5. Analysis of NMR data at different pH values. **(a)**: Food mixture samples of 60-MHz ^{1}H-NMR spectra from tuna extract (black). The chemical shift changes upon different pH of Histidine signals (orange: pH = 5, yellow green: pH = 6, green: pH = 7, blue: pH = 8). (b) Changes in root-mean-square deviation values of matching between food mixture versus standard (Histidine) upon the different pH values.

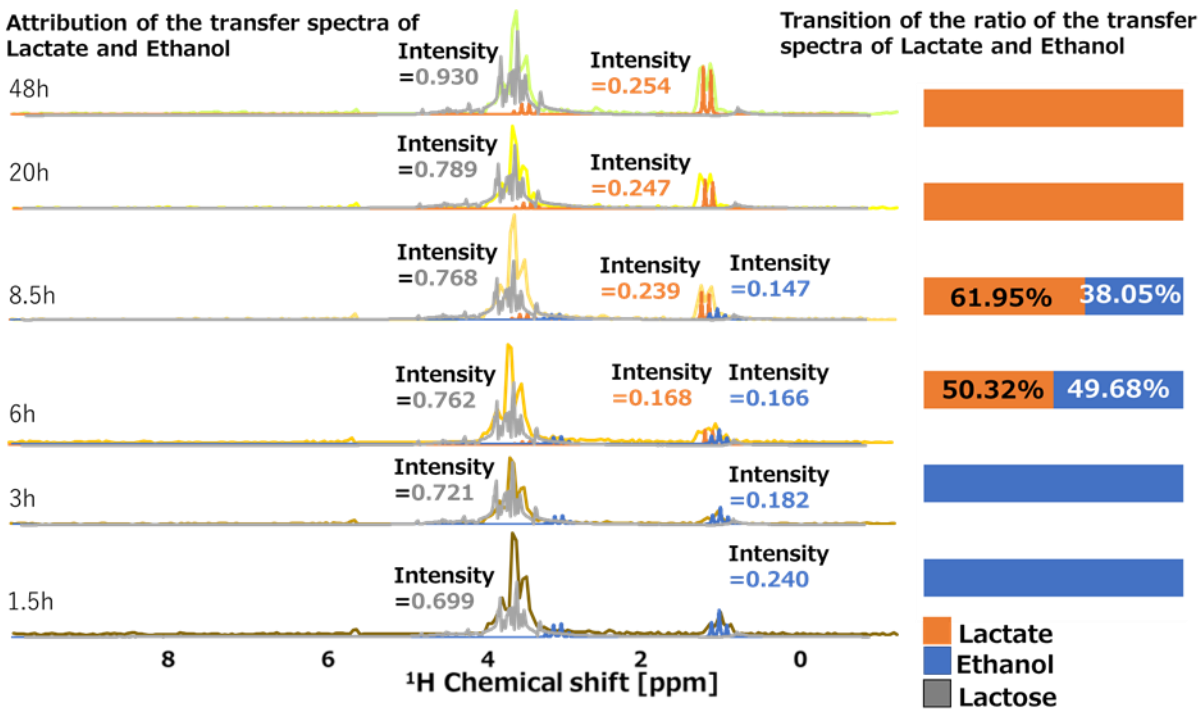

Figure S6. Transferred 60-MHz NMR spectra of milk fermentation process by the addition of yogurt samples within two days. Orange, blue, and gray represent the components of spectra by different component ratios of lactate, ethanol, and lactose, respectively.

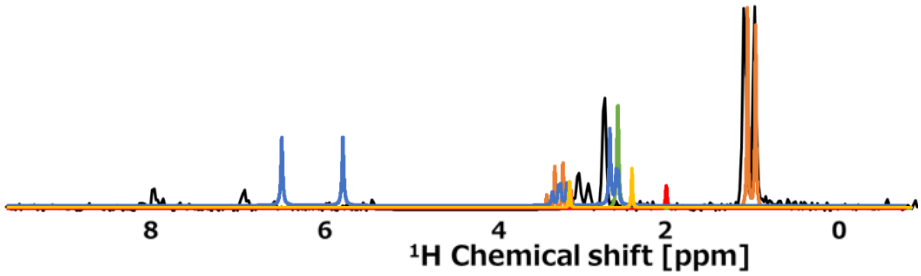

Figure S7. Generation of food mixture spectrum (60 MHz) using the Guided Ideographic Spin System Model Optimization (GISSMO) program. Red, green, orange, blue, and yellow represent the components of spectra by different component ratios of choline, trimethylamine *N*-oxide, lactate, histidine, and creatine, respectively. In this case, we did not perform pH adjustment indicated in Fig. S5.

Table S1. Peak annotation results of ^{1}H nuclear magnetic resonance (H NMR) spectra of the boiling process of carrot in agriculture.

Carrot's peaks	
ppm	**Annotation**
1.35	Lactate
1.49	Alanine
1.97	Quinate
2.11	Quinate
2.45	Malate
2.64	Malate
2.83	Aspartic acid
2.87	Asparagine
2.92	Asparagine
2.97	4-aminobutanoic acid
3.02	4-aminobutanoic acid
3.21	Glucose
3.35	Glucose
3.4	Glucose, Sucrose
3.54	Sucrose
3.64	Sucrose
3.78	Sucrose
3.83	Sucrose
3.92	Glucose, Sucrose
3.97	Fructose
4.06	Sucrose, Fructose
5.26	Glucose

Table S2. Peak annotation results of ^{1}H NMR spectra of the fermentation process of milk in the livestock industry.

Yogurt's peaks	
ppm	**Annotation**
1.35	Lactate
2.69	Citrate
2.83	Citrate
3.21	Carnithine
3.31	Lactose
3.54	Lactose
3.59	Lactose
3.64	Lactose
3.73	Lactose
3.83	Lactose
3.87	Lactose
3.93	Lactose
4.12	Lactose
5.26	Lactose

Table S3. Peak annotation results of ^{1}H NMR spectra sashimi process of fishes (tuna, sardine, and flatfish) in fishery.

Tuna's peaks

ppm	Annotation
0.97	Valine
1.35	Lactate
3.02	Creatine
3.21	Histidine
3.3	Histidine
3.87	Creatine
4.02	Histidine
5.26	Glucose
7.26	Histidine
8.35	Histidine

Sardine's peaks

ppm	Annotation
1.35	Lactate
3.02	Creatine
3.25	Trimethylamine *N*-oxide, Betaine
3.45	Taurine
3.92	Creatine
4.06	Lactate, Histidine
5.26	Glucose
8.16	Histidine

Flatfish's peaks

ppm	Annotation
1.35	Lactate
3.02	c
3.26	Trimethylamine *N*-oxide, Betaine
3.93	Creatine
5.27	Glucose
5.36	Maltose

Table S4. Fitting results for the NMR spectrum of the tested standard compounds. (a) Low-field (60 MHz) and (b) high-field (700 MHz) NMR.

(a) Low-field NMR (60 MHz)				
Standard compound name	**Chemical shift [ppm]**	**Intensity**	**Full width at half maximum [Hz]**	**T_2^* relaxation time [s]**
Choline	3.28	1023	10.01	0.0151
	4.59	1005	6.501	0.0035
Trimethylamine *N*-oxide	1.13	3526	10.11	0.0191
	4.55	2578	7.831	0.0115
	4.69	9484	8.321	0.0122
	5.53	3443	5.911	0.0007
	7.31	2551	10.12	0.0199
Lactate	1.17	7410	11.21	0.0213
	2.67	793	14.81	0.0243
	2.77	793	14.22	0.0231
Creatine	1.17	9787	6.011	0.0014
	4.36	5669	7.019	0.0080
	5.06	1411	8.201	0.0129
	5.30	5012	7.723	0.0085
	5.44	5117	1.009	0.0148
	5.53	2107	12.01	0.0209
	7.31	3714	12.13	0.0216
Histidine	2.45	6599	10.01	0.0180
	2.59	19328	6.014	0.0015
	3.30	5731	12.12	0.0223
	3.44	3999	12.91	0.0251
	6.72	8490	4.012	0.0007
	7.47	7412	9.031	0.0149
(b) High-field NMR (700 MHz)				
Standard compound name	**Chemical shift [ppm]**	**Intensity**	**Full width at half maximum [Hz]**	**T_2^* relaxation time [s]**
Choline	2.95	1144	4.716	0.2886
	4.50	5791	4.830	0.3869
Trimethylamine *N*-oxide	3.01	2515	4.900	0.3394
	4.51	9335	5.000	0.5611
Lactate	0.23	349197	4.796	0.2748
	0.31	2552	4.984	0.2505
	1.54	2634	4.918	0.3644
Creatine	0.23	438916	4.911	0.2795
	0.84	19849	4.871	0.3563
	0.87	21566	4.746	0.3448
	3.13	22811	4.678	0.3094
	3.15	25044	4.911	0.3170
	3.25	1932132	4.337	0.2768
	4.14	782603	4.731	0.2642
Histidine	0.04	6267	4.881	0.1627
	0.06	4717	4.811	0.3512
	0.13	460770	5.000	0.4311
	0.21	4770	4.911	0.4255
	0.71	25198	4.792	0.3736
	0.75	21911	4.812	0.4405
	0.83	130577	5.000	0.3013
	0.99	100294	4.811	0.3020
	1.05	60984	4.712	0.2973
	1.13	132421	4.982	0.2881
	1.27	7783	4.787	0.2541
	1.29	13628	4.992	0.1828
	1.60	9035	4.992	0.3146
	2.91	25257	4.812	0.4971
	2.94	24068	5.000	0.4839
	3.52	11466	5.000	0.1779
	6.70	11006	5.000	0.0628
	7.45	3967	4.780	0.0665

Table S5. Fitting results in the NMR spectrum of the tuna sample. (a) low-field NMR (60 MHz), (b) High-field NMR (700 MHz).

(a) Low-field NMR (60 MHz)

Sample name	Chemical shift [ppm]	Intensity	Full width at half maximum [Hz]	T_2^* relaxation time [s]
Tuna	1.484	8015	3.443	0.0050
	1.625	7931	3.313	0.0049
	3.383	4440	3.443	0.0050
	3.594	1104	1.399	0.0049
	3.711	1499	3.443	0.0048
	4.344	3511	3.340	0.0047
	4.508	1803	4.000	0.0048
	7.653	844	0.3443	0.0042
	8.167	893	4.344	0.0049

(b) High-field NMR (700 MHz)

Sample name	Chemical shift [ppm]	Intensity	Full width at half maximum [Hz]	T_2^* relaxation time [s]
Tuna	1.175	18922	5.134	0.128
	3.621	7291	4.167	0.109
	3.709	1821	4.238	0.112
	3.812	2058	4.288	0.182
	6.785	3740	5.000	0.080
	7.825	2121	6.000	0.039

Table S6 Fitting results for the NMR spectrum of the sardine sample. (a) Low-field (60 MHz) and (b) high-field (700 MHz) NMR.

(a) Low-field NMR (60 MHz)

Sample name	Chemical shift [ppm]	Intensity	Full width at half maximum [Hz]	T_2^* relaxation time [s]
Sardine	2.484	12784	5.134	0.002
	2.625	14272	4.167	0.002
	4.406	15528	4.238	0.002
	4.547	1409	4.288	0.013
	4.781	8340	5.000	0.002
	5.344	1851	6.000	0.027
	7.313	11408	9.000	0.019
	7.906	1264	9.000	0.019

(b) High-field NMR (700 MHz)

Sample name	Chemical shift [ppm]	Intensity	Full width at half maximum [MHz]	T_2^* relaxation time [s]
Sardine	2.400	14692	12.60	0.305
	2.619	10153	24.26	0.298
	3.193	12101	8.721	0.287
	6.092	1504	6.224	0.061
	6.967	463	5.825	0.196
	7.008	457	5.840	0.100
	7.076	518	6.010	0.291
	7.240	473	6.044	0.247

Table S7. Fitting results for the NMR spectrum of the flatfish sample. (a) Low-field (60 MHz) and (b) high-field (700 MHz) NMR.

(a) Low-field NMR (60 MHz)

Sample name	Chemical shift [ppm]	Intensity	Full width at half maximum [MHz]	T_2^* relaxation time [s]
Flatfish	1.78	459	5.840.	0.015
	2.48	1555	12.60	0.016
	2.63	1516	12.60	0.001
	4.41	3821	24.26	0.019
	4.64	10316	12.60	0.020
	5.02	464	24.26	0.022
	5.20	451	5.134	0.016
	5.34	2735	9.000	0.018
	7.31	699	9.000	0.024

(b) High-field NMR (700 MHz)

Sample name	Chemical shift [ppm]	Intensity	Full width at half maximum [MHz]	T_2^* relaxation time [s]
Flatfish	0.09	1517	5.003	0.3338
	2.20	4300	1.259	0.0754
	2.26	2440	2.619	0.1811
	3.42	4249	2.623	0.1855
	3.45	2608	2.435	0.1680
	3.56	3599	1.987	0.1280
	3.69	2371	2.018	0.1409
	3.88	2320	2.313	0.0421
	4.65	1195	2.187	0.1769