"Supporting Information"

Empirical Kernel Map Approach to Nonlinear Underdetermined Blind Separation of Sparse Nonnegative Dependent Sources: Pure Components Extraction from Nonlinear Mixtures Mass Spectra

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Taylor expansion of nonlinear blind source separation problem

The underdetermined nonlinear nonnegative blind source separation (uNNBSS) problem with dependent sources is described as:

$$\mathbf{x}_t = \mathbf{f}\left(\mathbf{s}_t\right) \qquad t = 1,...,T \tag{1}$$

where $\mathbf{x}_t \in \mathbb{R}_{0+}^{N\times 1}$ stands for nonnegative measurement vector comprised of intensities acquired at some of T mass-to-charge (m/z) channels, $\mathbf{s}_t \in \mathbb{R}_{0+}^{M\times 1}$ stands for unknown vector comprised of intensities of M nonnegative sources. $\mathbf{f}: \mathbb{R}_{0+}^{M} \to \mathbb{R}_{0+}^{N}$ is an unknown multivariate mapping such that $\mathbf{f}\left(\mathbf{s}_t\right) = \left[f_1\left(\mathbf{s}_t\right) \ldots f_N\left(\mathbf{s}_t\right) \right]^T$ and $\left\{ f_n : \mathbb{R}_{0+}^{M} \to \mathbb{R}_{0+} \right\}_{n=1}^{N}$. We need to expand vector valued function with vector argument (1) into Taylor series around some reference point $\mathbf{s}_0$. Thereby, expansion goes up to arbitrary order K. Without loss of generality we assume $\mathbf{s}_0=\mathbf{0}_{M\times 1}$ and $\mathbf{f}(\mathbf{s}_0)=\mathbf{0}_{N\times 1}$. $\mathbf{0}_{M\times 1}$ and $\mathbf{0}_{N\times 1}$ stand for column vectors of dimensions M and N with all entries equal to zero. To simplify notation in derivations related to Taylor expansion of (1) we shall drop a column index t. For example, instead of $\mathbf{x}_t$ and $\mathbf{s}_t$ we shall respectively use $\mathbf{x}$ and $\mathbf{s}$. In the literature one mostly finds Taylor expansion based on first- (Jacobian) and second (Hessian) order derivatives of vector valued function with vector argument and first derivative of matrix function with matrix argument. It is argued in [S1] that very occasionally one might need third- and higher-order derivatives of vector- or matrix-valued functions with vector or matrix arguments. The main reason for that is notational complexity. It is argued in [S2] and [S3], in chapters 9 and 10, to use procedure based on differentials when calculating first and second

derivatives of discussed functions. For higher order terms no recommendation is given. In what follows we present an approach to derivation of the Taylor expansion of vector valued function with vector argument up to arbitrary order K by using tensorial notation, [S4]. That is legitimate given the fact that k^{th} term, k=1,...,K, in Taylor expansion of vector valued function with vector argument is a tensor of order k+1. To this end, higher-order arrays (tensors) will be denoted with underlined uppercase bold letters. For example $\underline{\mathbf{X}} \in \mathbb{R}_{0+}^{I_1 \times I_2 \times I_3}$ refers to a third order nonnegative tensor with dimensions I_1, I_2 and I_3. Uppercase bold letters denote matrices, lowercase bold letters denote vectors and italic lowercase letters denote scalars. Having in mind (1) we can write the kth order derivative as a tensor of order 1+k:[1]

$$\underline{\mathbf{G}}^k \in \mathbb{R}_{0+}^{N \times \overbrace{M \times \ldots \times M}^{k \text{ times}}} \tag{2}$$

We now introduce mode-r product of an Rth order tensor $\underline{\mathbf{T}} \in \mathbb{R}^{I_1 \times I_2 \times \ldots \times I_R}$ and matrix $\mathbf{W} \in \mathbb{R}^{J_1 \times J_2}$ that is defined when number of columns of matrix is equal to the dimension of the tensor in mode r, that is J_2=I_r. It yields a new tensor $\underline{\mathbf{Y}} = \underline{\mathbf{T}} \times_r \mathbf{W}$, such that $\underline{\mathbf{Y}} \in \mathbb{R}^{I_1 \times \ldots \times I_{r-1} \times J_2 \times I_{r+1} \times \ldots \times I_R}$, [S5].[2] We can now express contribution of the kth order term in Taylor expansion as:

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[1] Element of the derivative tensor indexed by (n, m_1, ... , m_k), where n=1, ... , N, m_1=1,..., M, ... , m_k=1,..., M is given as: $\left[\underline{\mathbf{G}}^k\right]_{nm_1 \ldots m_k} = \dfrac{\partial^k f_n(\mathbf{s})}{\partial s_{m_1} \ldots \partial s_{m_k}}$.

[2] For example, mode-*2* product of a 3-way tensor $\underline{\mathbf{T}} \in \mathbb{R}^{I_1 \times I_2 \times I_3}$ and a matrix $\mathbf{W} \in \mathbb{R}^{D \times I_2}$ is a 3-way tensor $\underline{\mathbf{Y}} = \underline{\mathbf{T}} \times_2 \mathbf{W} \in \mathbb{R}^{I_1 \times D \times I_3}$, calculated element-wise as: $y_{i_1,d,i_3} = \sum_{i_2=1}^{I_2} t_{i_1,i_2,i_3} \cdot w_{d,i_2}$.

$$\mathbf{x}^k = \frac{1}{k!}\underline{\mathbf{G}}^k \times_2 \mathbf{s}^T \ldots \times_{k+1} \mathbf{s}^T \tag{3}$$

where T denotes transpose operation and (4) is known as Tucker tensor model [S5, S6]. Thereby, $\frac{1}{k!}\underline{\mathbf{G}}^k$ stands for a core tensor and $\mathbf{s}^T$ for factors. Since $\mathbf{s}^T$ is $1\times M$ row vector mode-*2* to mode-(*k+1*) multiplications of derivative tensor $\underline{\mathbf{G}}^k$ with row vectors $\mathbf{s}^T$ contracts higher order dimensions yielding as final result an $N\times 1$ column vector. It is also possible to use mode-*1* unfolding of $\underline{\mathbf{G}}^k \times_2 \mathbf{s}^T \ldots \times_{k+1} \mathbf{s}^T$ that yields:[3]

$$\mathbf{x}^k = \frac{1}{k!}\mathbf{G}^k_{(1)}\left[\mathbf{s}\underbrace{\otimes \ldots \otimes}_{k-1\ times}\mathbf{s}\right] \tag{4}$$

where $\mathbf{G}^k_{(1)} \in \mathbb{R}_{0+}^{N\times M^{(k)}}$ denotes a matrix obtained by mode-*1* unfolding of tensor $\underline{\mathbf{G}}^k$, $\otimes$ denotes Kronecker product and $\mathbf{s}\underbrace{\otimes \ldots \otimes}_{k-1\ times}\mathbf{s}$ yields $M^{(k)}\times 1$ vector, whereas $M^{(k)} = \binom{M+k-1}{k}$.[4] Hence we can formally write a *K*th order Taylor expansion of vector valued function with vector argument as:

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[3] The mode-*r* unfolding of tensor $\underline{\mathbf{T}}$ produces matrix $\mathbf{T}_{(r)} \in \mathbb{R}^{I_r \times \prod_{k,k\neq r} I_k}$ which consists of mode-*r* vectors stacked in the matrix as columns. There are various possibilities for ordering of mode-*r* vectors into columns of $\mathbf{T}_{(r)}$, but particular ordering is not important, as long it is consistent through all computations, [S5].

[4] Kronecker product $\mathbf{s}\underbrace{\otimes \ldots \otimes}_{k-1\ times}\mathbf{s}$ yields $M^k\times 1$ vector. However, monomials such as $s_i s_j$ and $s_j s_i$ stand for the same monomial. That is why M^k has to be substituted by $M^{(k)} = \binom{M+k-1}{k}$.

$$\mathbf{x} = \sum_{k=1}^{K} \frac{1}{k!} \underline{\mathbf{G}}^{k} \times_2 \mathbf{s}^T \ldots \times_{k+1} \mathbf{s}^T = \sum_{k=1}^{K} \frac{1}{k!} \mathbf{G}_{(1)}^{k} \left[\mathbf{s} \underbrace{\otimes \ldots \otimes}_{k-1 \ times} \mathbf{s} \right] \tag{5}$$

Elements of the column vector $\mathbf{s} \underbrace{\otimes \ldots \otimes}_{k-1 \ times} \mathbf{s}$ are monomials of the order k, that is:

$$\mathbf{s} \underbrace{\otimes \ldots \otimes}_{k-1 \ times} \mathbf{s} = \left[\left\{ s_{m_1}^{q_1} \ldots s_{m_p}^{q_p} \right\}_{m_1,\ldots,m_p=1;p=1}^{M;\ k} \right] \ s.t.\ q_1,\ldots,q_p \in \{0,1,\ldots,p\} \ \ and \ \sum_{i=1}^{p} q_i = k \tag{6}$$

Eq. (6) confirms that nonlinear mappings induce higher order (nonlinear) terms. In case of linear mapping, k=1, (6) becomes argument (source) vector $\mathbf{s}$. Thus, Taylor expansion (5) can be written as:

$$\mathbf{x} = \mathbf{G}\overline{\mathbf{s}} \tag{7}$$

where $\mathbf{G} \in \mathbb{R}_{0+}^{N \times \sum_{k=1}^{K} M^{(k)}}$ stands for block matrix written as:

$$\left[\mathbf{G}_{(1)}^{1} \left| \frac{1}{2} \mathbf{G}_{(1)}^{2} \right| \ldots \left| \frac{1}{K!} \mathbf{G}_{(1)}^{K} \right. \right], \tag{8}$$

where "|" stands for the separation between the blocks. $\overline{\mathbf{s}} \in \mathbb{R}_{0+}^{\sum_{k=1}^{K} M^{(k)} \times 1}$ is column vector of the form:

$$\overline{\mathbf{s}} = \left[\mathbf{s} \;\; \mathbf{s} \otimes \mathbf{s} \;\; \ldots \;\; \mathbf{s} \underbrace{\otimes \ldots \otimes}_{k-1 \;\; times} \mathbf{s} \right]^T \tag{9}$$

Equation (7) represents uLNBSS problem that is equivalent to uNNBSS problem (1). The equivalent uLNBSS problem (7) is comprised of new source vector $\overline{\mathbf{s}} \in \mathbb{R}_{0+}^{\sum_{k=1}^{K} M^{(k)} \times 1}$ that further is comprised of M original sources and $\sum_{k=2}^{K} M^{(k)}$ higher order monomials. These are new sources correlated with the original ones. Thus, original uNNBSS problem (1) characterized by N mixtures and $M>N$ dependent sources is equivalent to uLNBSS problem (7) characterized by the same number of N mixtures but with $\sum_{k=1}^{K} M^{(k)}$ dependent sources. Since underdetermined BSS problem with M dependent sources is hard to solve, [S7], underdetermined problem with $\sum_{k=1}^{K} M^{(k)}$ dependent sources is basically intractable. It means that without further constraints uNNBSS problem (1) is also intractable.[5]

References:

S1. Magnus J R. On the concept of matrix derivative. *J. Multivariate Analysis* 2010; **101**: 2200-2206.

S2. Magnus J R, Neudecker H. Matrix Differential Calculus with Applications to Simple, Hadamard, and Kronecker Products. *J. Mathematical Psychology* 1985; **29**: 474-492.

[5] It is worth noticing that nonlinear BSS method [S8] is also performed in EKM-induced RKHS , whereas sources in RKHS are constrained by mutual decorrelation and individual correlation (autocorrelation functions of individual sources differ from delta function). Given the fact that in addition to original sources higher order monomials are also present in RKHS we find mutual decorrelation constraint problematic even though source signals in [S8] were not constrained to be nonnegative.

S3. Magnus J R, Neudecker H. *Matrix Differential Calculus with Applications in Statistics and Economics*. Revised edition. John Wiley: Chichester, UK, 1999.

S4. Kiers H A L. Towards a standardized notation and terminology in multiway analysis. *J. Chemometrics* 2000; **14**: 105-122.

S5. Kolda T G, Bader B W. Tensor Decompositions and Applications. *SIAM Review* 2009; **51**: 455-500.

S6. Tucker L R. Some mathematical notes on three-mode factor analysis. *Psychometrika* 1966; **31**: 279-311.

S7. Kopriva I, Jerić I, Brkljačić L. Nonlinear mixture-wise expansion approach to underdetermined blind separation of nonnegative dependent sources. *J. Chemometrics* 2013; **27**: 189-197.

S8. Harmeling S, Ziehe A, Kawanabe M. Kernel-Based Nonlinear Blind Source Separation. *Neural Comput.* 2003; **15**: 1089-1124.

Table S-1. Numerical study for nonlinear blind source separation problem comprised of N=3 mixtures, M=8 sources, L=3 overlapped sources and T=1000 observations. Comparative performance analysis of NMU, NMF_L0, EKM-NMU, EKM-NMF_L0, PTs-EKM-NMU and PTs-EKM-NMF_L0 algorithms. Sources are generated according to mixed state sparse probabilistic model with probability of zero state was ρ_m=0.5 $\forall m$=1,...,25 and mean value of the exponential distribution of the amplitudes μ_m=1.5×10^{-3} $\forall m$=1,...,25. Four metrics used in comparative performance analysis were: number of assigned components with normalized correlation coefficient greater than or equal to 0.6, mean value of correlation coefficient over all correctly assigned components, minimal value of correlation coefficient and number of pure components assigned incorrectly (that occurs due to poor separation). Mean values and variance are reported and estimated over 10 Monte Carlo runs. The best result in each metric is in bold. The first three metrics are calculated only for correctly assigned components. That is why NMU and NMF_L0 seem to have comparable performance.

	NMU	**NMF_L0**	**EKM-NMU**	**EKM-NMF_L0**	**PTs_EKM-NMU**	**PTs-EKM-NMF_L0**
correlation G.E. 0.6	2.7±0.67	2.6±1.35	3.3±0.48	3.3±0.48	3.5±0.53	**3.8±0.63**
mean correlation	0.67±0.02	0.61±0.1	0.67±0.03	0.62±0.03	0.67±0.02	**0.68±0.03**
minimal correlation	**0.52±0.03**	0.42±0.06	0.5±0.03	0.42±0.03	0.5±.03	0.46±0.03
incorrect assignments	3.2±0.63	3.0±0.47	2.8±0.42	2.00±0.47	2.6±0.52	**1.7±0.67**

Table S-2. Normalized cross-correlation coefficients between 25 pure components ($\mathbf{s}_1$ to $\mathbf{s}_{25}$) generated in nonlinear chemical reaction of peptide synthesis. Thereby, pairs of pure components are identified with normalized correlation coefficient above 0.1.Their mass spectra are shown in Figure S-4.

	$\mathbf{s}_2$	$\mathbf{s}_6$	$\mathbf{s}_7$	$\mathbf{s}_9$	$\mathbf{s}_{10}$	$\mathbf{s}_{12}$
$\mathbf{s}_1$	0.9839	0.1416	0.1218	0.1796	0.1072	0.3343
	$\mathbf{s}_6$	$\mathbf{s}_7$	$\mathbf{s}_9$	$\mathbf{s}_{10}$	$\mathbf{s}_{12}$	
$\mathbf{s}_2$	0.1418	0.1268	0.1797	0.1075	0.3305	
	$\mathbf{s}_{16}$	$\mathbf{s}_{17}$	$\mathbf{s}_{18}$			
$\mathbf{s}_3$	0.3575	0.3103	0.1716			
	$\mathbf{s}_6$	$\mathbf{s}_{19}$	$\mathbf{s}_{21}$			
$\mathbf{s}_4$	0.3077	0.3947	0.4005			
	$\mathbf{s}_7$					
$\mathbf{s}_5$	0.7824					
	$\mathbf{s}_9$					
$\mathbf{s}_7$	0.3297					
	$\mathbf{s}_{13}$					
$\mathbf{s}_8$	0.1293					
	$\mathbf{s}_{12}$	$\mathbf{s}_{22}$				
$\mathbf{s}_{11}$	0.2666	0.1622				
	$\mathbf{s}_{17}$					
$\mathbf{s}_{14}$	0.1024					
	$\mathbf{s}_{22}$					
$\mathbf{s}_{15}$	0.1349					
	$\mathbf{s}_{17}$					
$\mathbf{s}_{16}$	0.9783					
	$\mathbf{s}_{18}$					
$\mathbf{s}_{17}$	0.1186					
	$\mathbf{s}_{21}$					
$\mathbf{s}_{19}$	0.9962					
	$\mathbf{s}_{24}$	$\mathbf{s}_{25}$				
$\mathbf{s}_{23}$	0.4409	0.4339				
	$\mathbf{s}_{25}$					

s_{24}	0.3008					

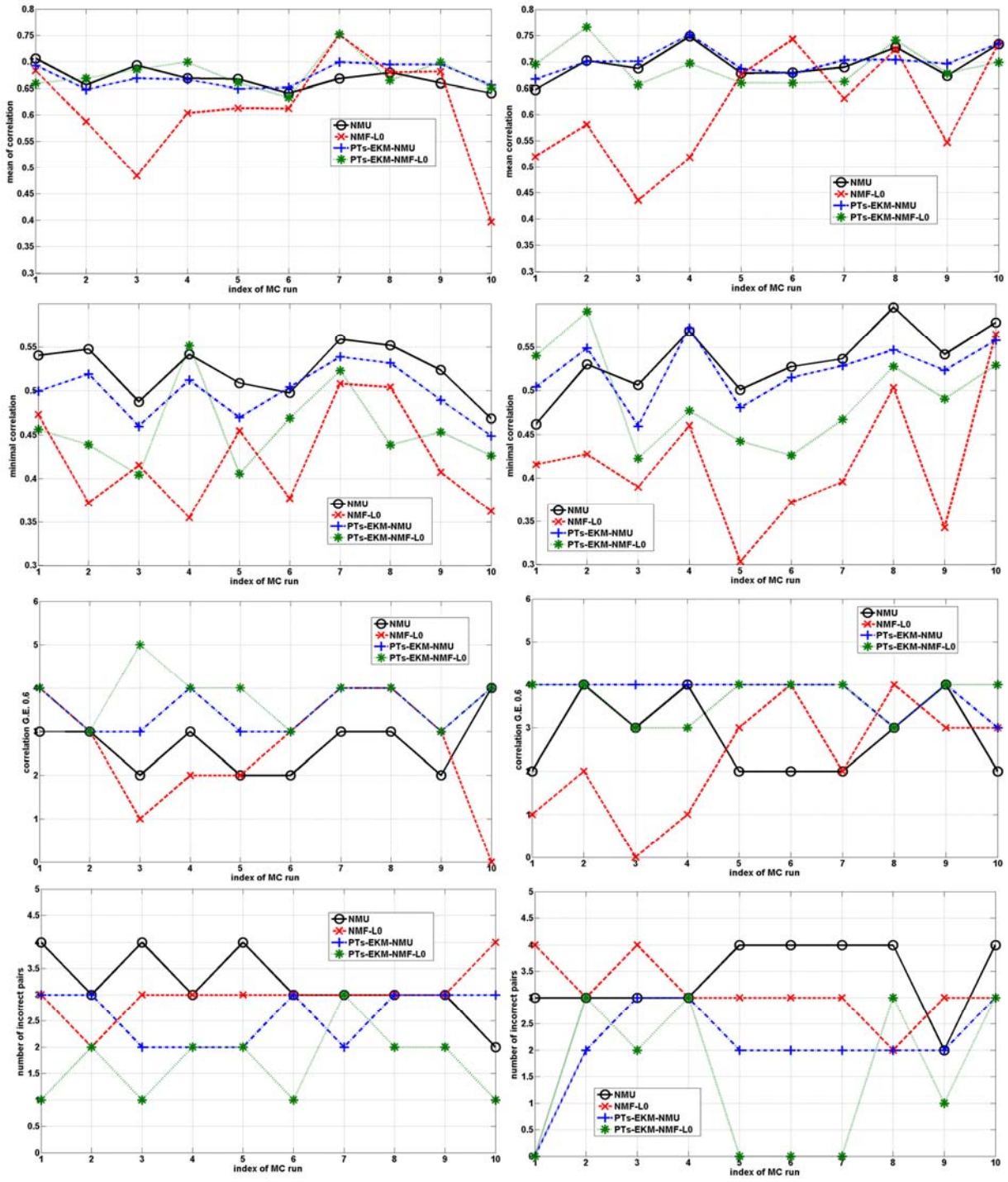
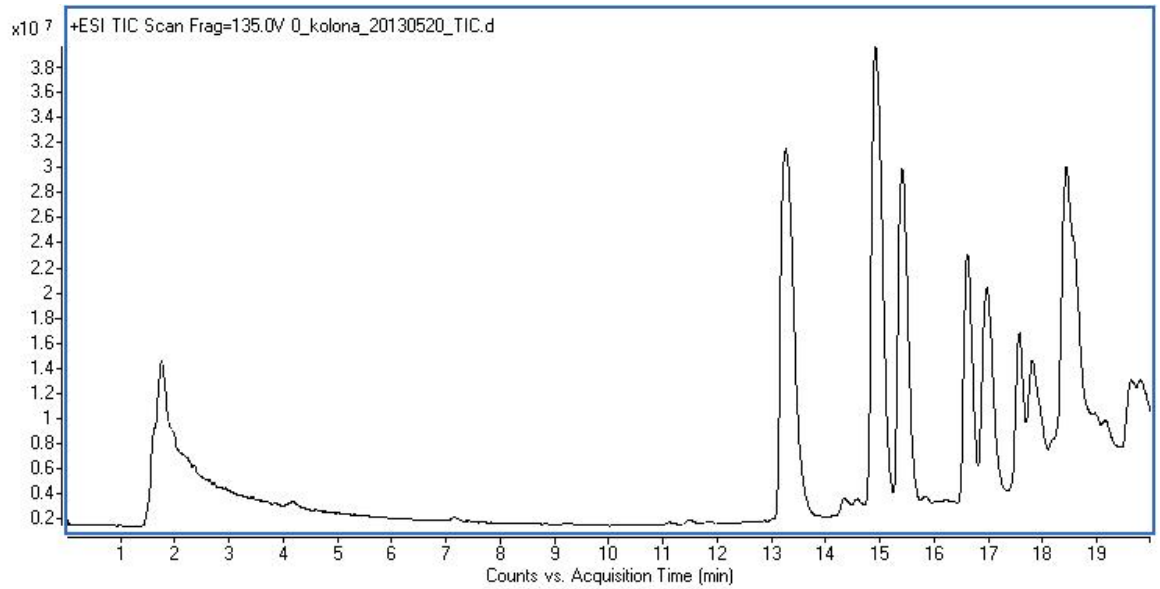


Figure S-1.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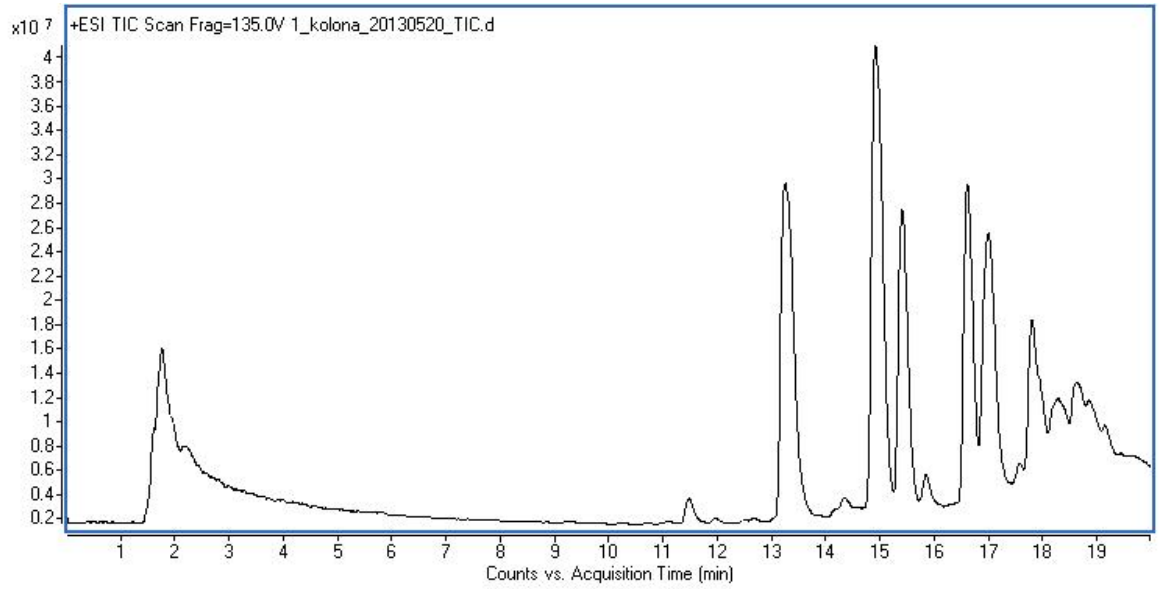
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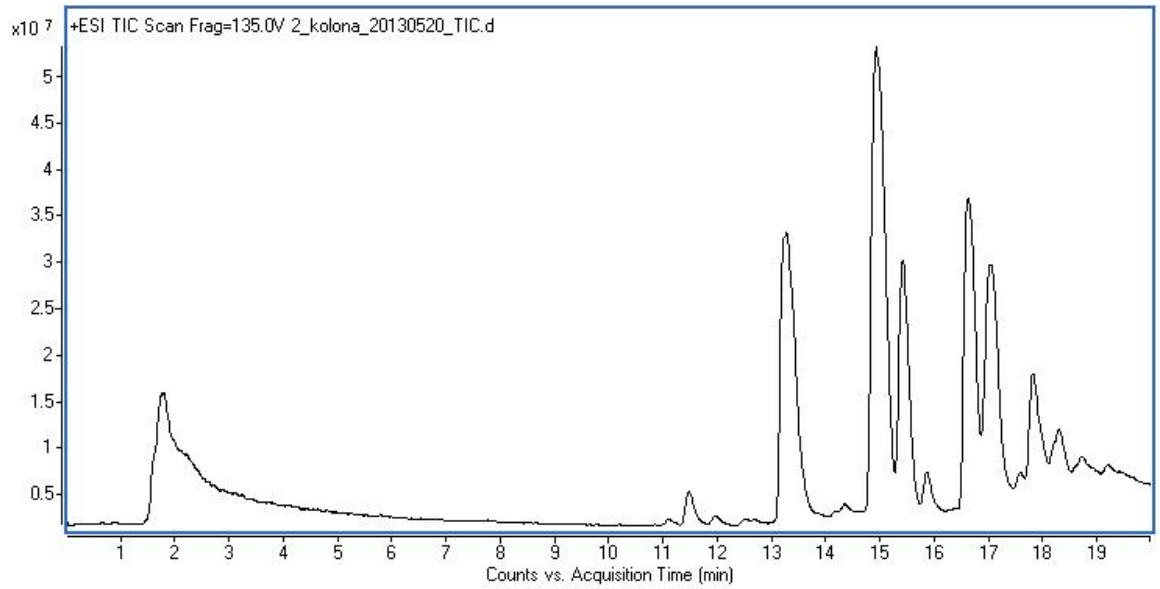
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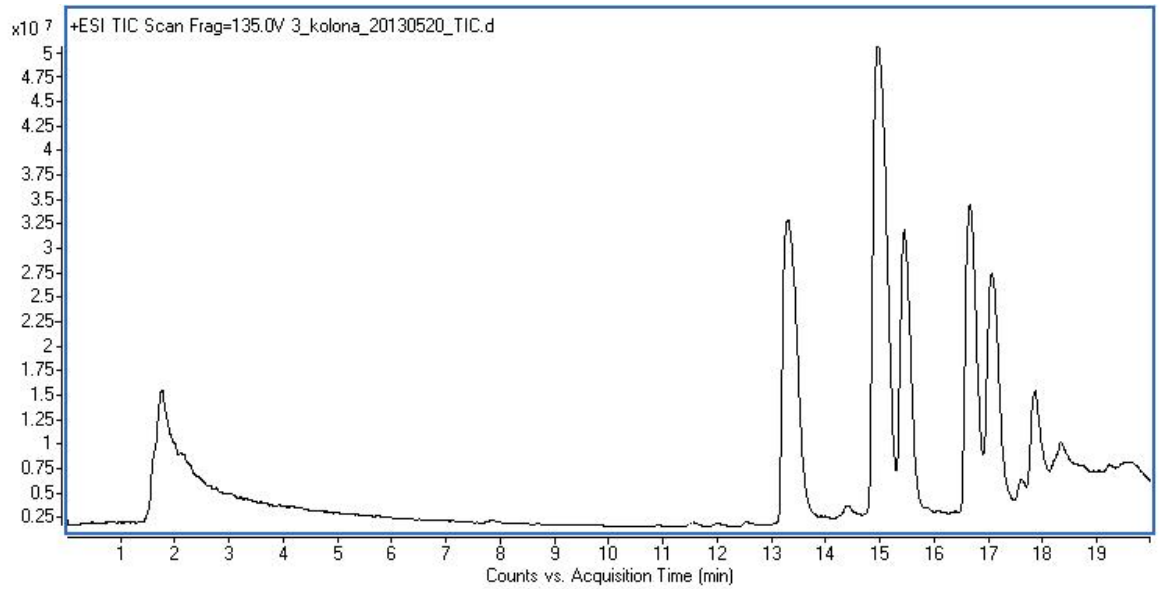
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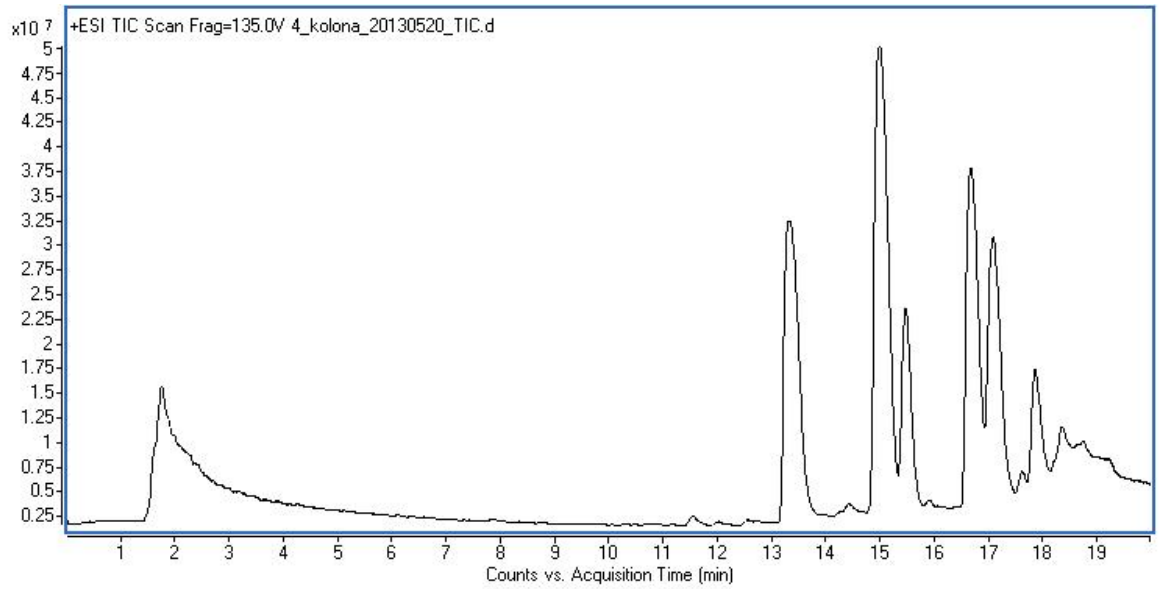
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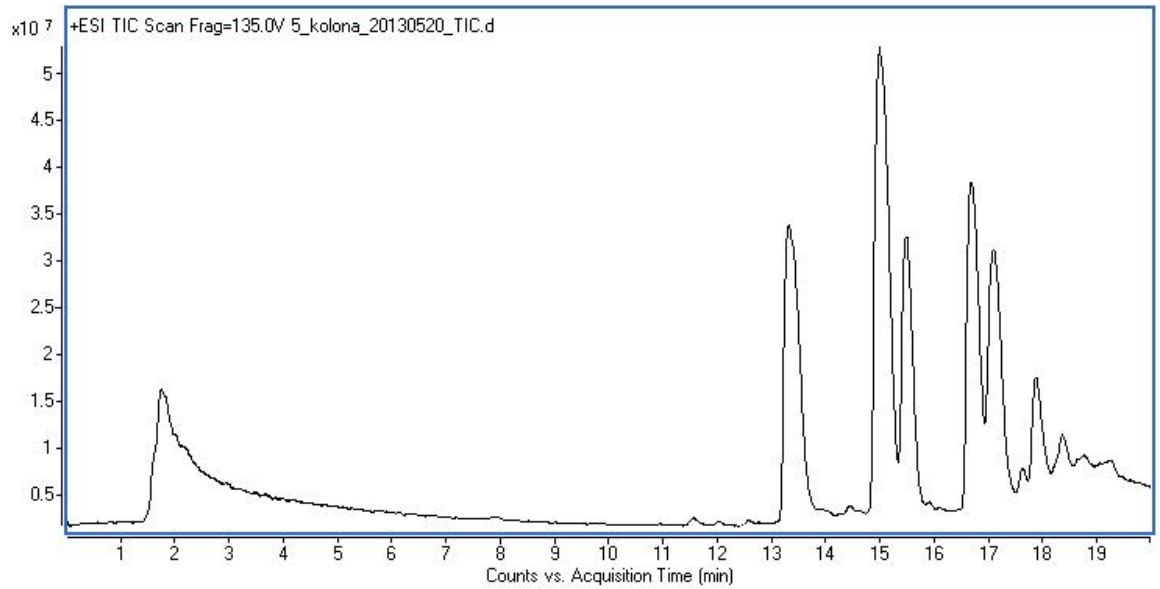
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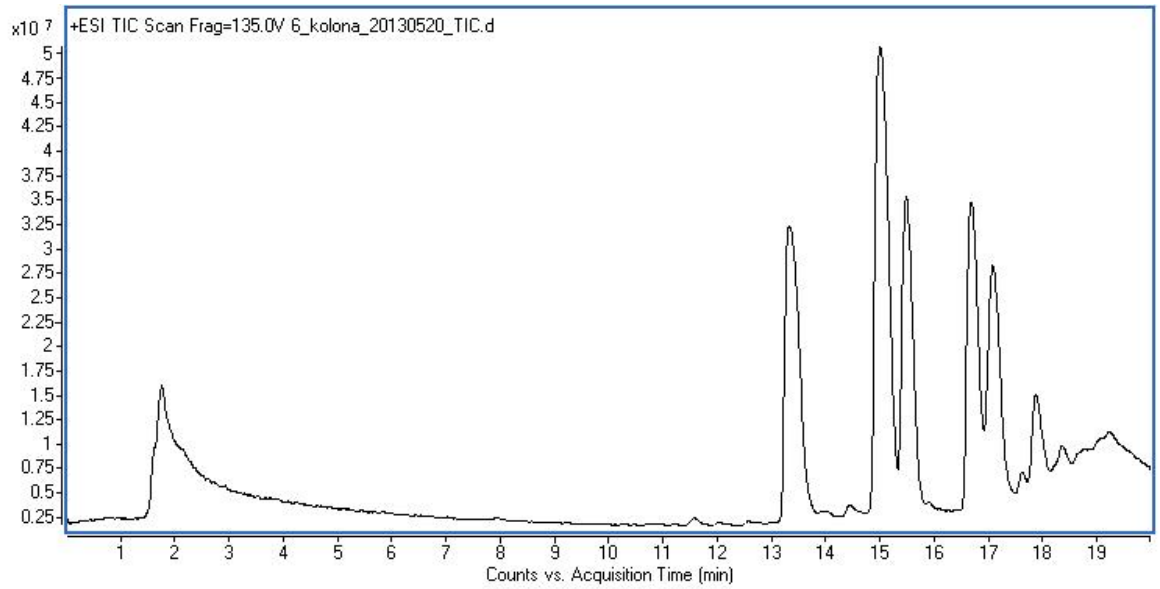
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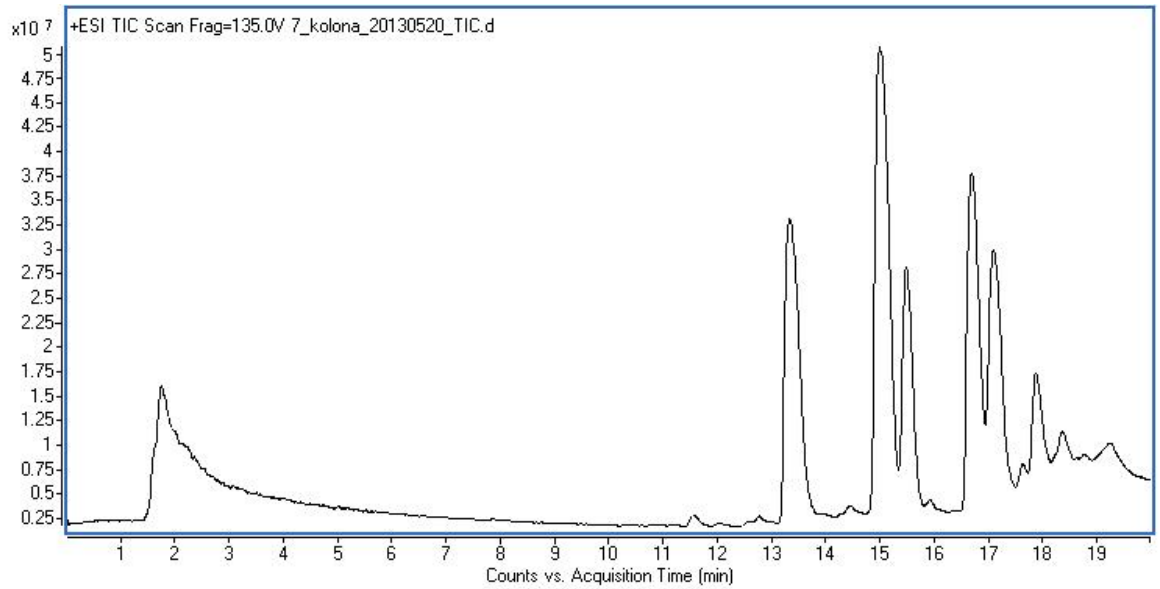
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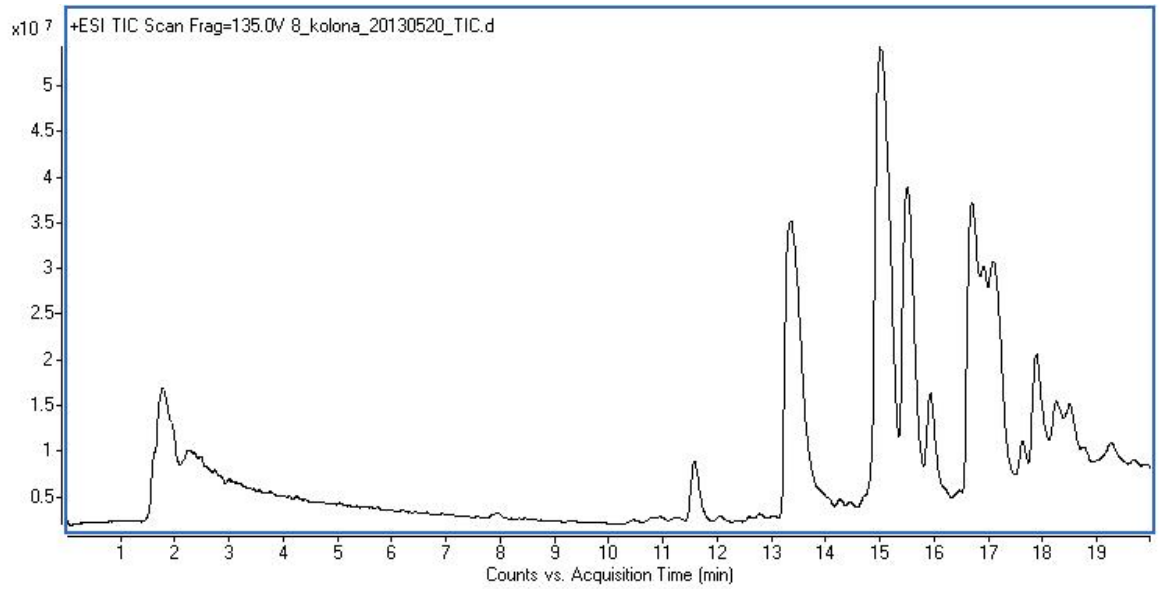
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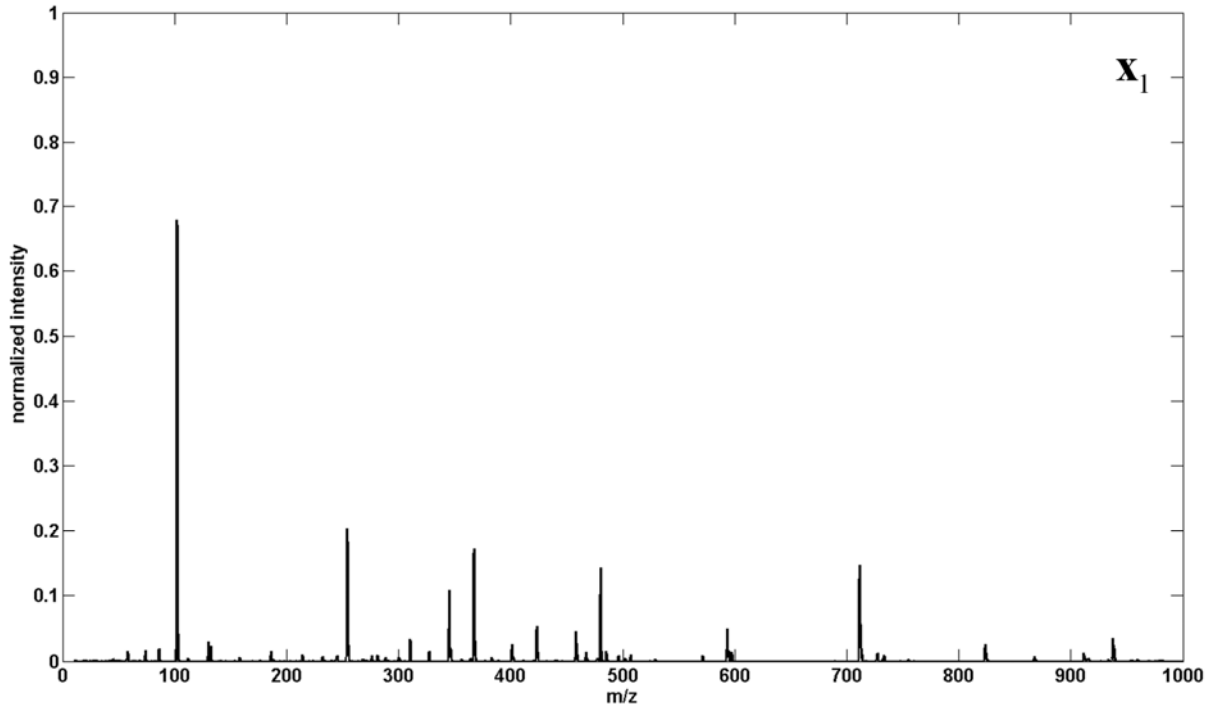
Figure S-2.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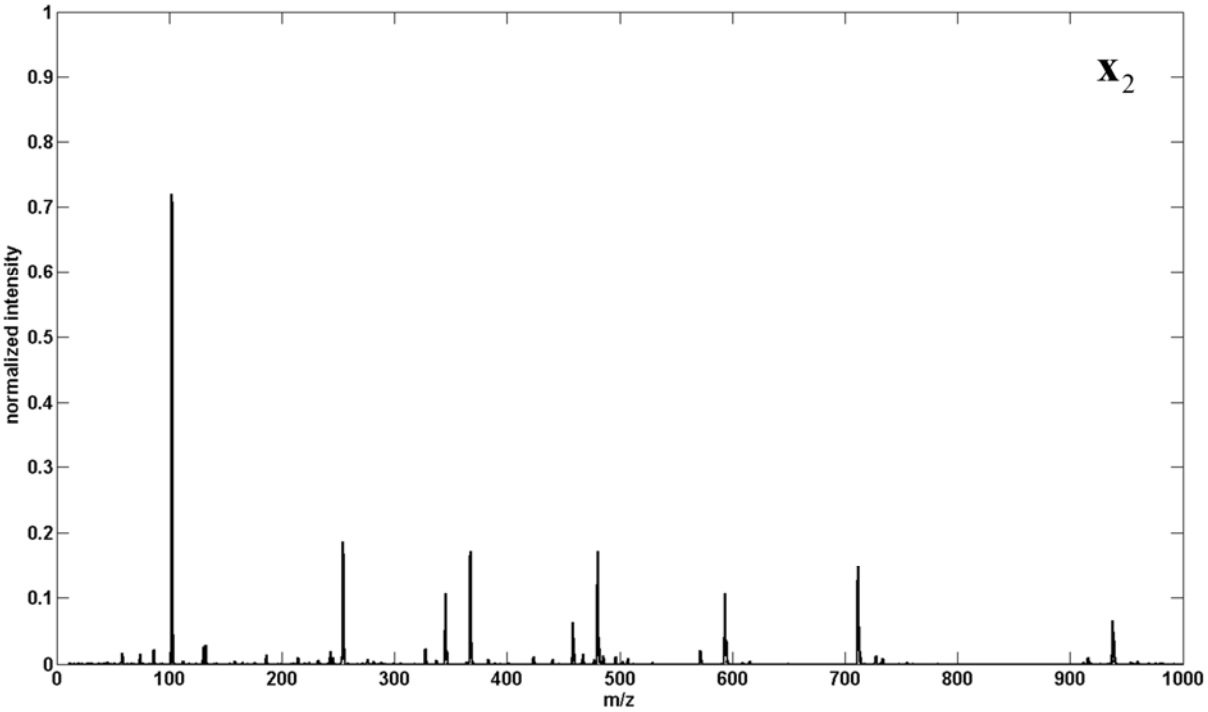
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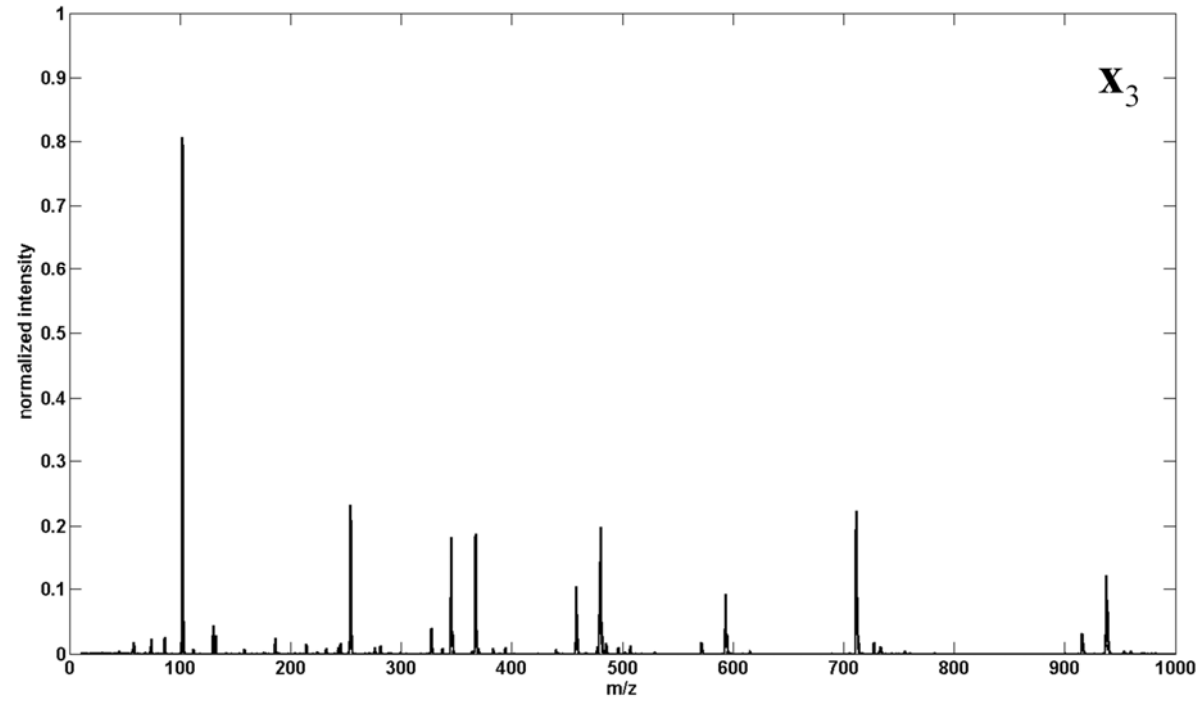
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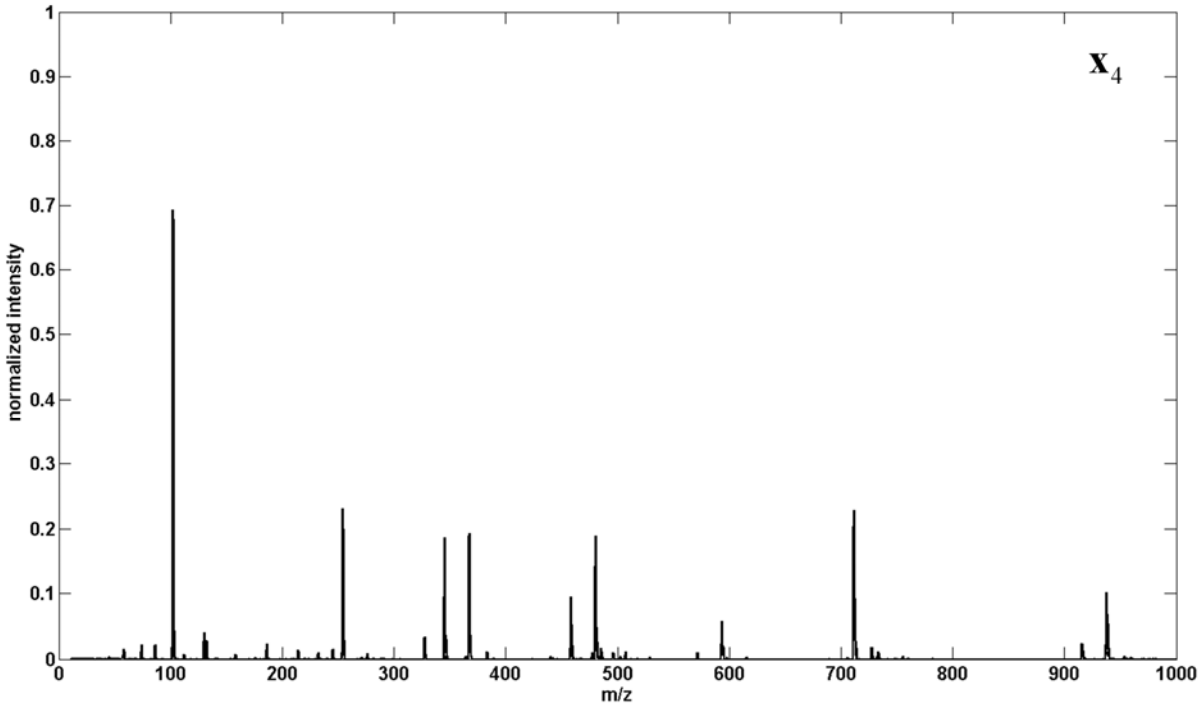
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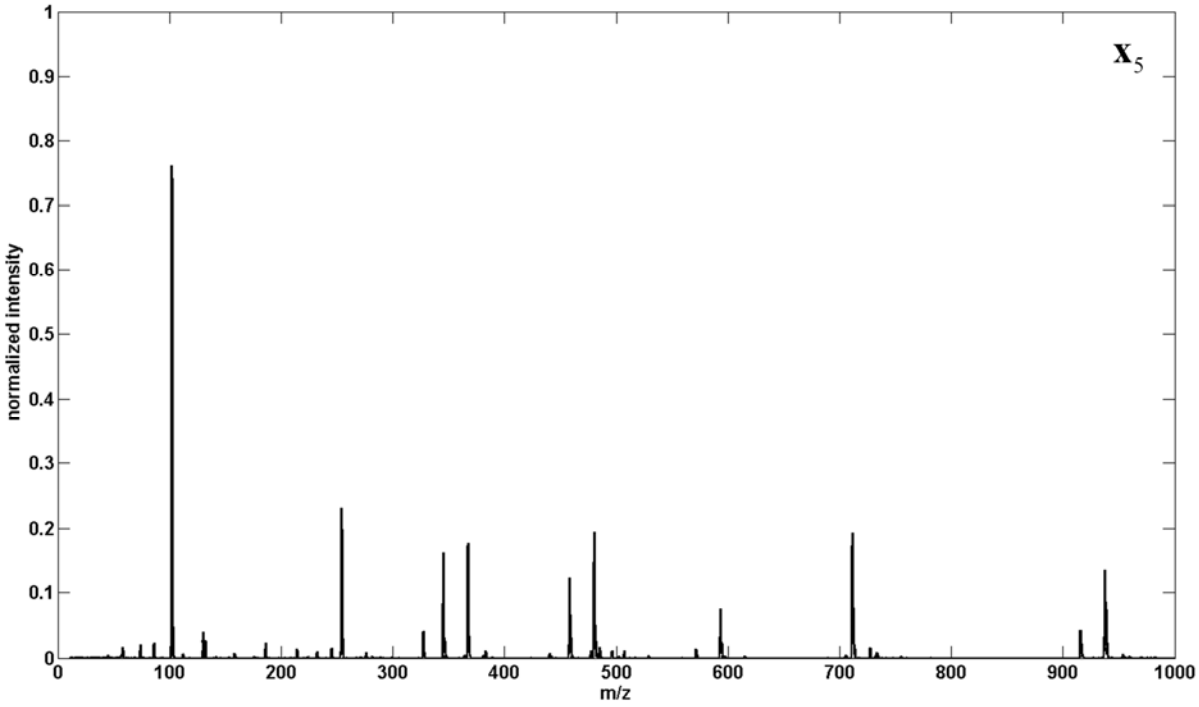
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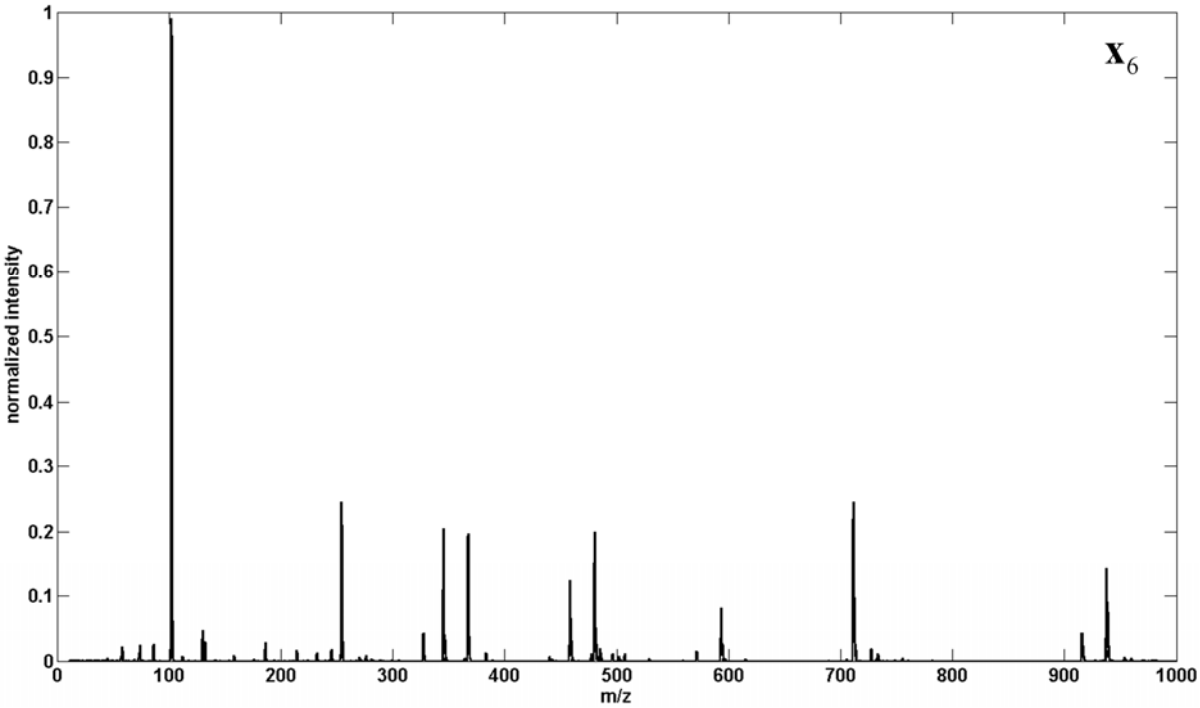
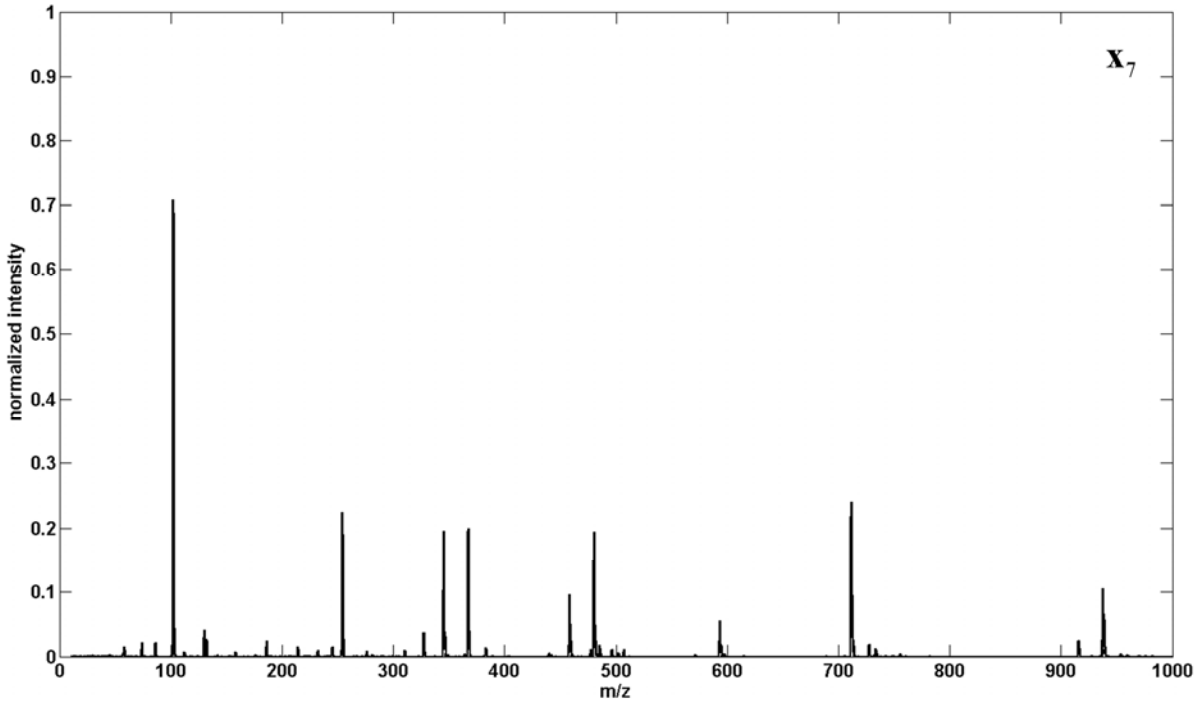
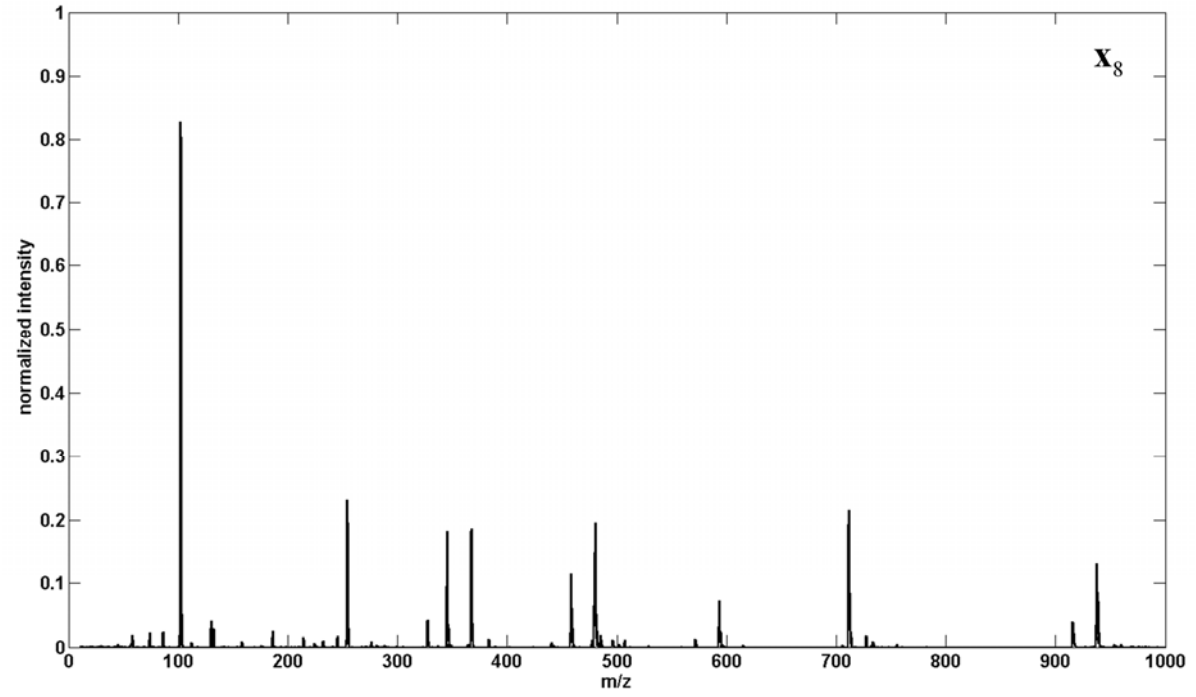
400
500
600
700
800
900
1000


$\mathbf{x}_6$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000


$\mathbf{x}_7$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000


$\mathbf{x}_8$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

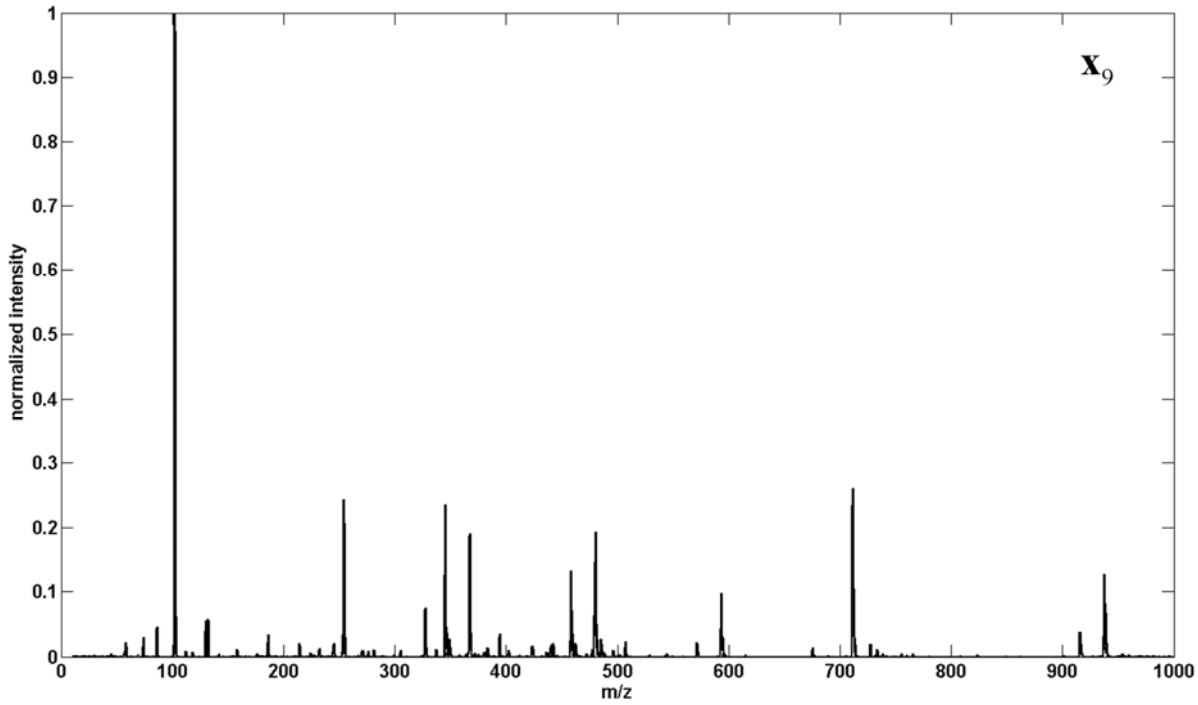
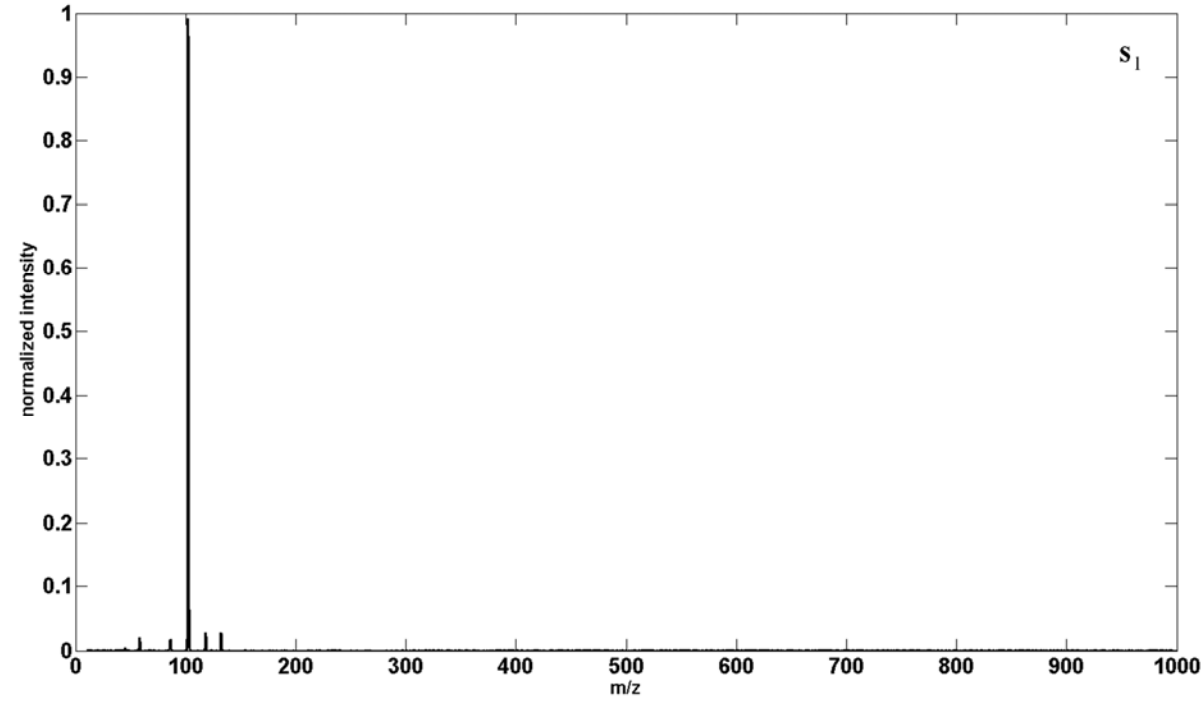
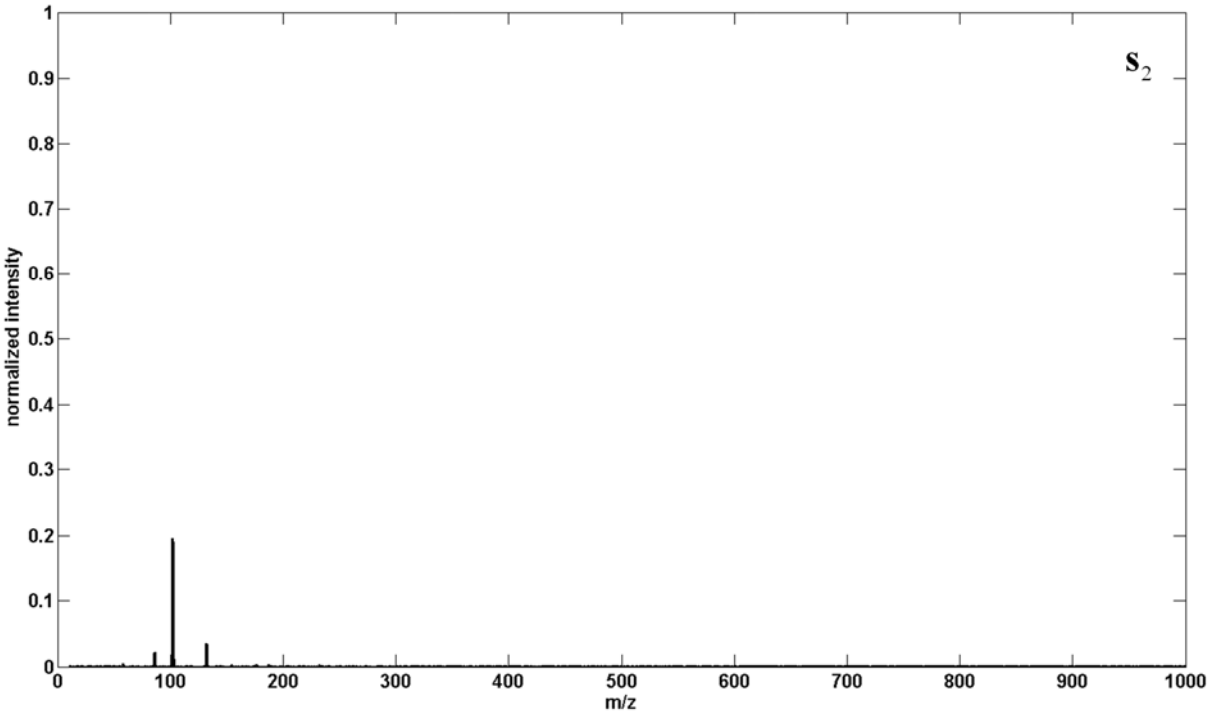
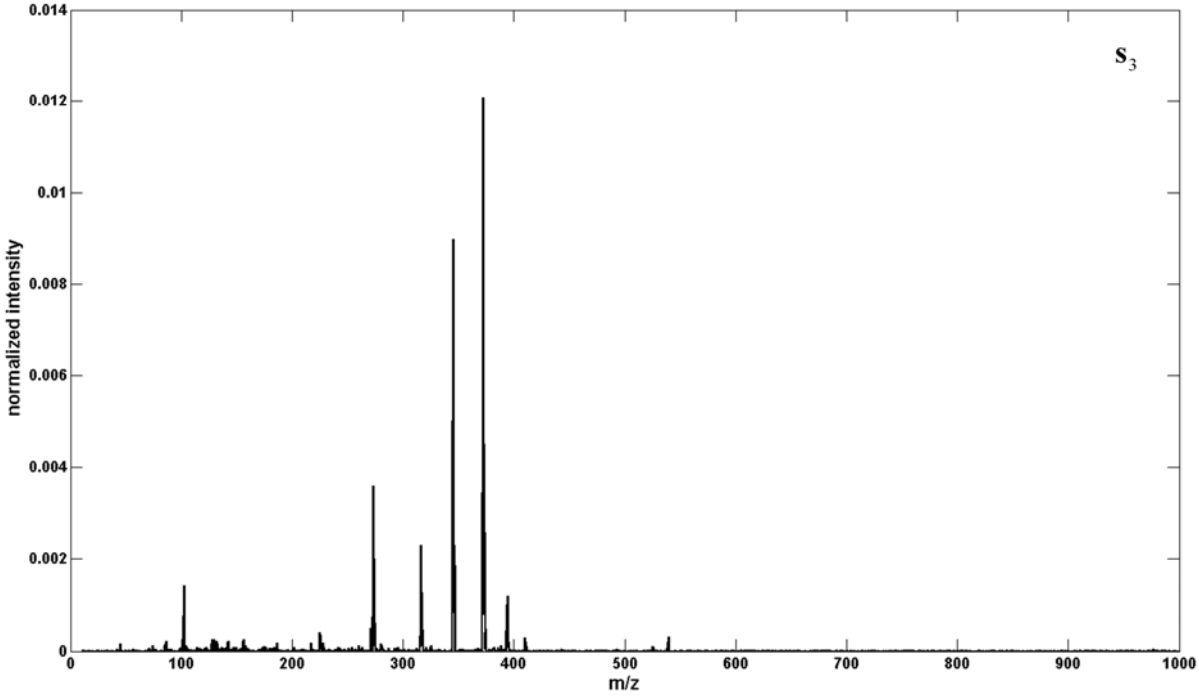
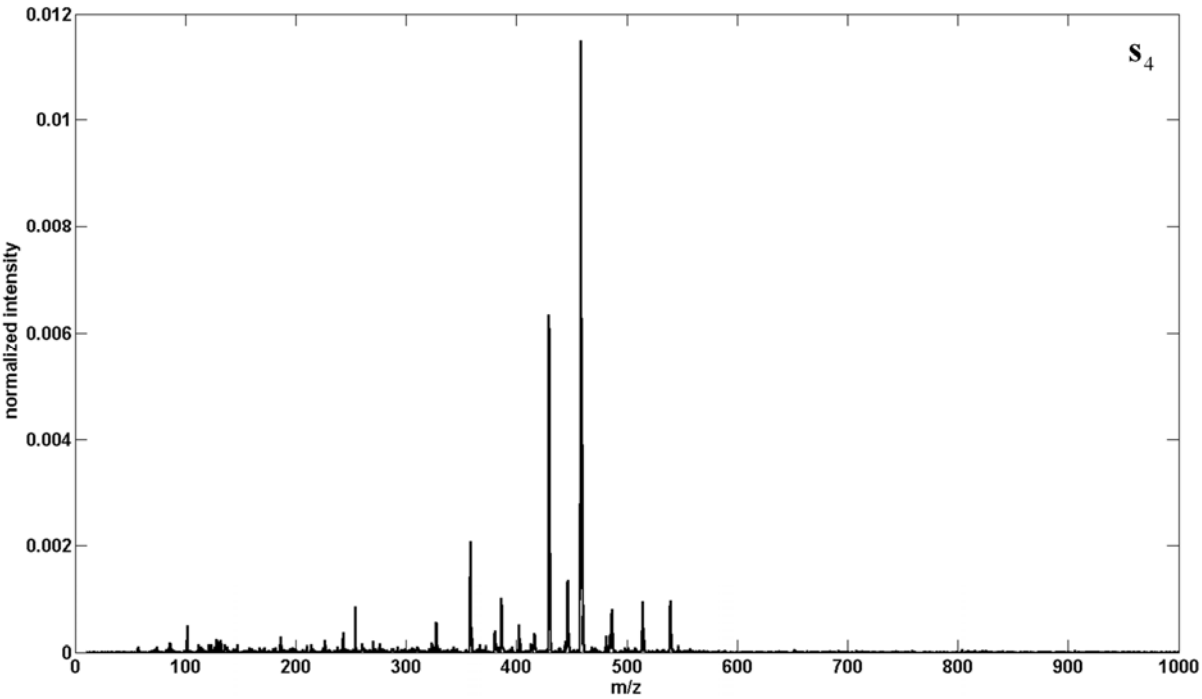

$\mathbf{x}_9$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

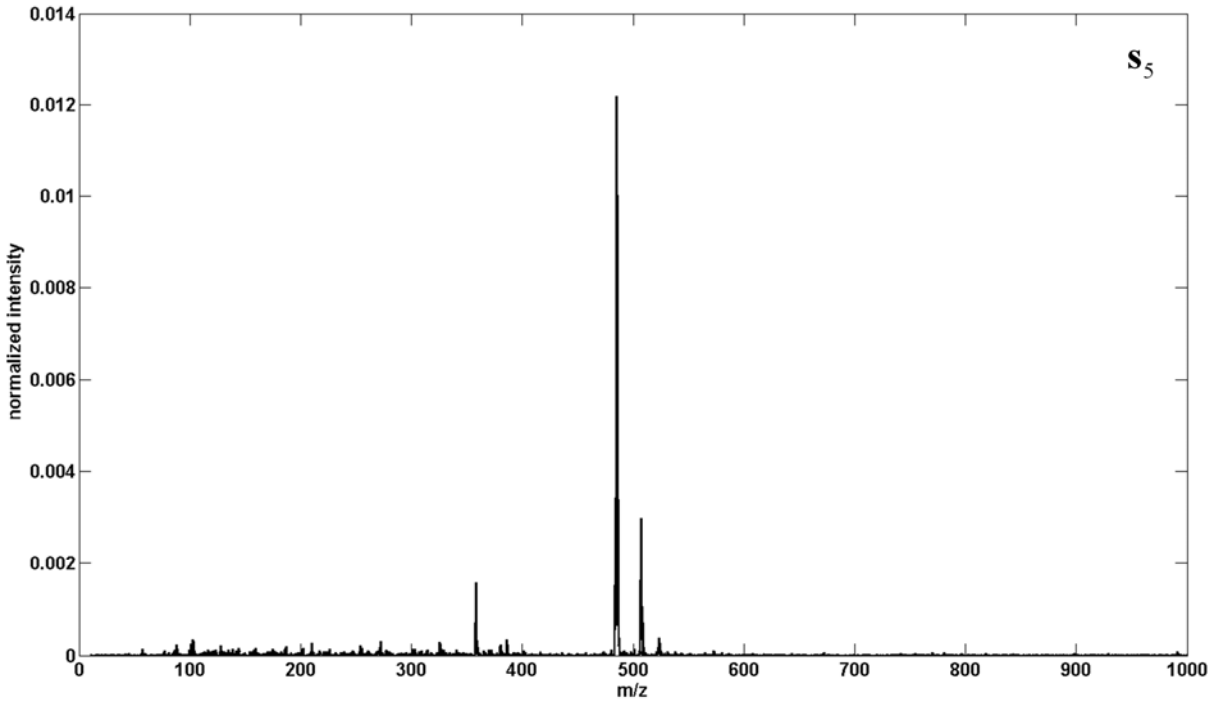
Figure S-3.

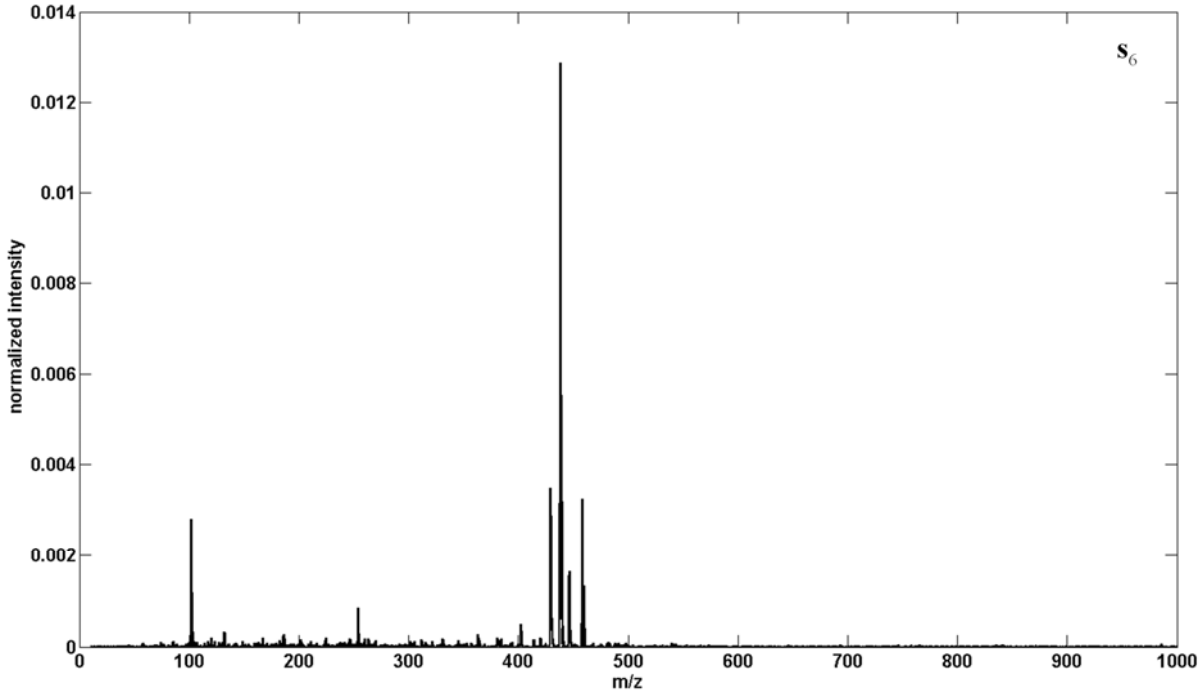

s1
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

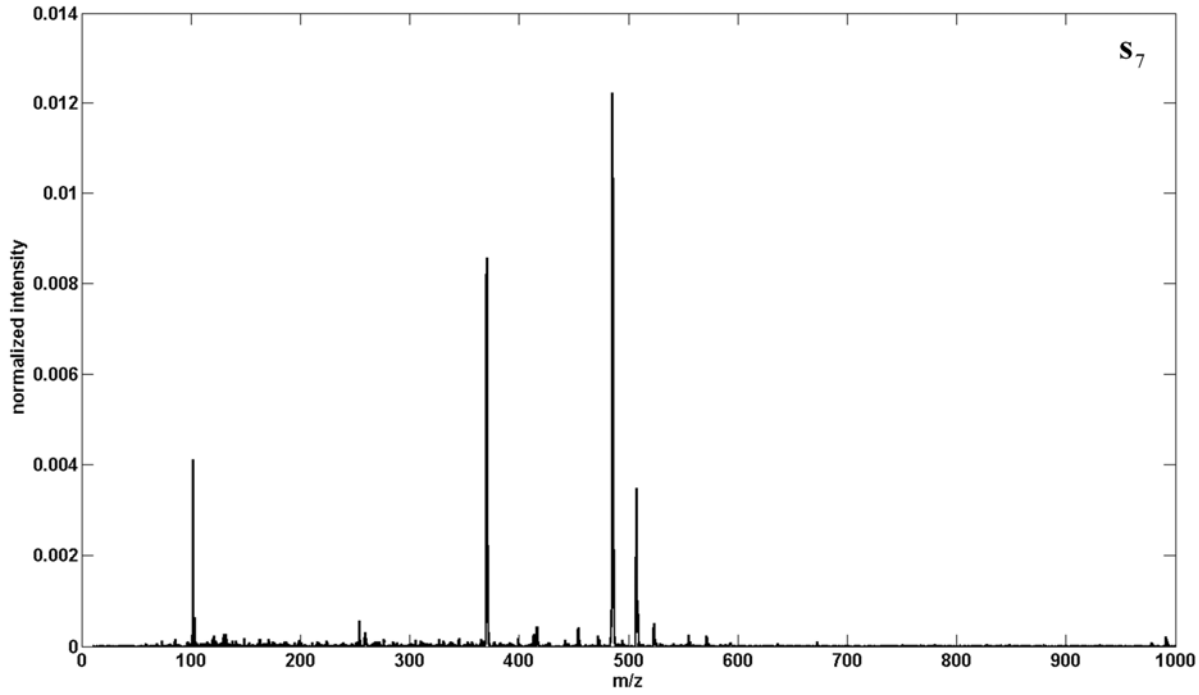

s2
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

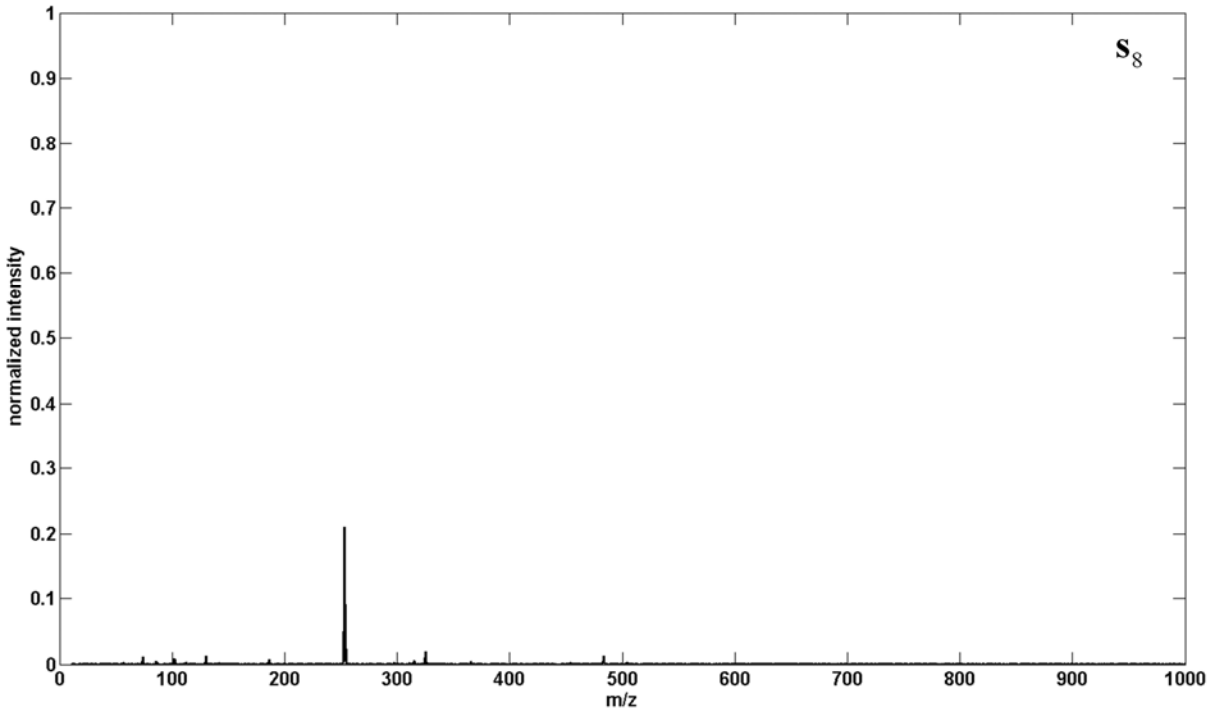

S3
normalized intensity
0.014
0.012
0.01
0.008
0.006
0.004
0.002
0
0
100
200
300
400
500
600
700
800
900
1000
m/z

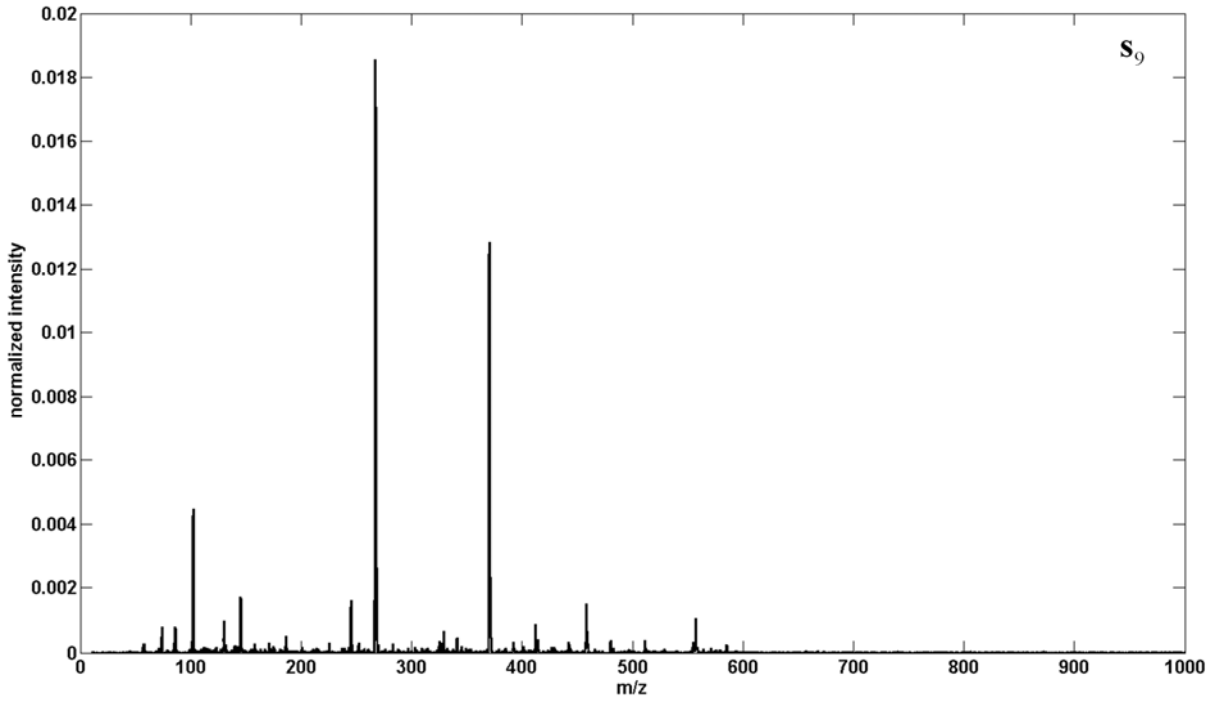

S4
normalized intensity
0.012
0.01
0.008
0.006
0.004
0.002
0
0
100
200
300
400
500
600
700
800
900
1000
m/z

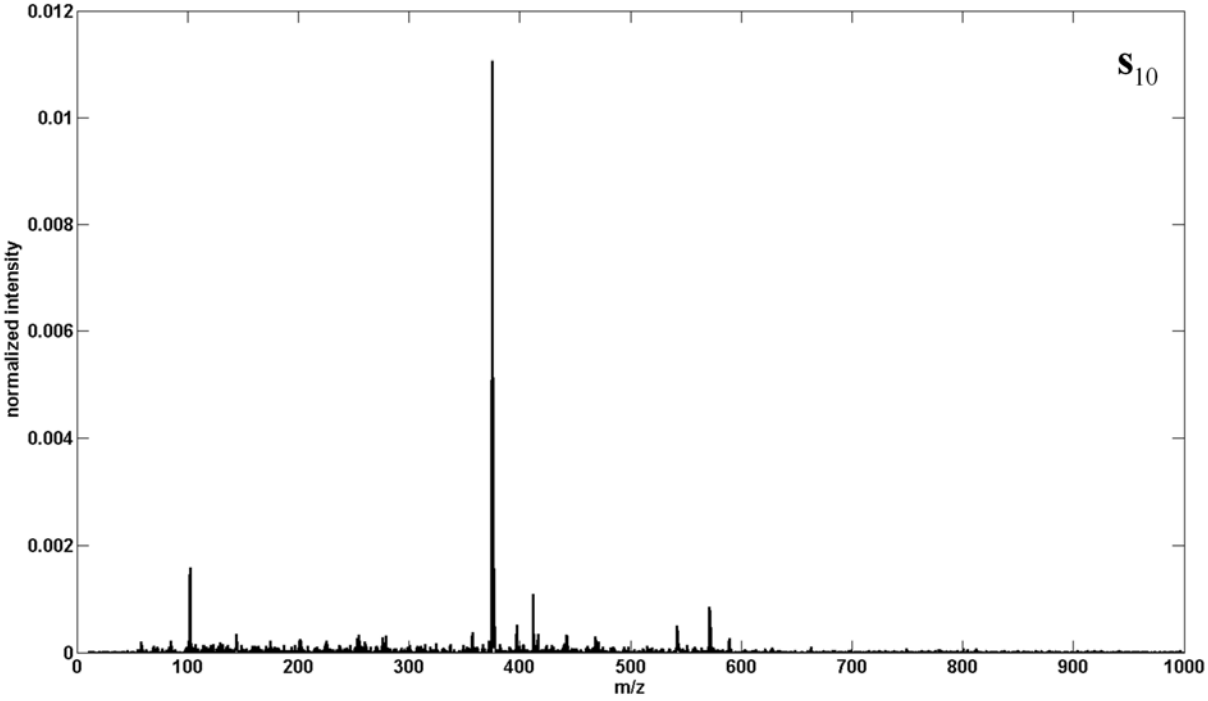

s_5
normalized intensity
m/z
0.014
0.012
0.01
0.008
0.006
0.004
0.002
0
0
100
200
300
400
500
600
700
800
900
1000

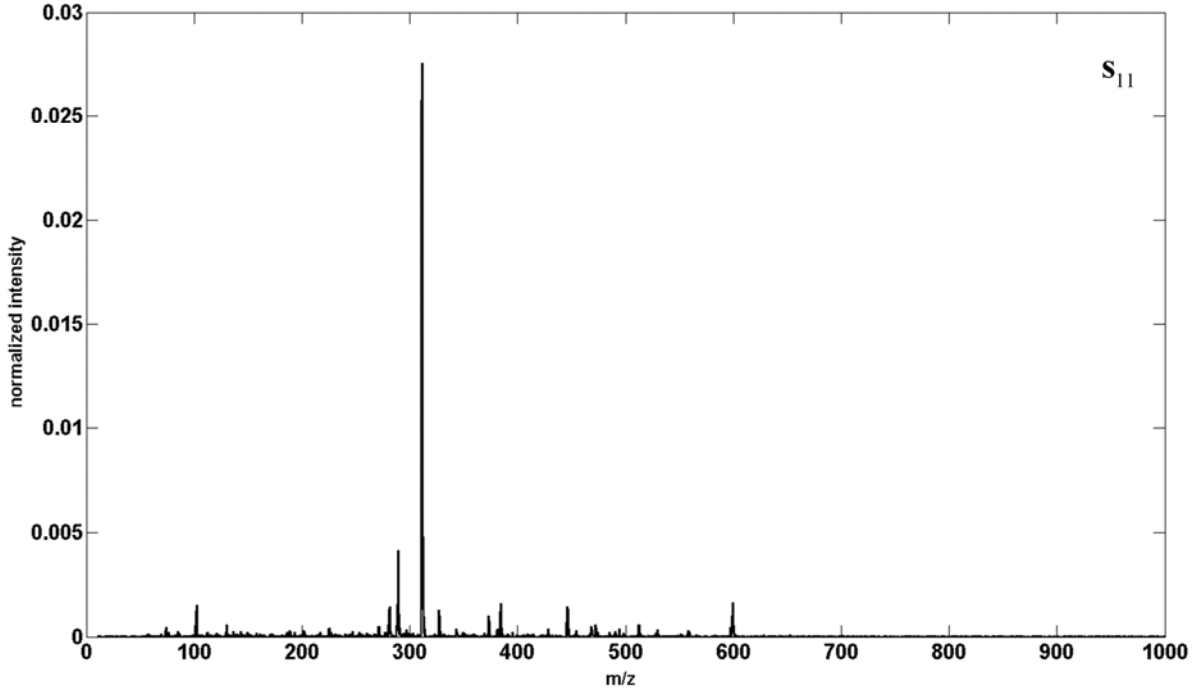

$\mathbf{s}_6$
normalized intensity
m/z
0.014
0.012
0.01
0.008
0.006
0.004
0.002
0
0
100
200
300
400
500
600
700
800
900
1000

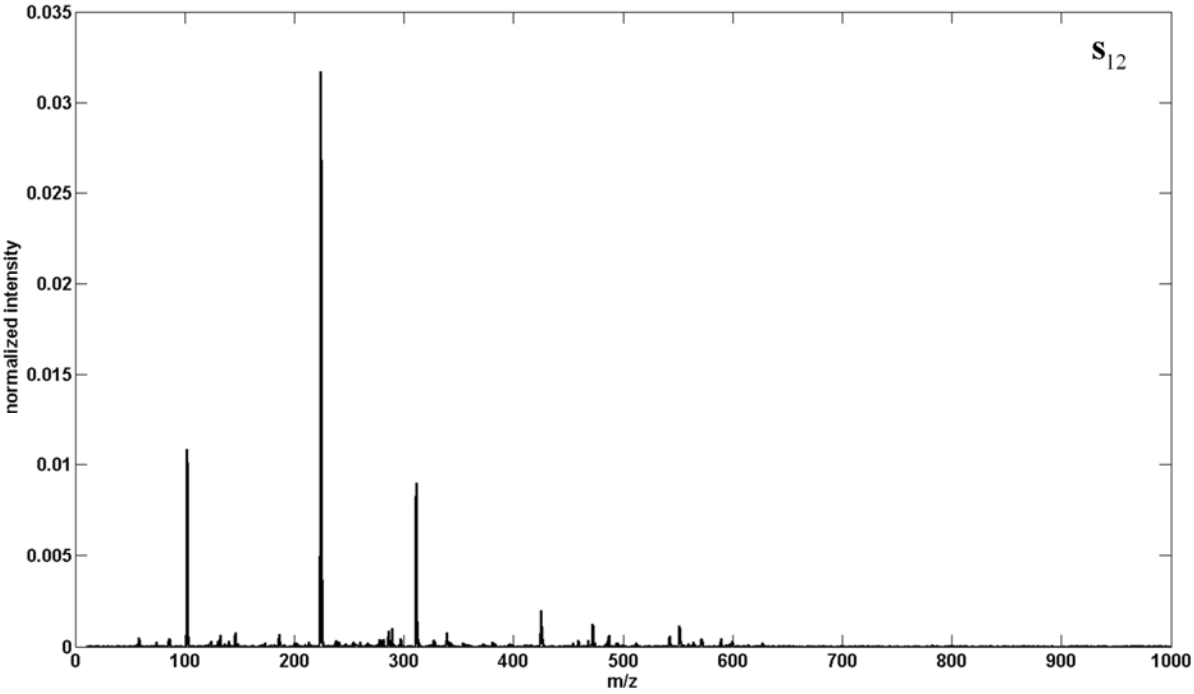

$\mathbf{s}_7$
normalized intensity
m/z
0.014
0.012
0.01
0.008
0.006
0.004
0.002
0
0
100
200
300
400
500
600
700
800
900
1000

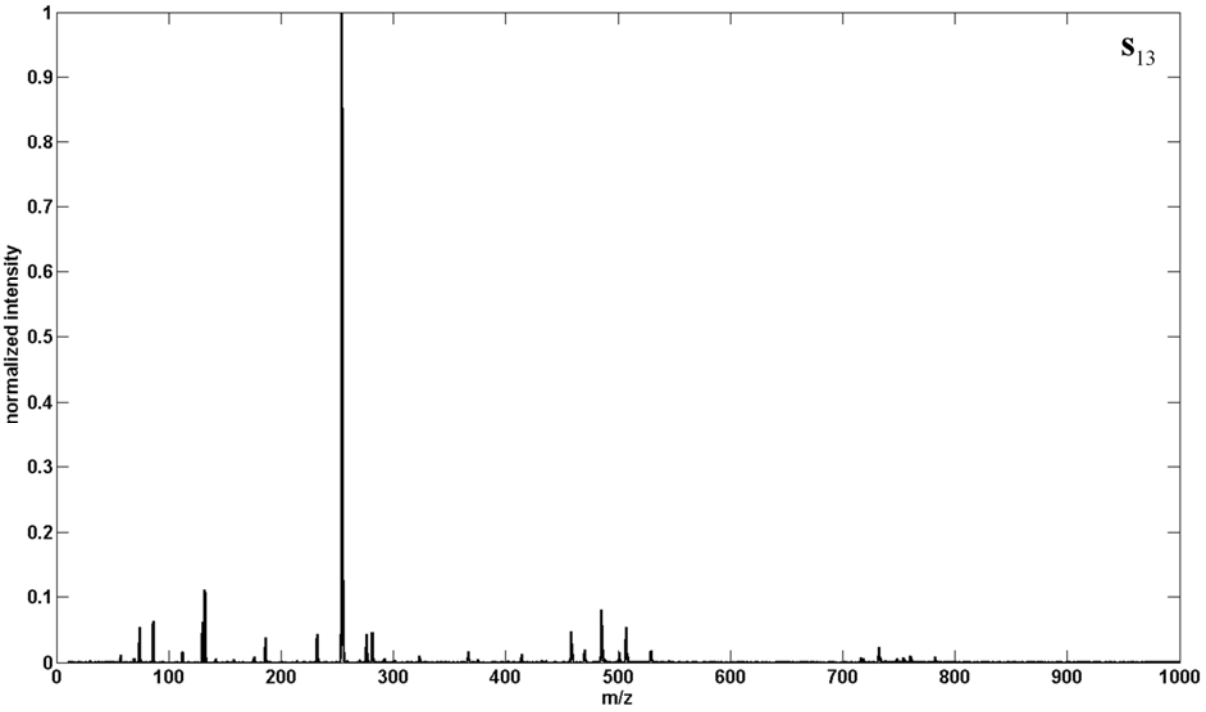

S8
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

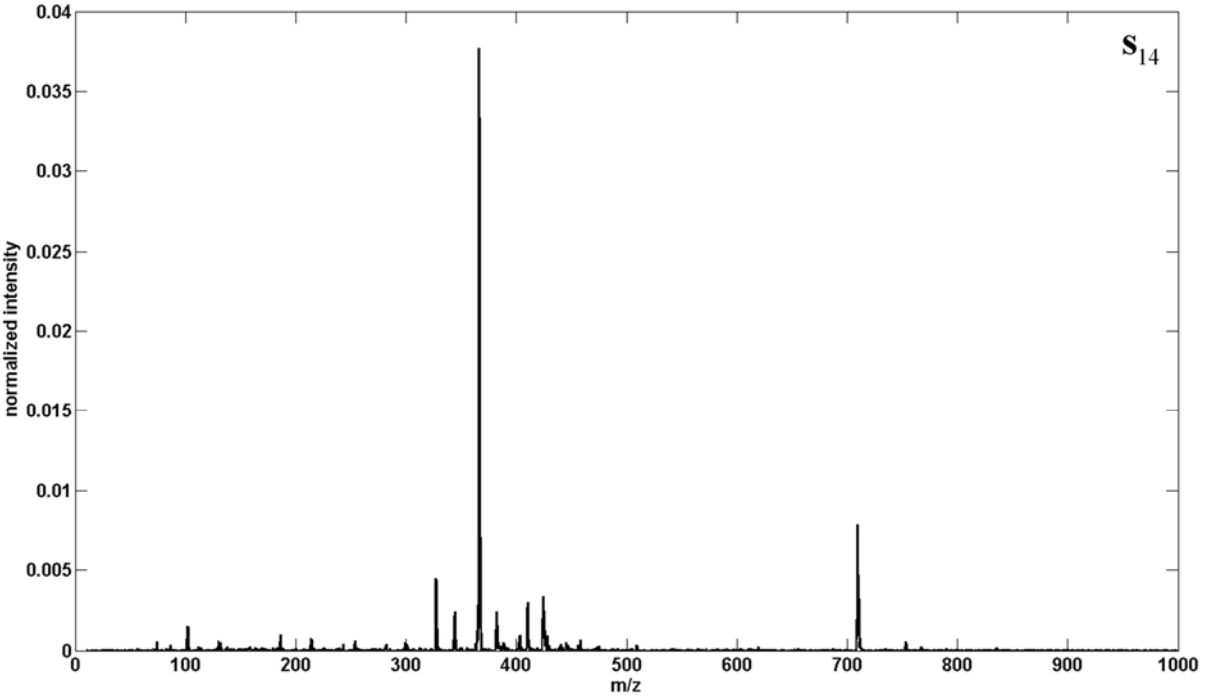

S9
normalized intensity
m/z
0
0.002
0.004
0.006
0.008
0.01
0.012
0.014
0.016
0.018
0.02
0
100
200
300
400
500
600
700
800
900
1000

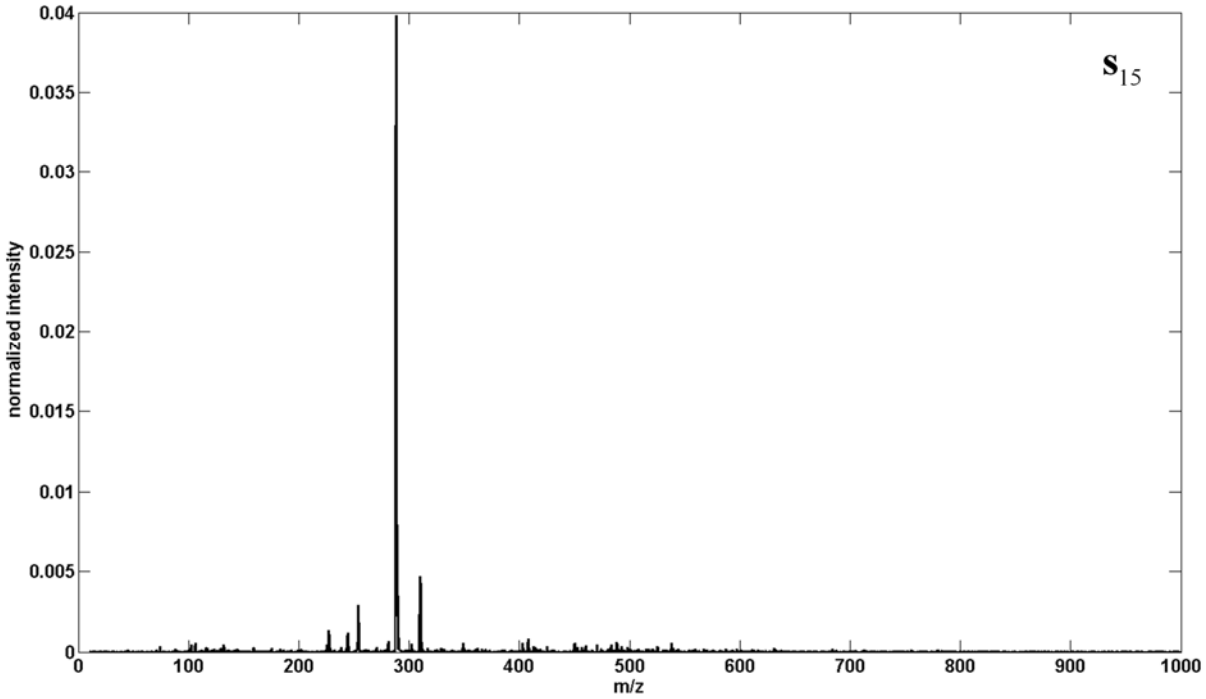

$\mathbf{S}_{10}$
normalized intensity
0.012
0.01
0.008
0.006
0.004
0.002
0
0
100
200
300
400
500
600
700
800
900
1000
m/z

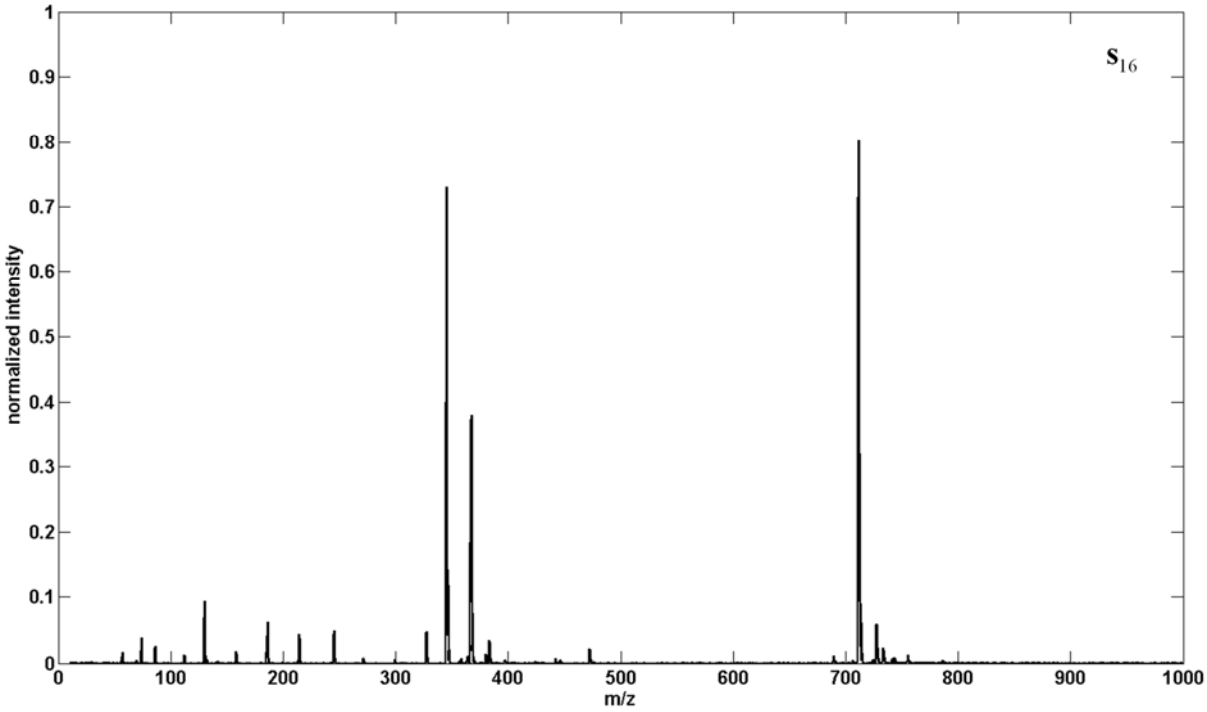

s_{11}
normalized intensity
m/z
0.03
0.025
0.02
0.015
0.01
0.005
0
0
100
200
300
400
500
600
700
800
900
1000

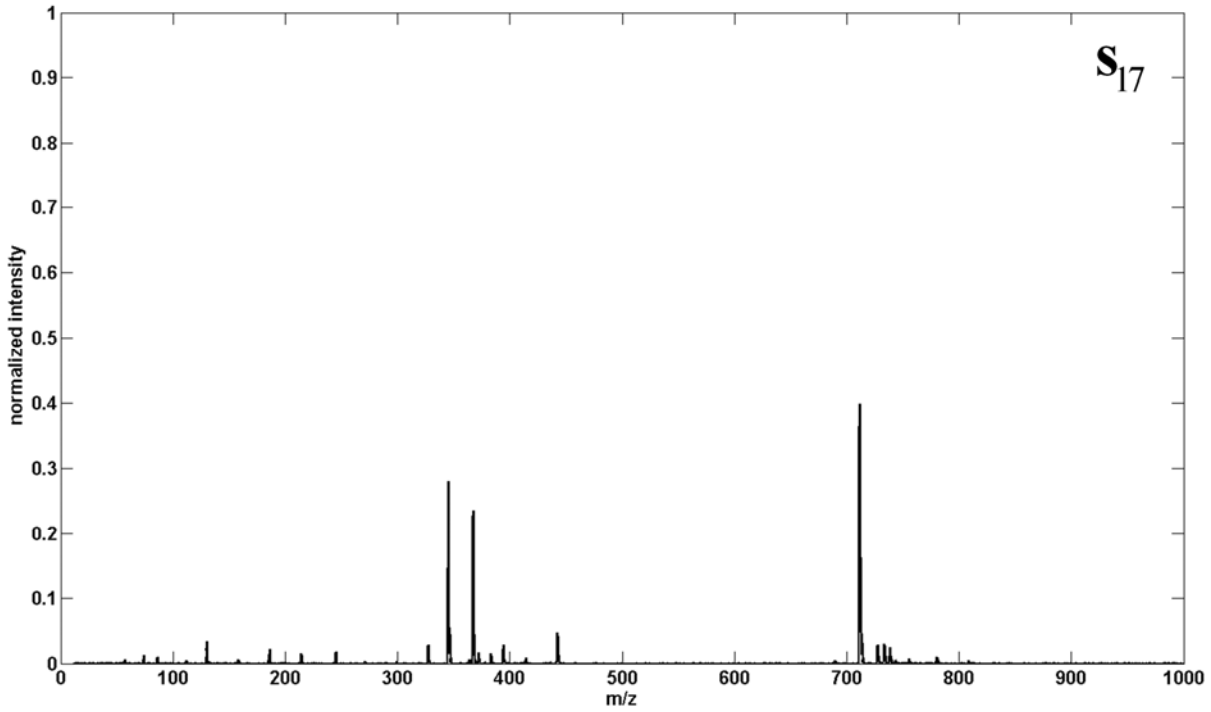

s_{12}
normalized intensity
m/z
0.035
0.03
0.025
0.02
0.015
0.01
0.005
0
0
100
200
300
400
500
600
700
800
900
1000

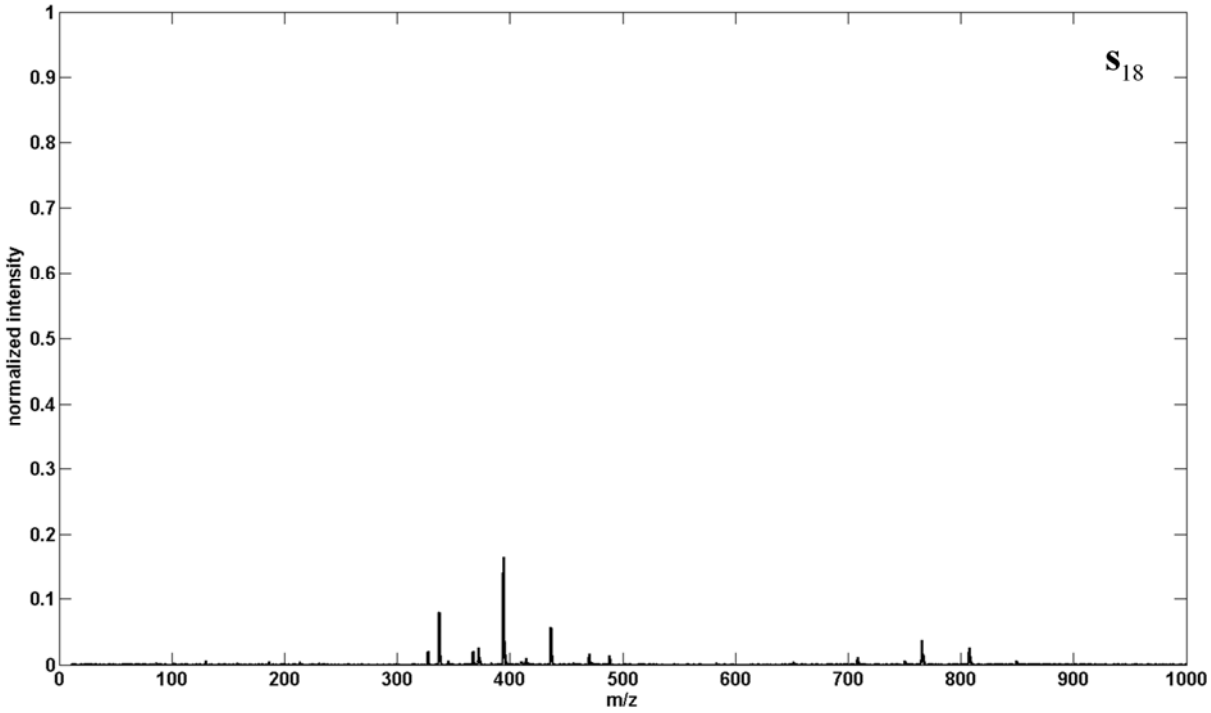

$\mathbf{s}_{13}$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

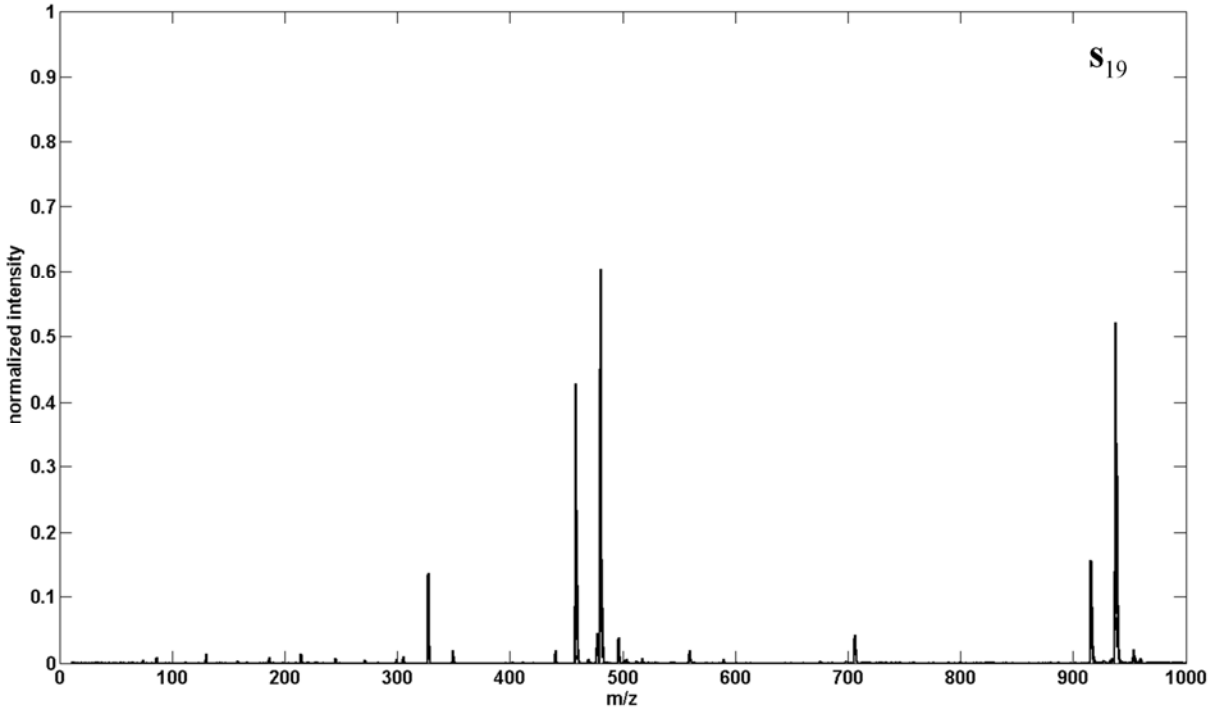

$\mathbf{s}_{14}$
normalized intensity
m/z
0
0.005
0.01
0.015
0.02
0.025
0.03
0.035
0.04
0
100
200
300
400
500
600
700
800
900
1000

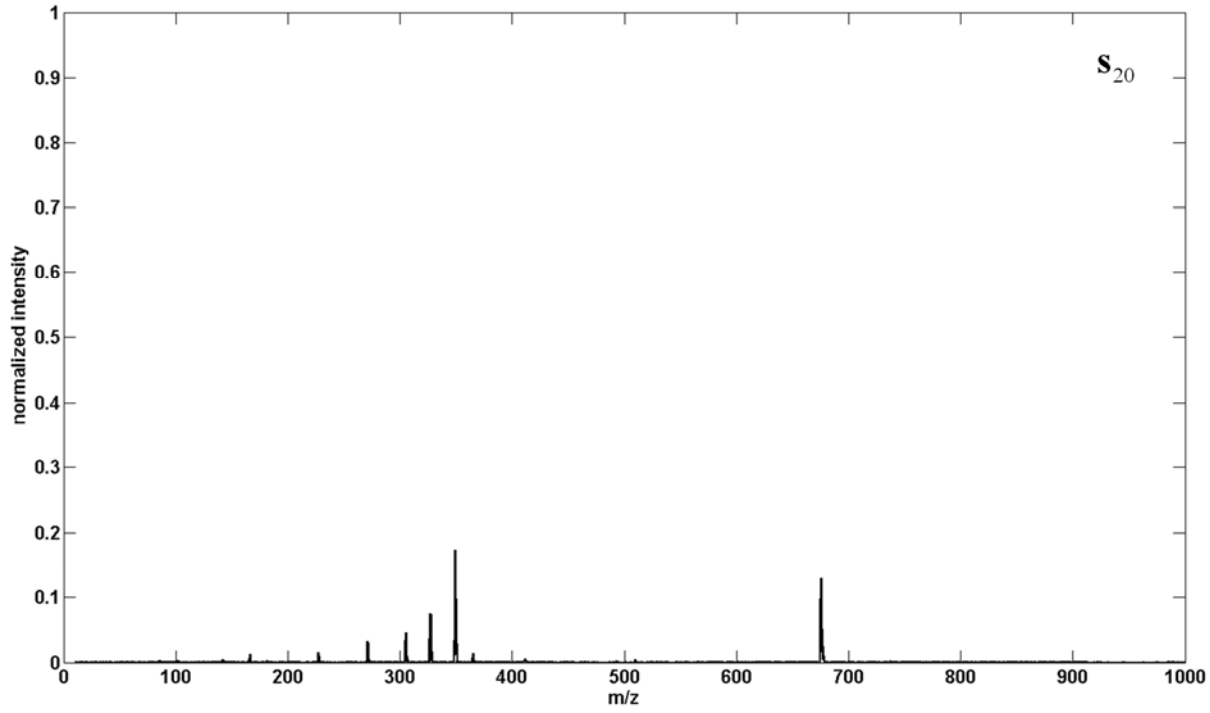

s15
normalized intensity
m/z
0.04
0.035
0.03
0.025
0.02
0.015
0.01
0.005
0
100
200
300
400
500
600
700
800
900
1000

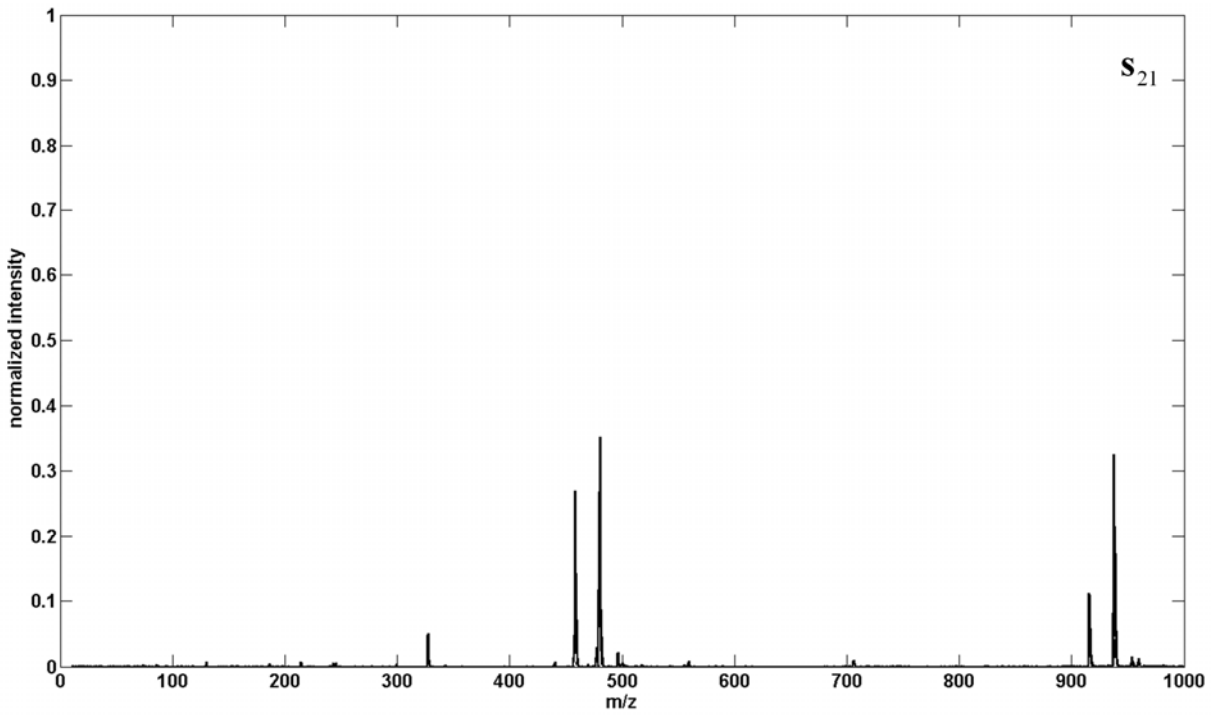

s16
normalized intensity
m/z
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
100
200
300
400
500
600
700
800
900
1000

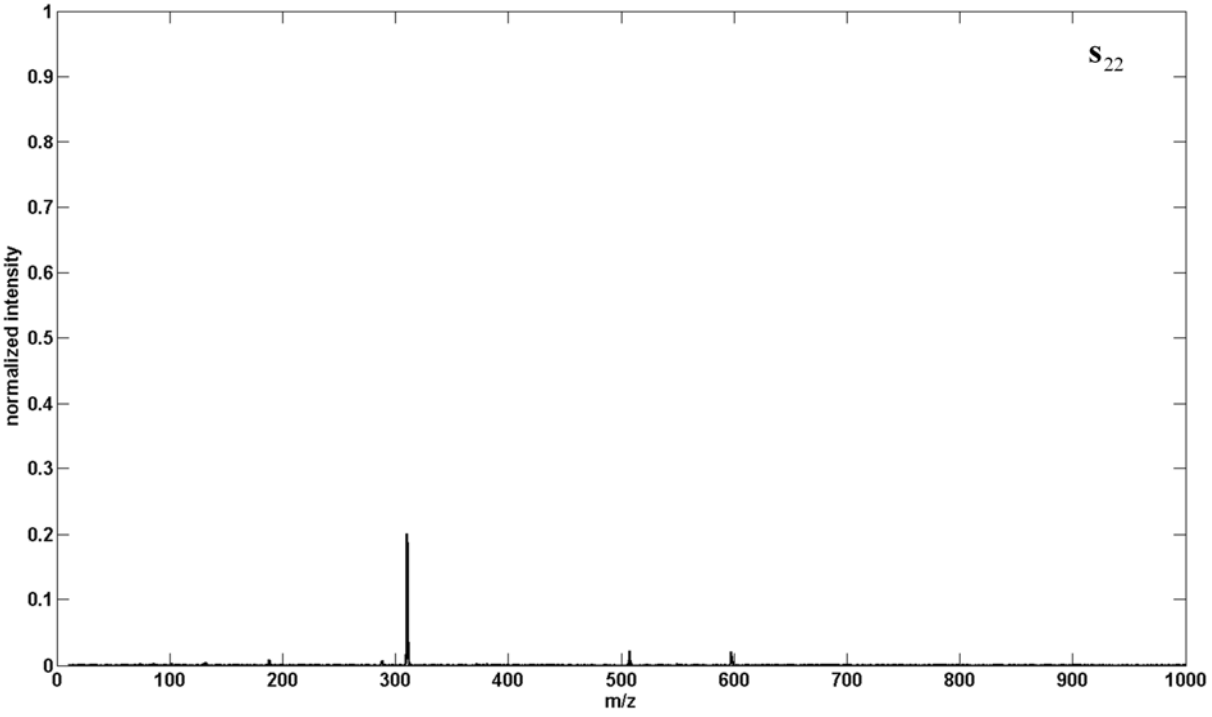

s_{17}
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

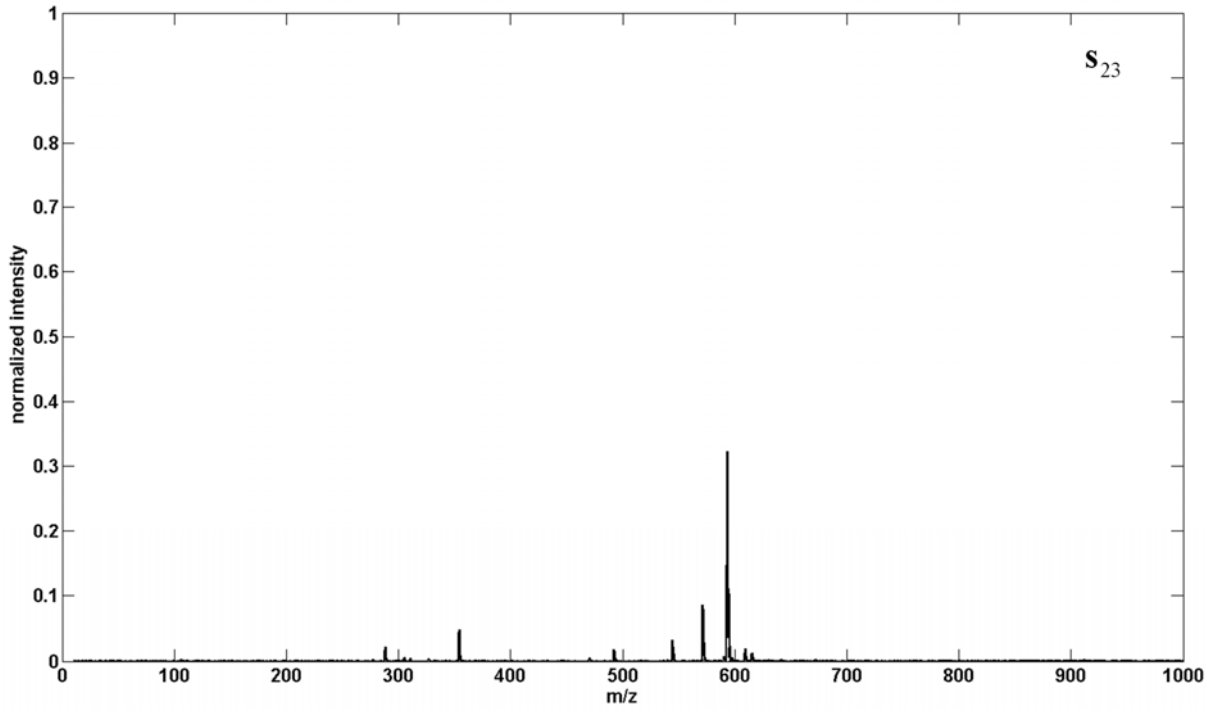

s_{18}
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

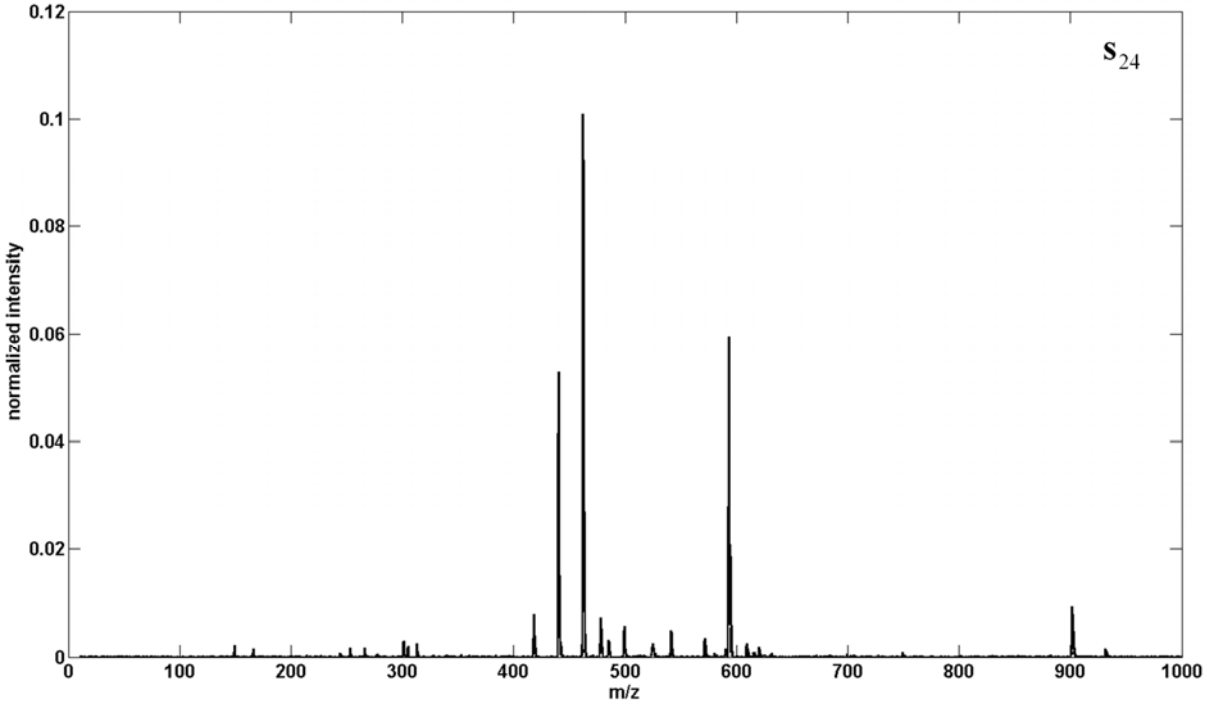

s19
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000


s20
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000


$\mathbf{s}_{21}$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000


$\mathbf{s}_{22}$
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000


s_{23}
normalized intensity
m/z


s_{24}
normalized intensity
m/z

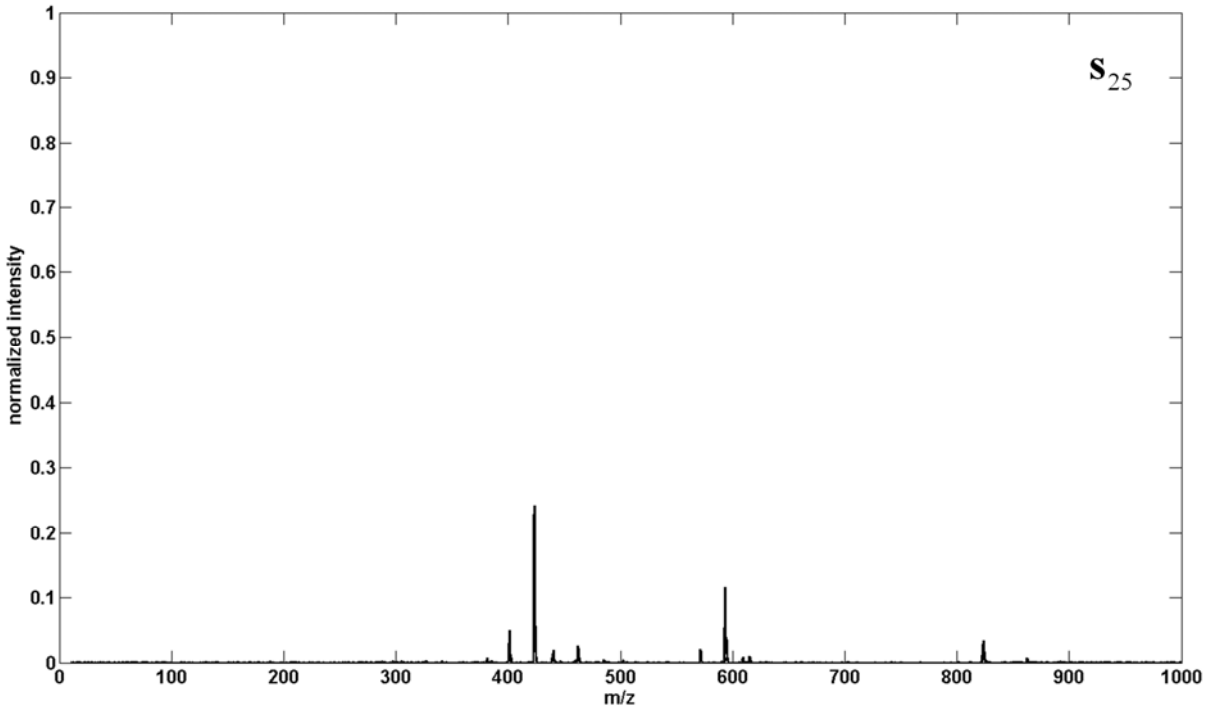


Figure S-4.

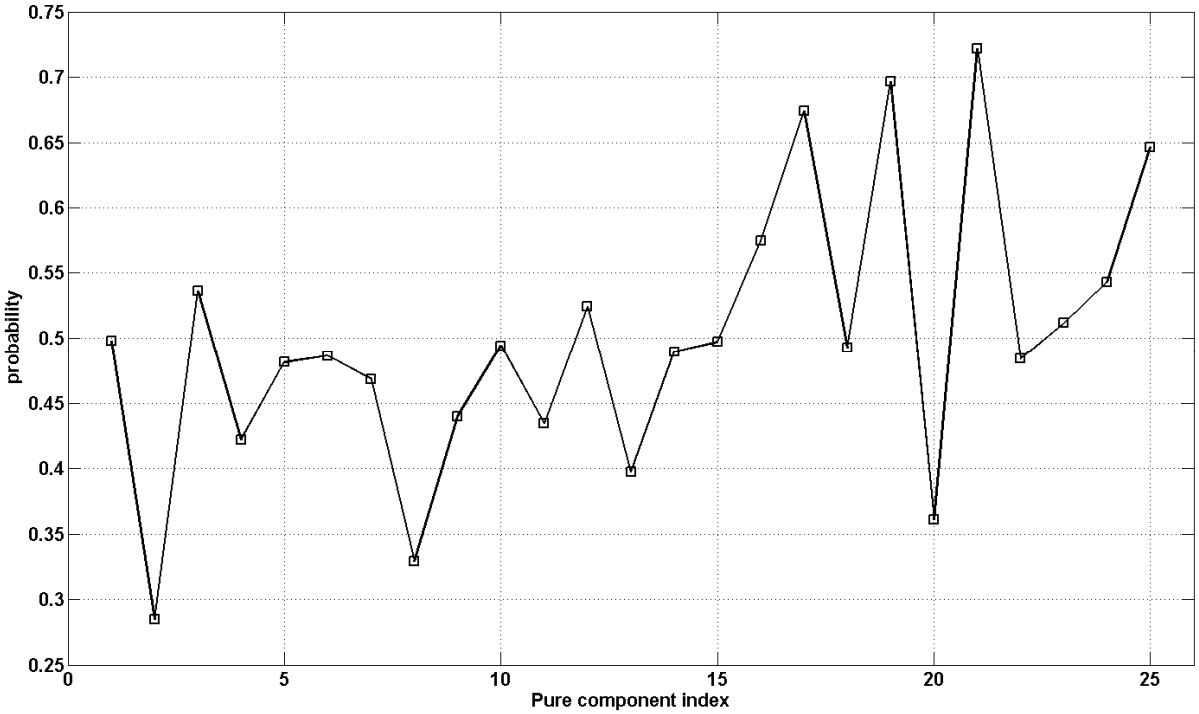


Figure S-5.

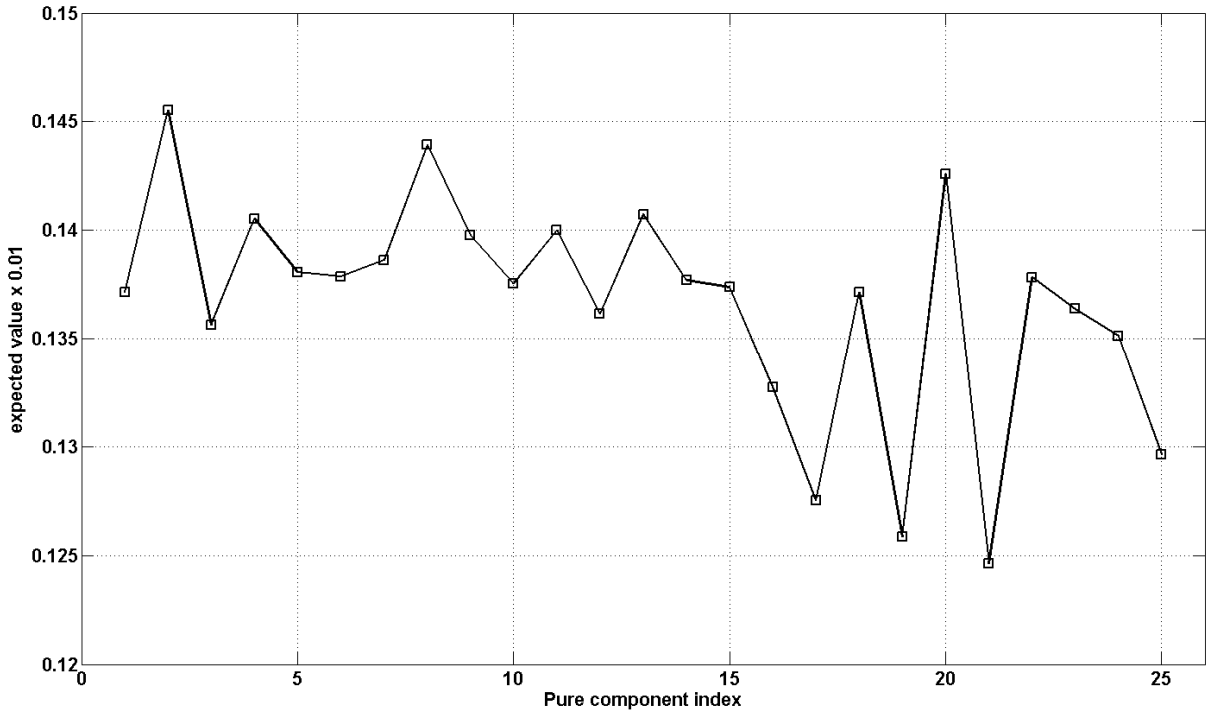


Figure S-6.

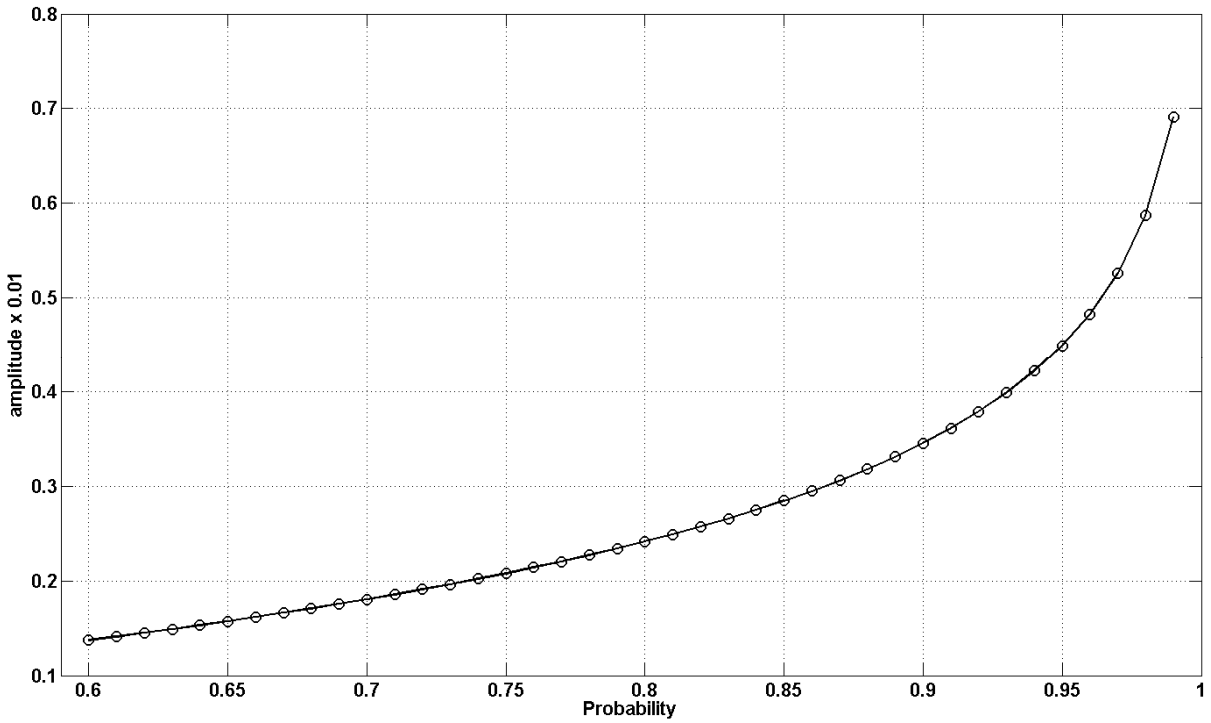
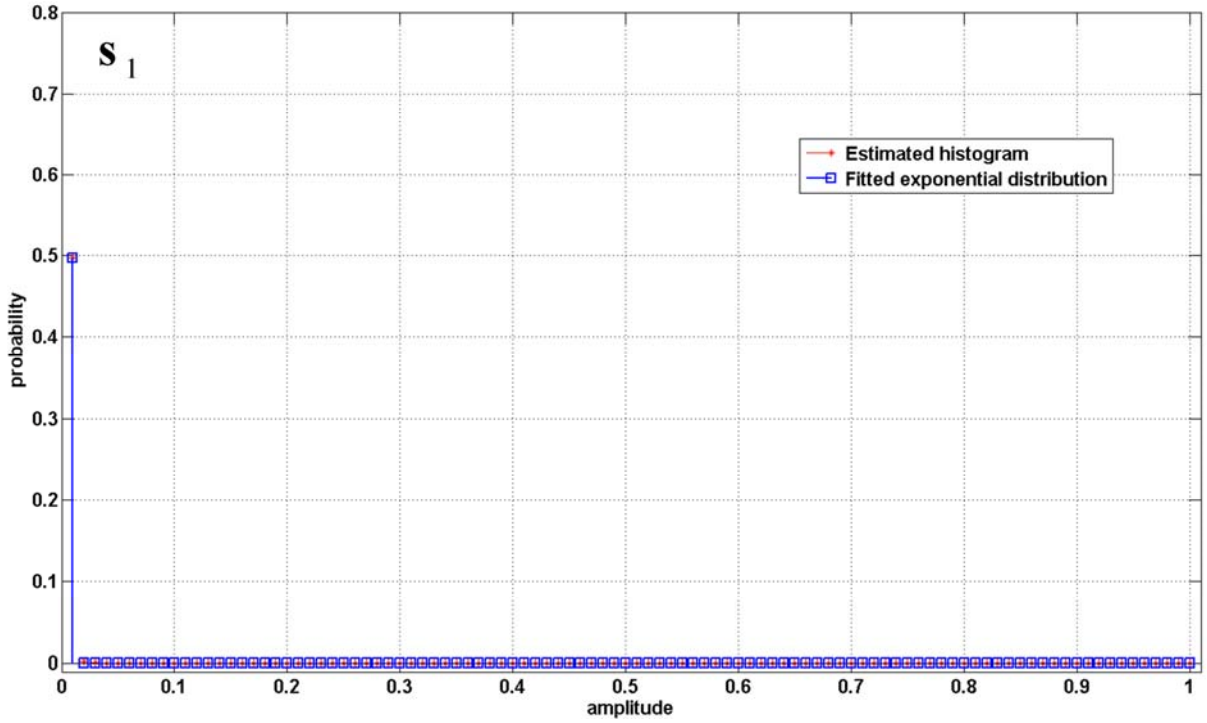
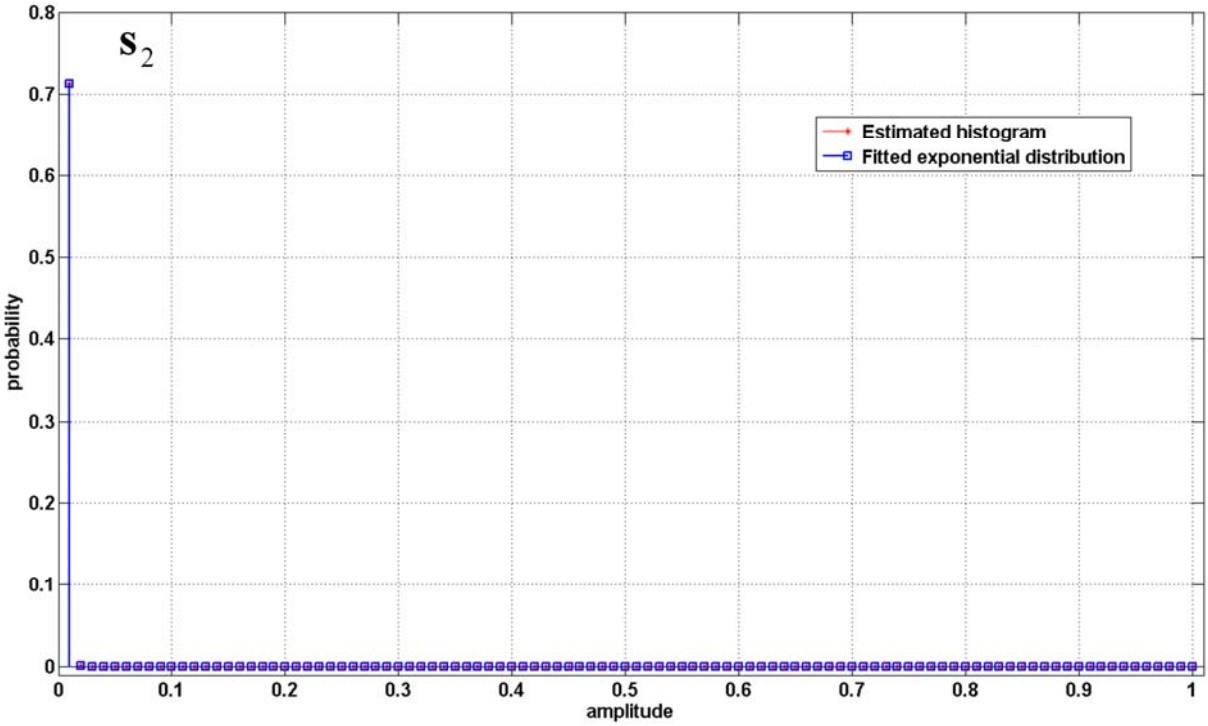
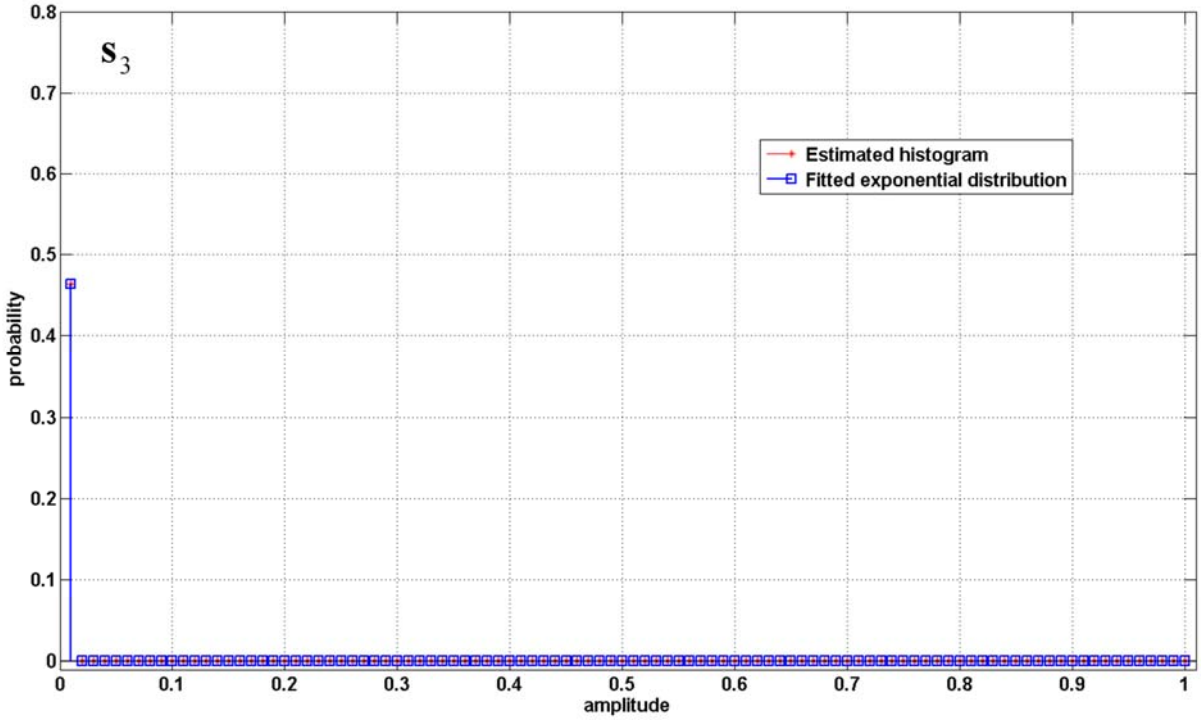
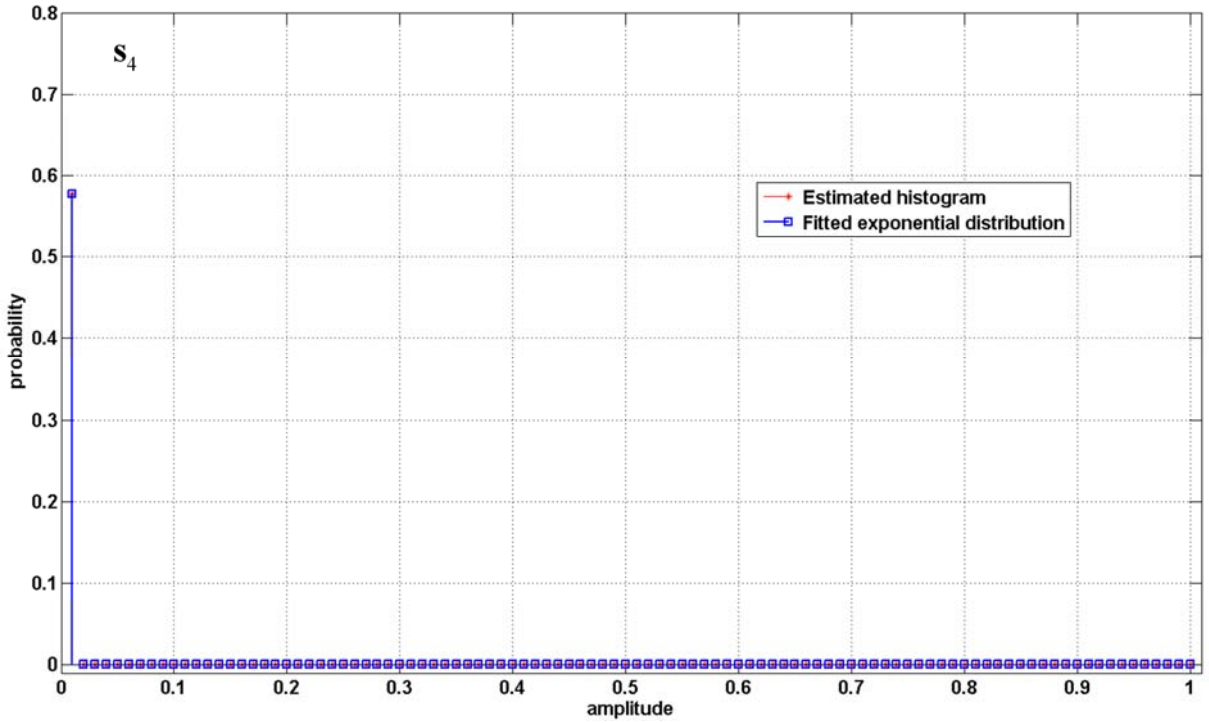


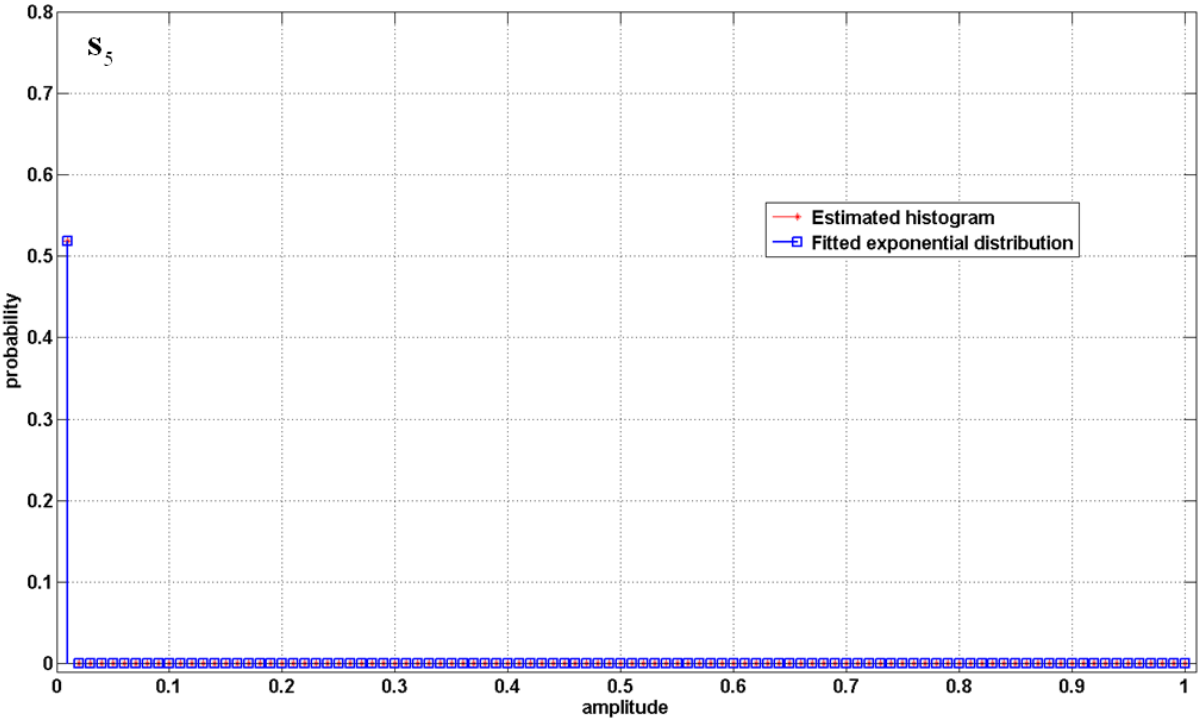
Figure S-7.

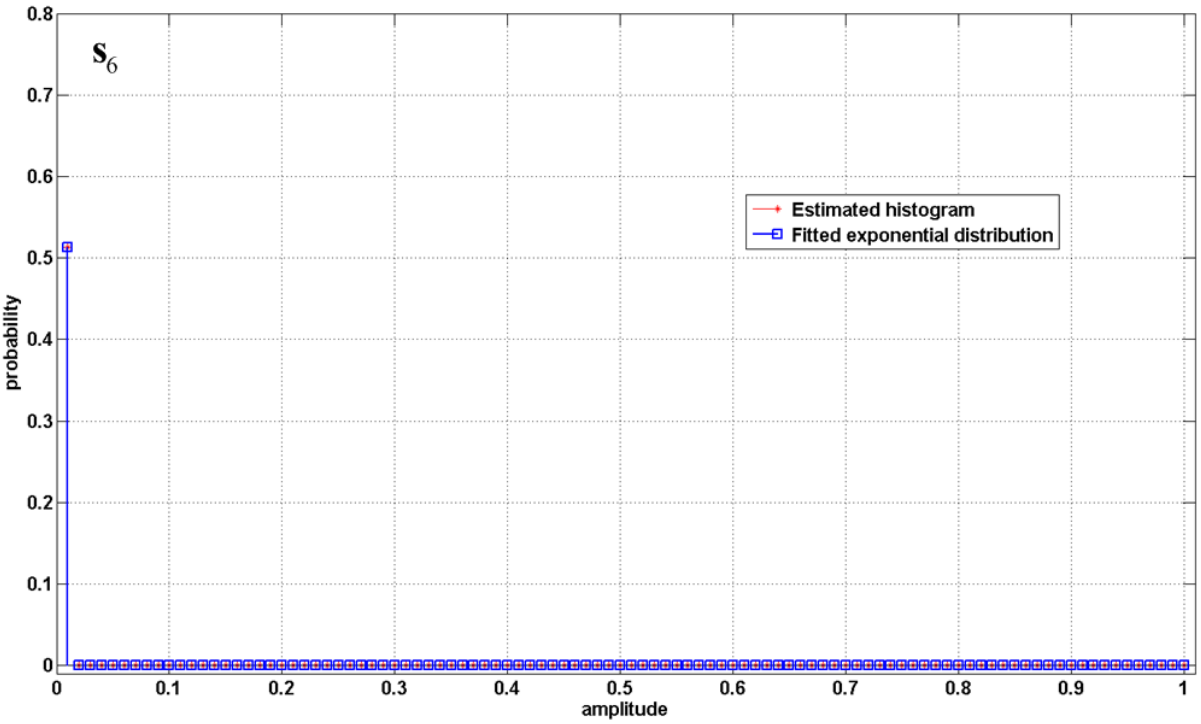

s 1
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

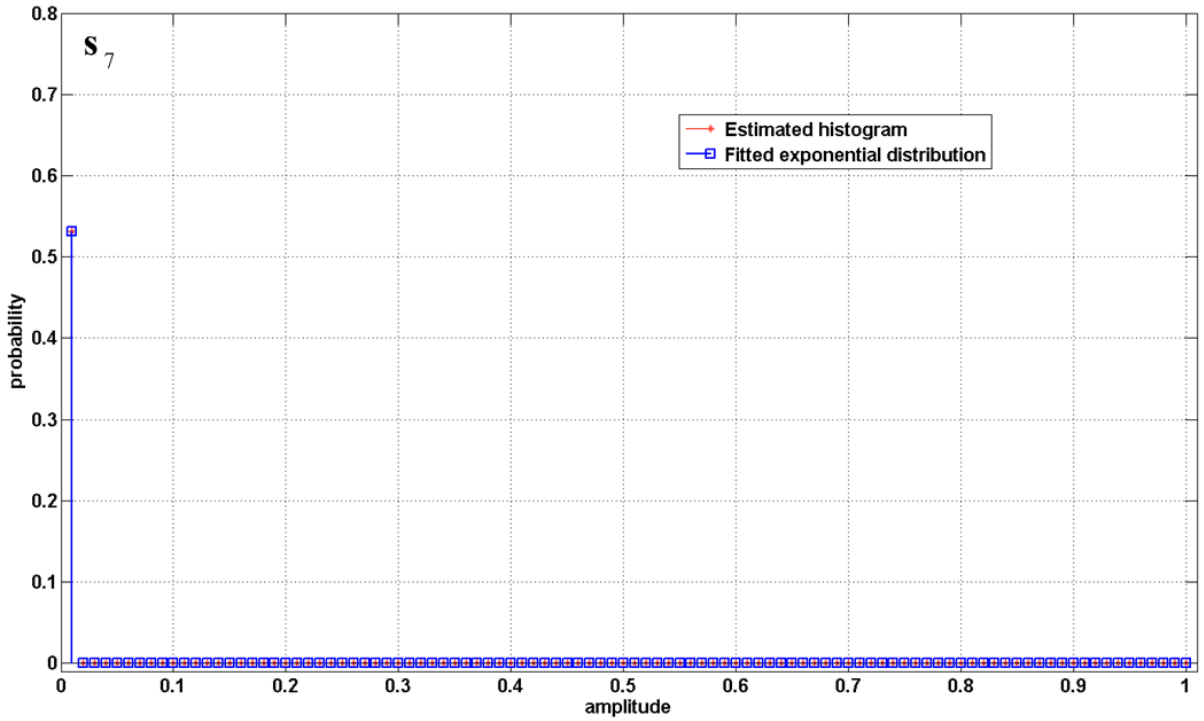

s 2
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

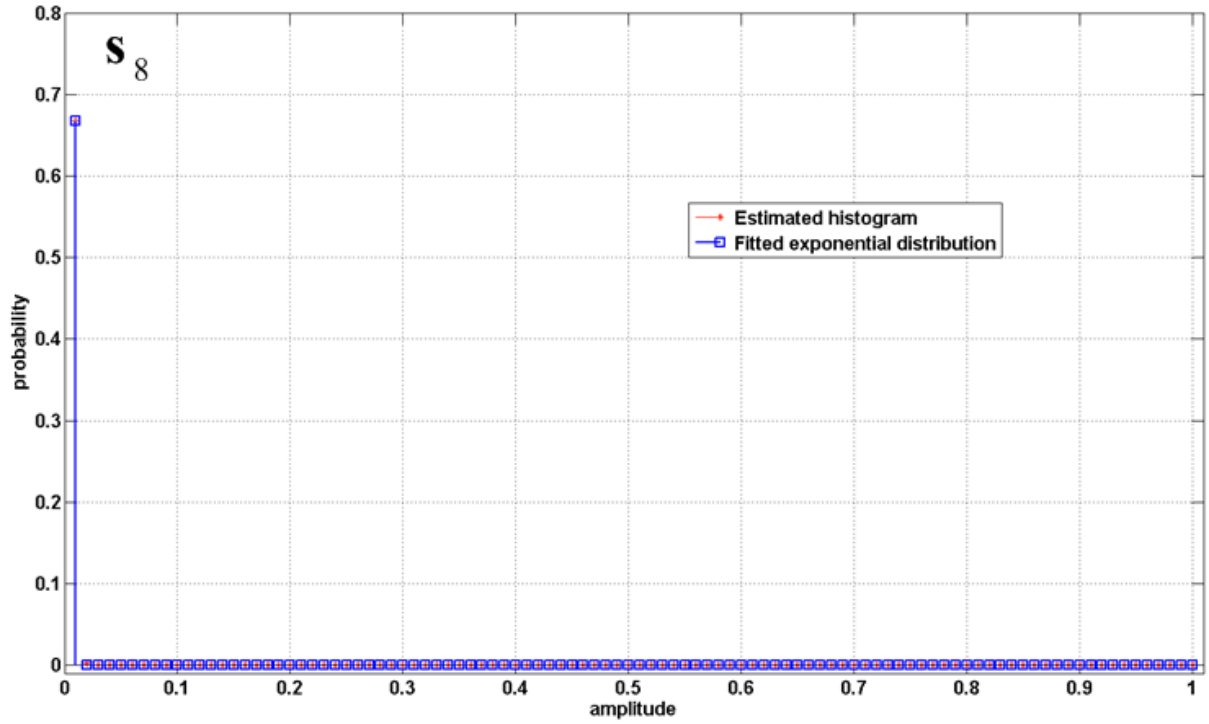

s_3
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

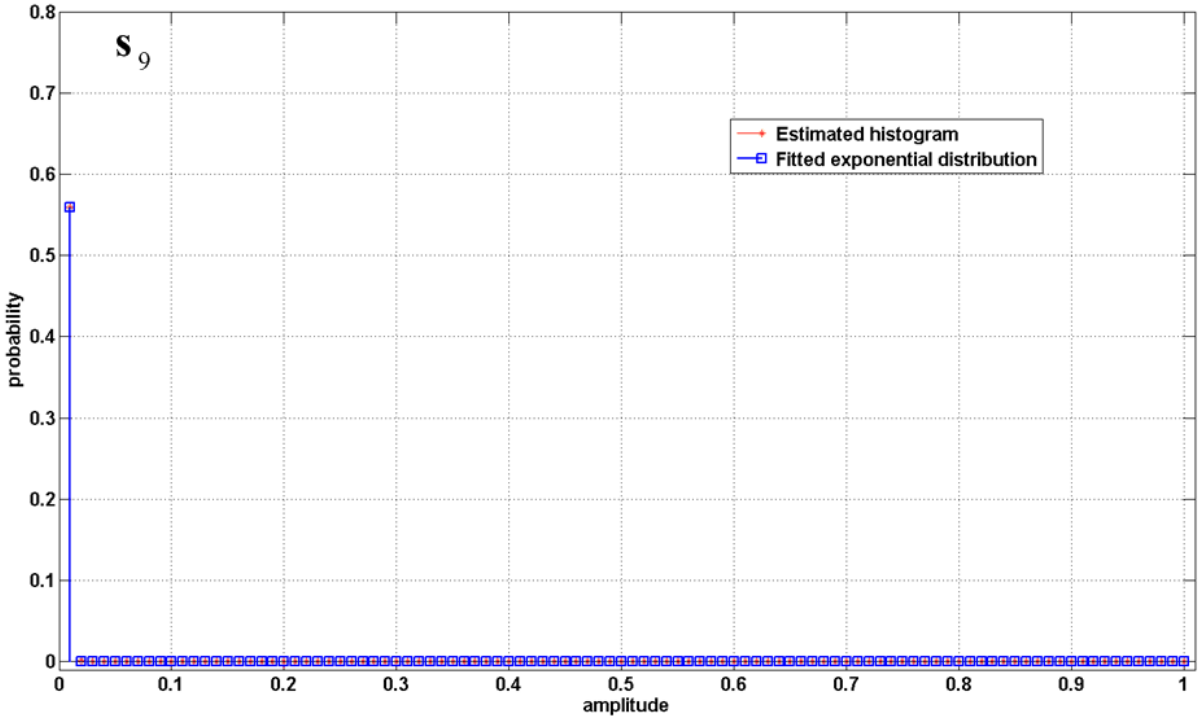

s_4
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

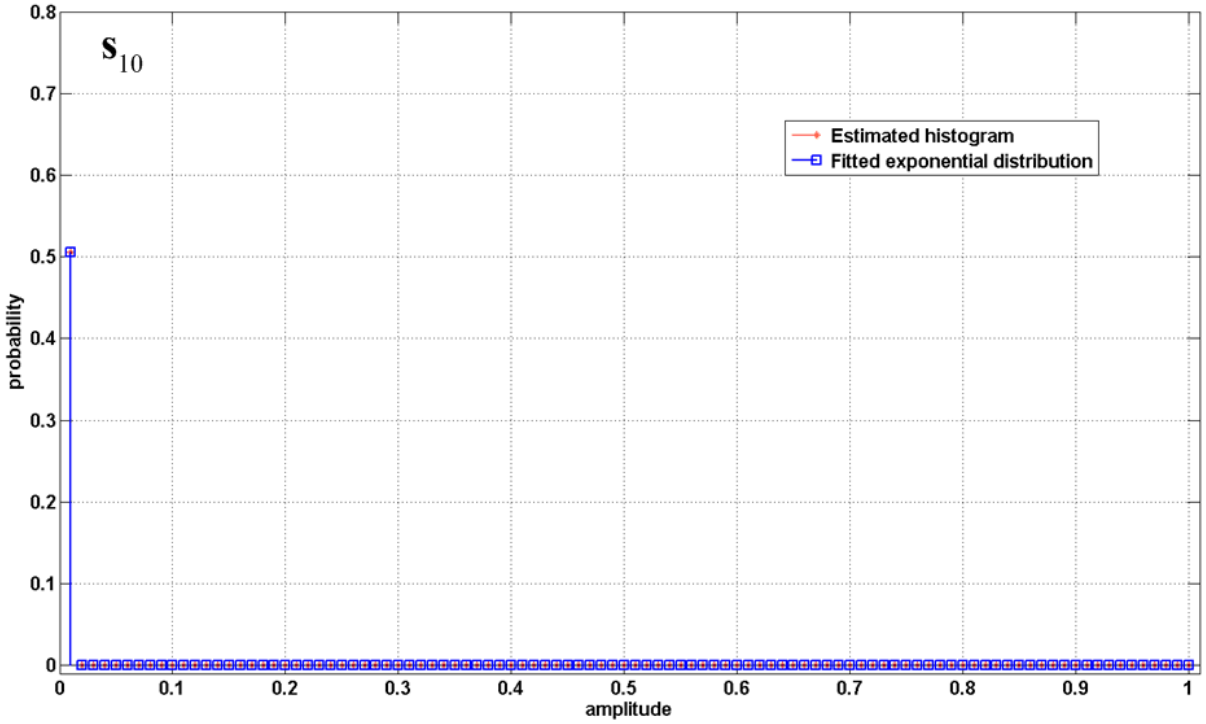

s_5
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

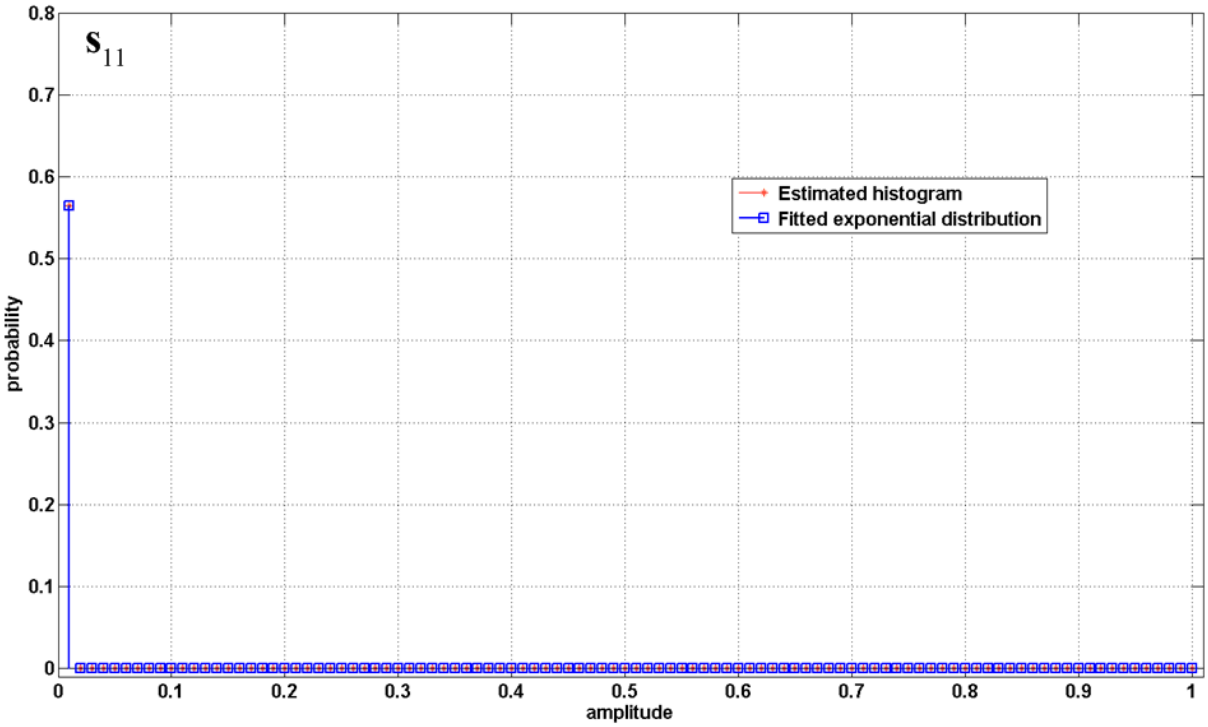

s_6
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

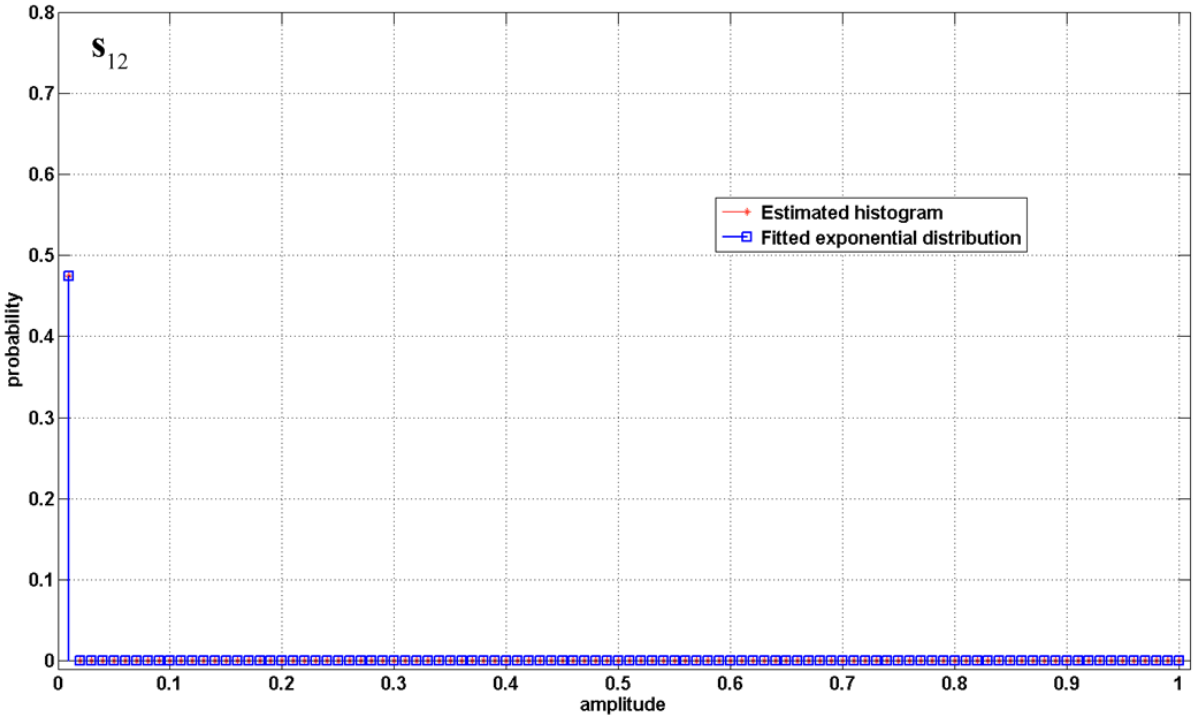

s 7
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

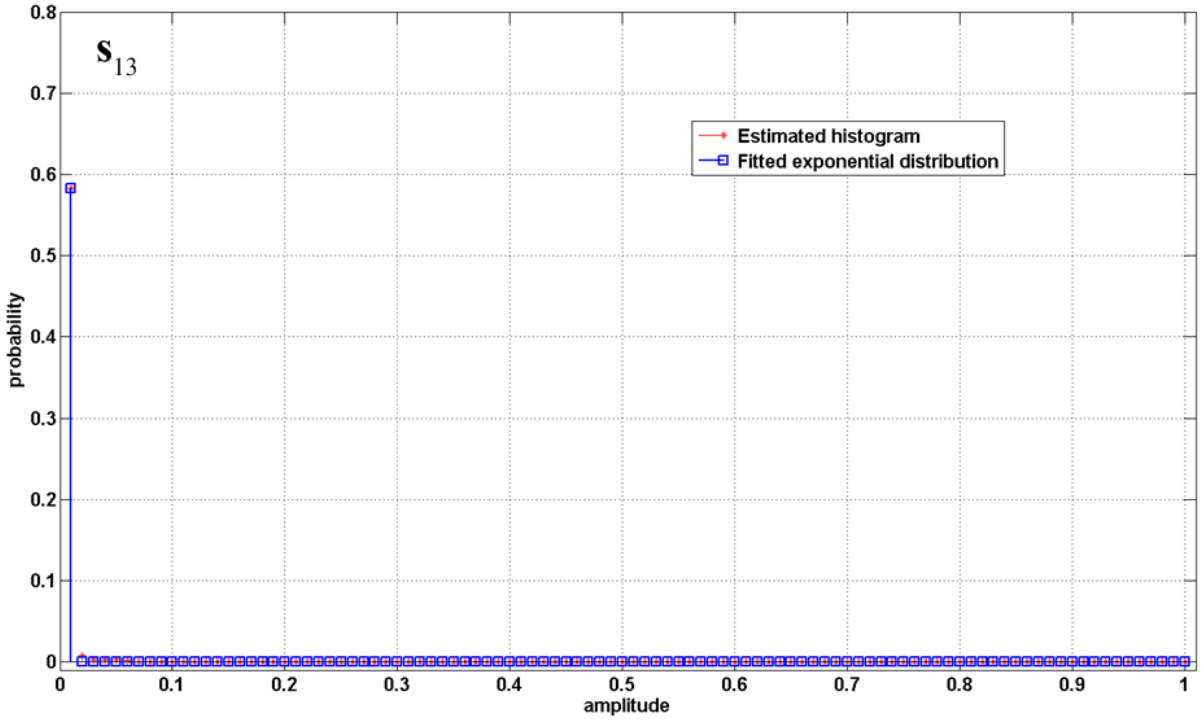

s 8
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

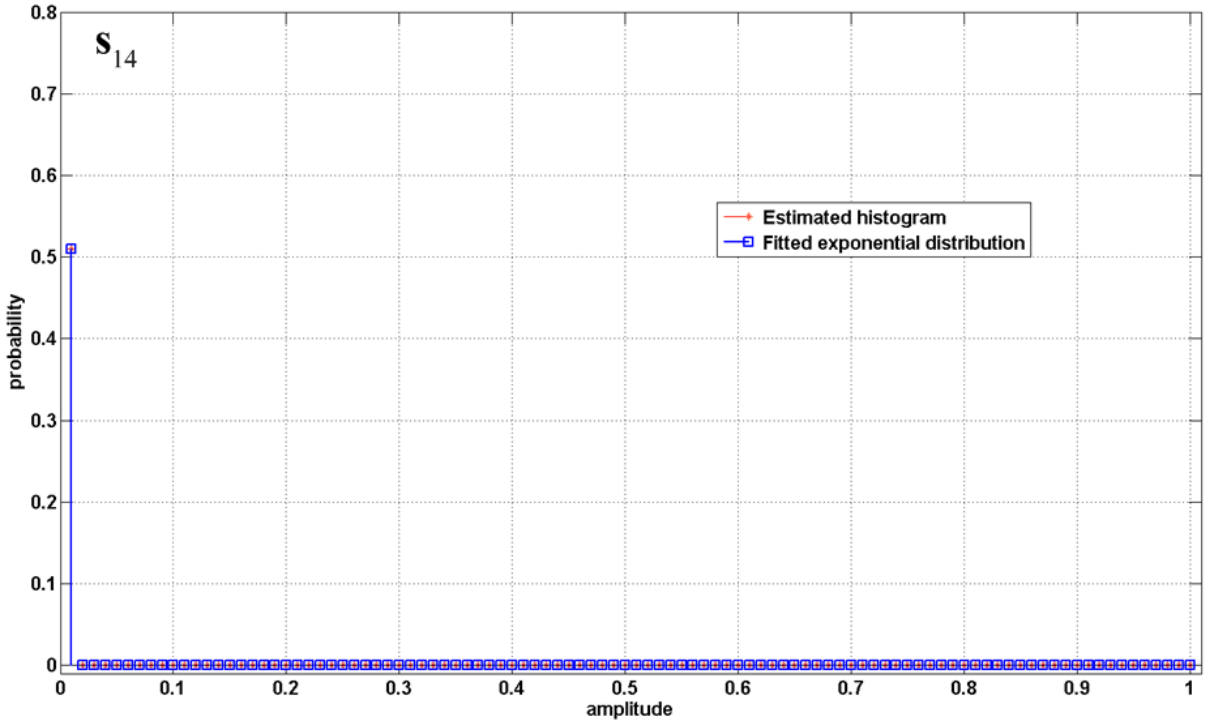

s 9
Estimated histogram
Fitted exponential distribution
probability
amplitude
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

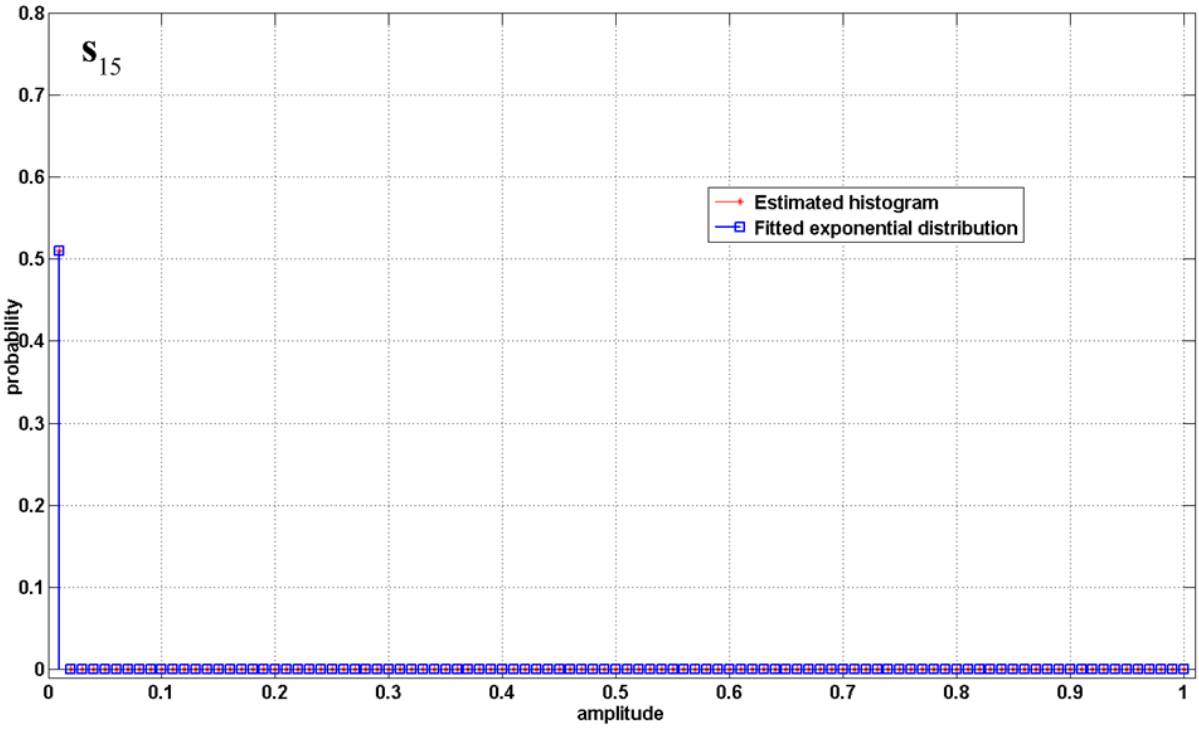

s10
Estimated histogram
Fitted exponential distribution
probability
amplitude
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

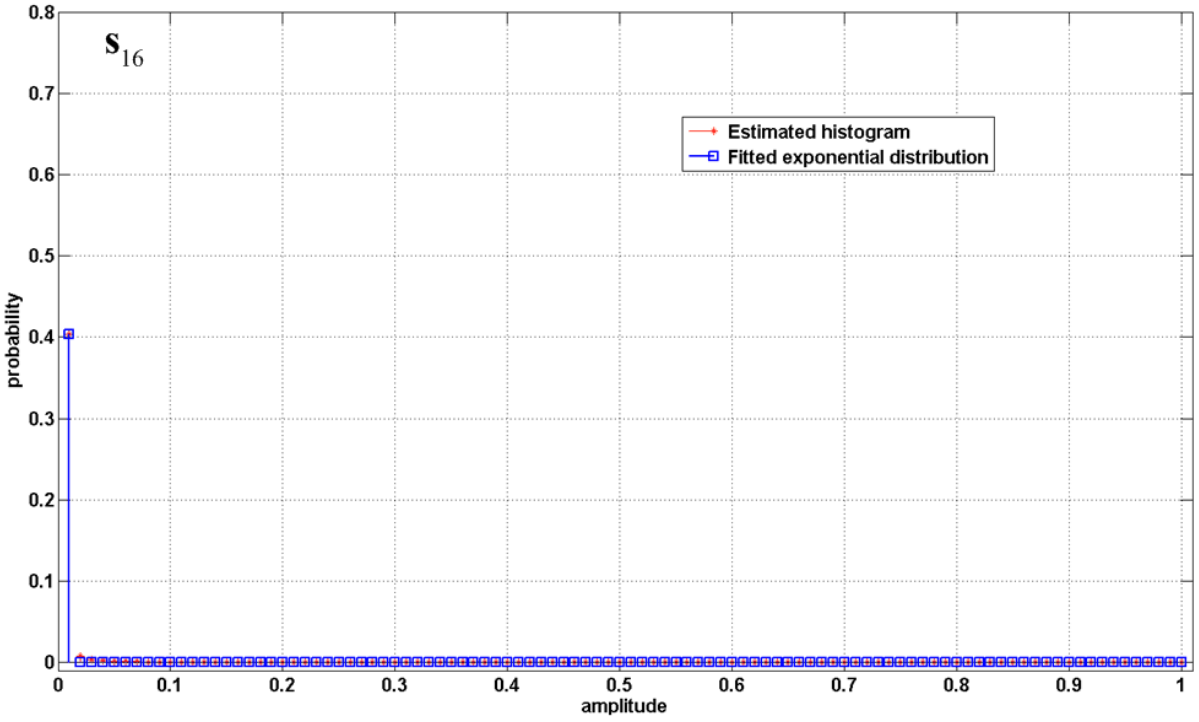

s_{11}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

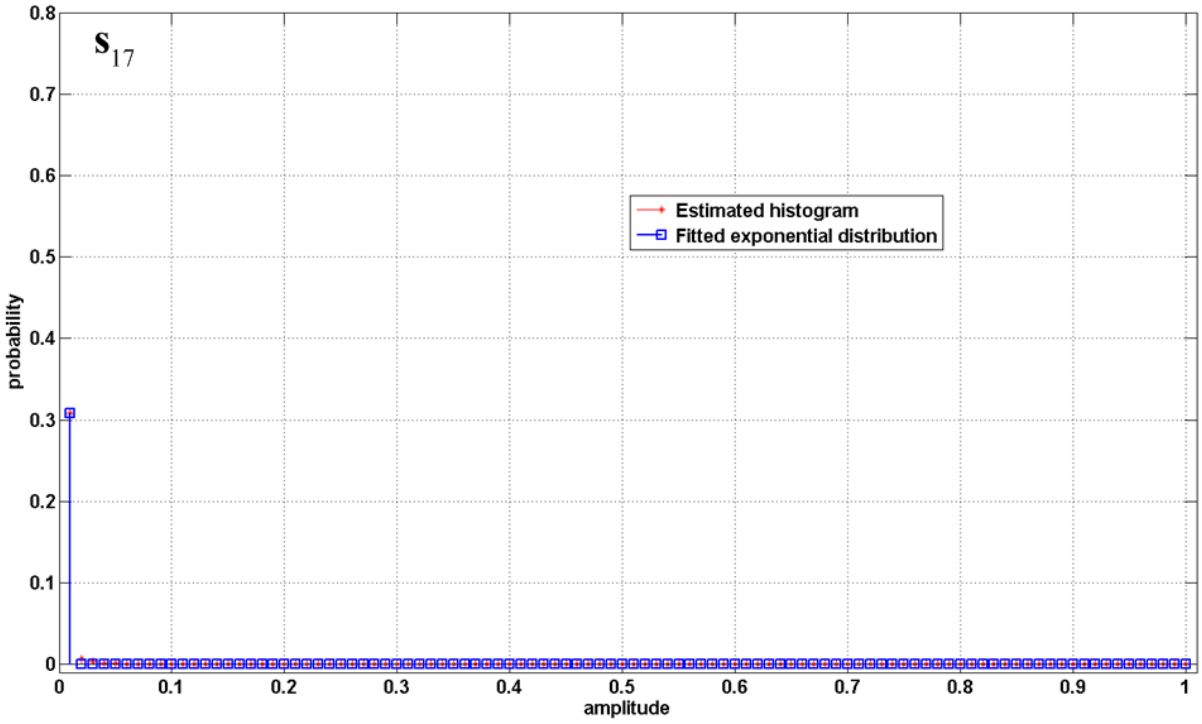

s_{12}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

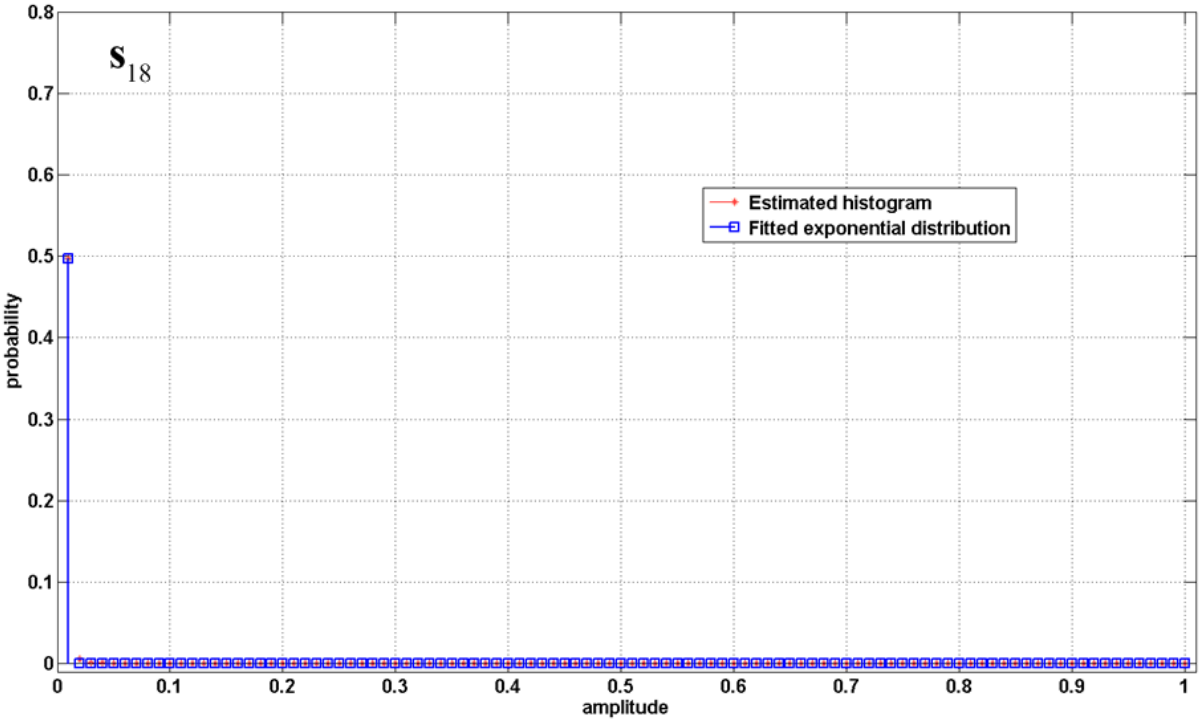

s13
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

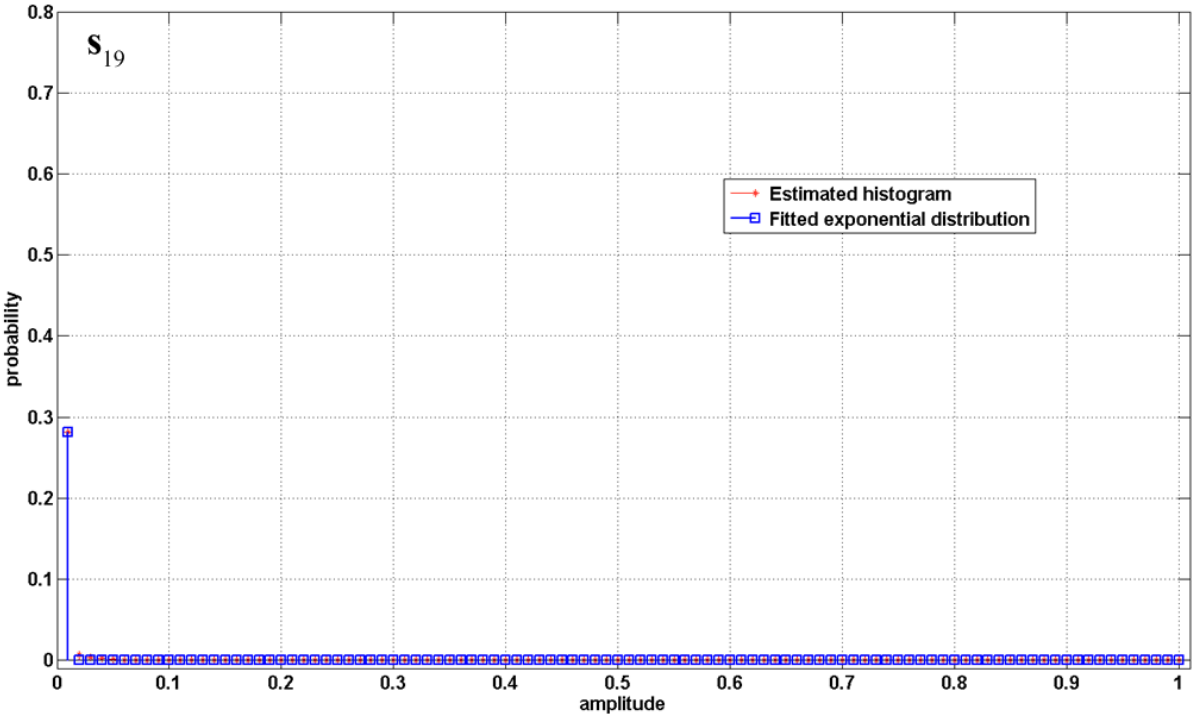

s14
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

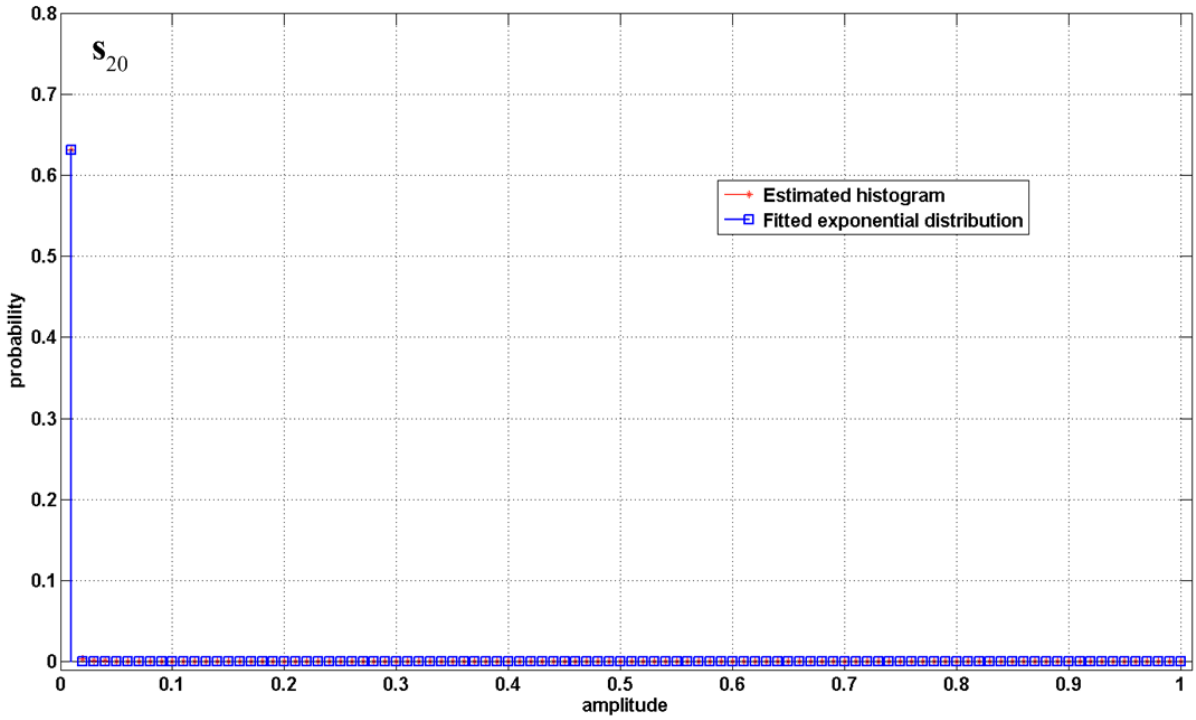

s_{15}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

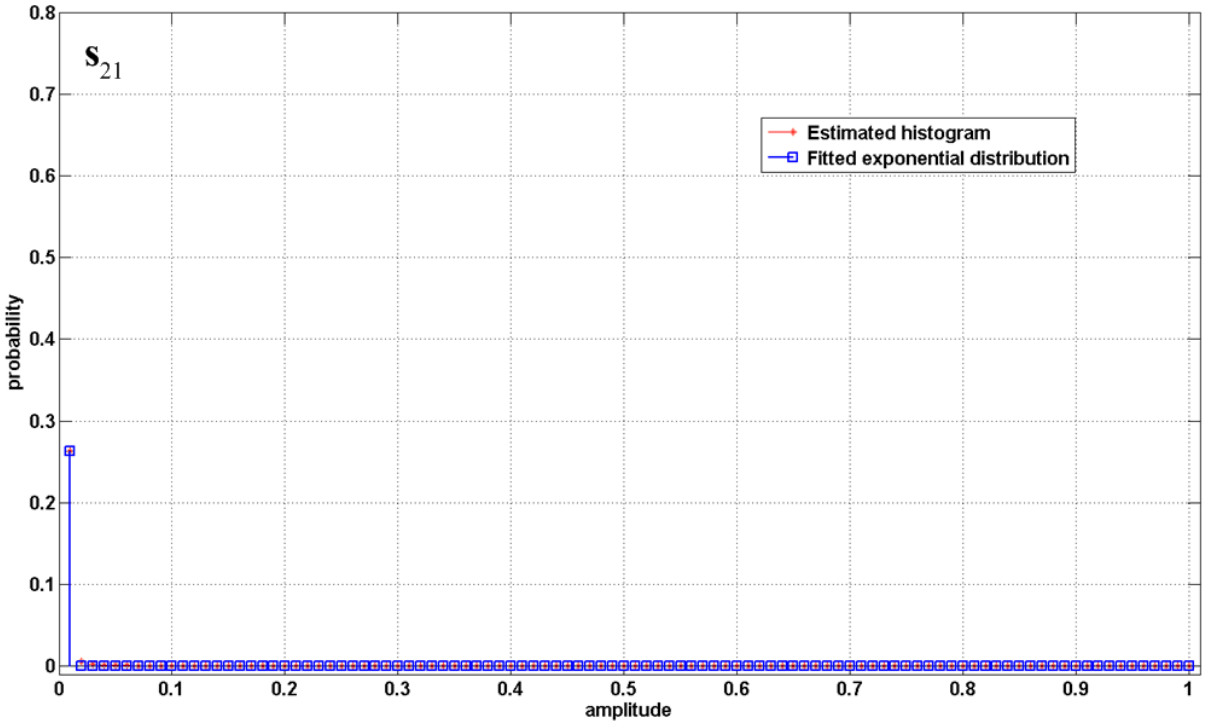

s_{16}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

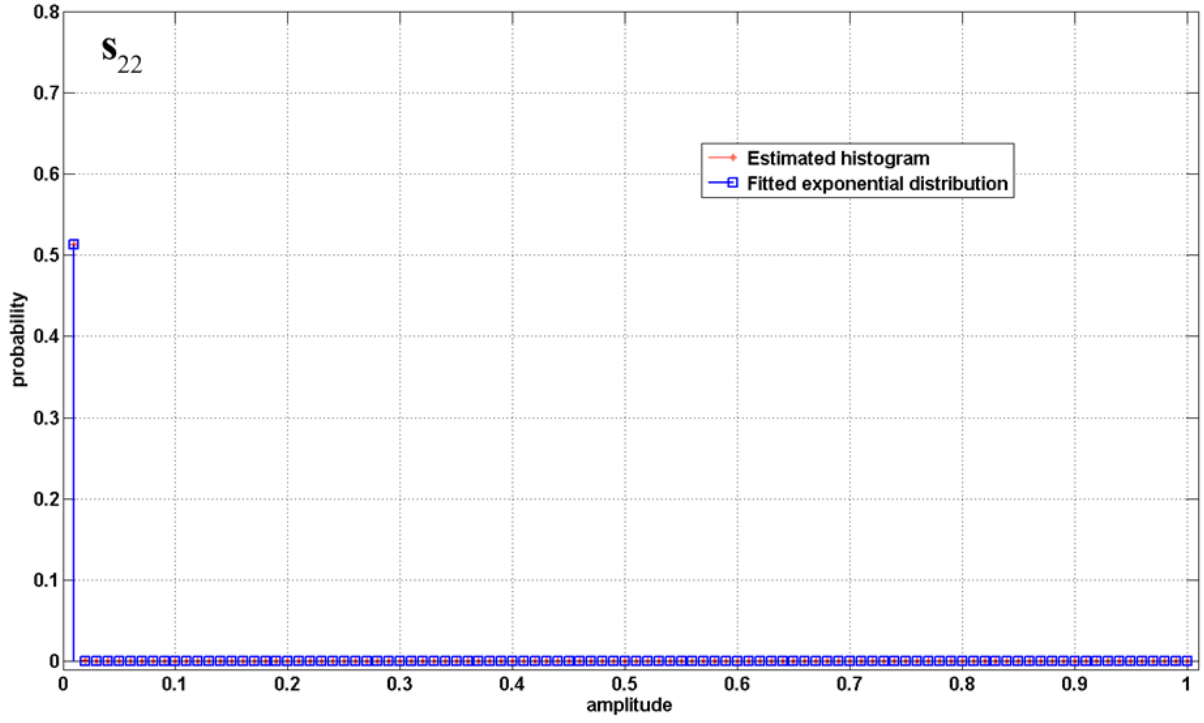

s_{17}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

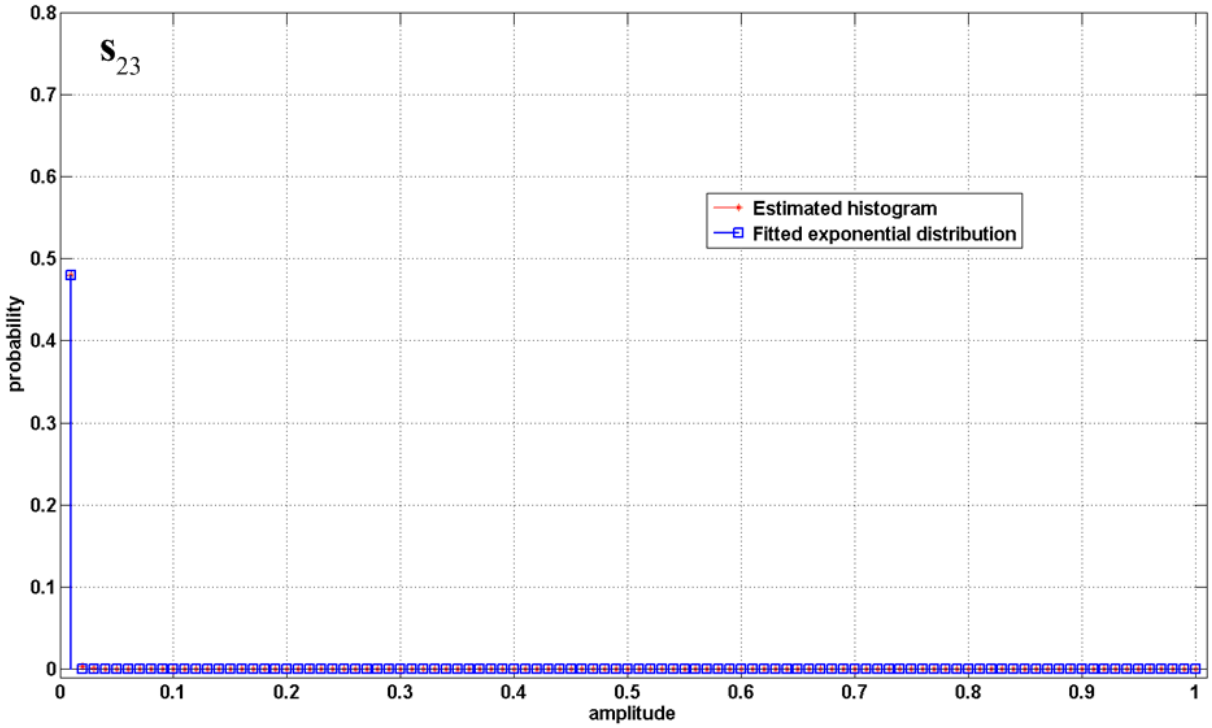

s_{18}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

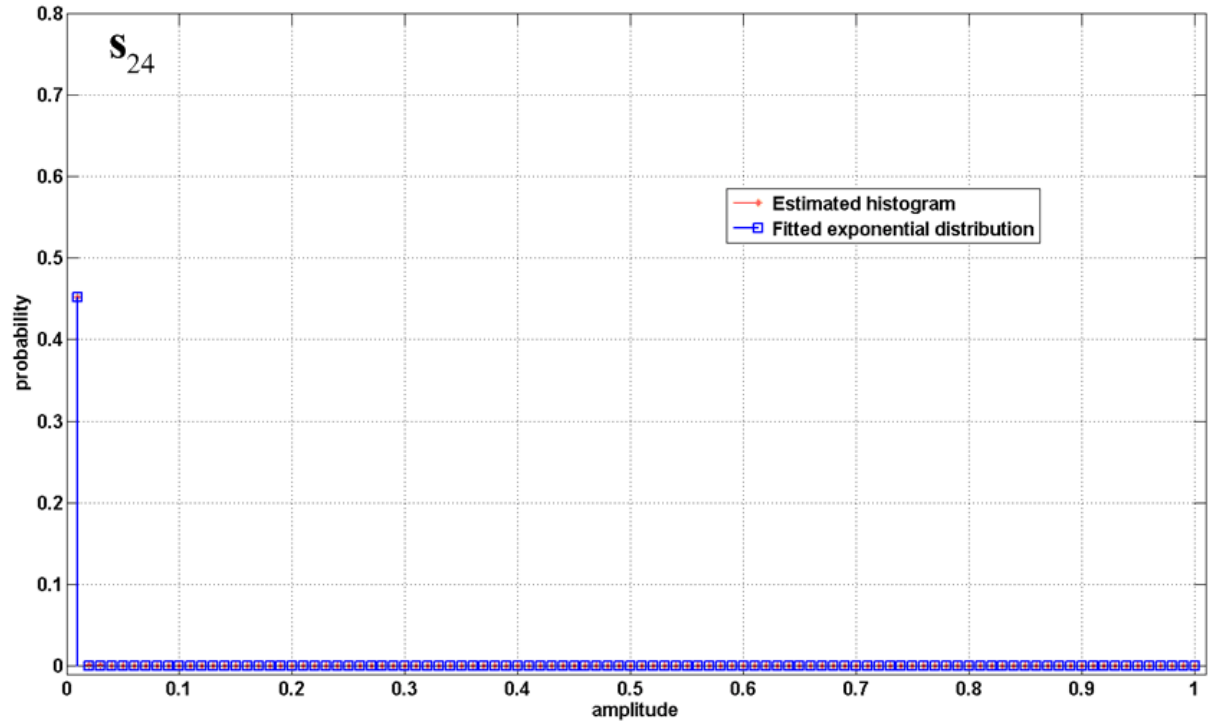

s_{19}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1


s_{20}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1


S21
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1


S22
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1


s_{23}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1


s_{24}
Estimated histogram
Fitted exponential distribution
probability
amplitude
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

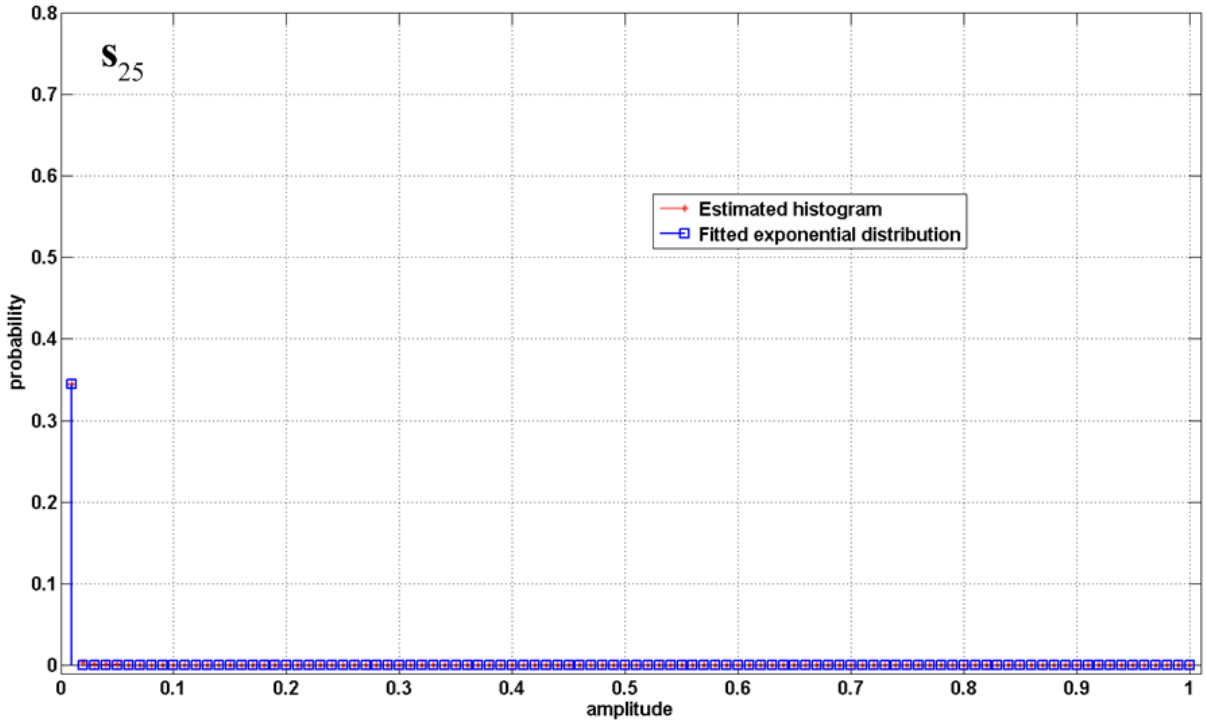
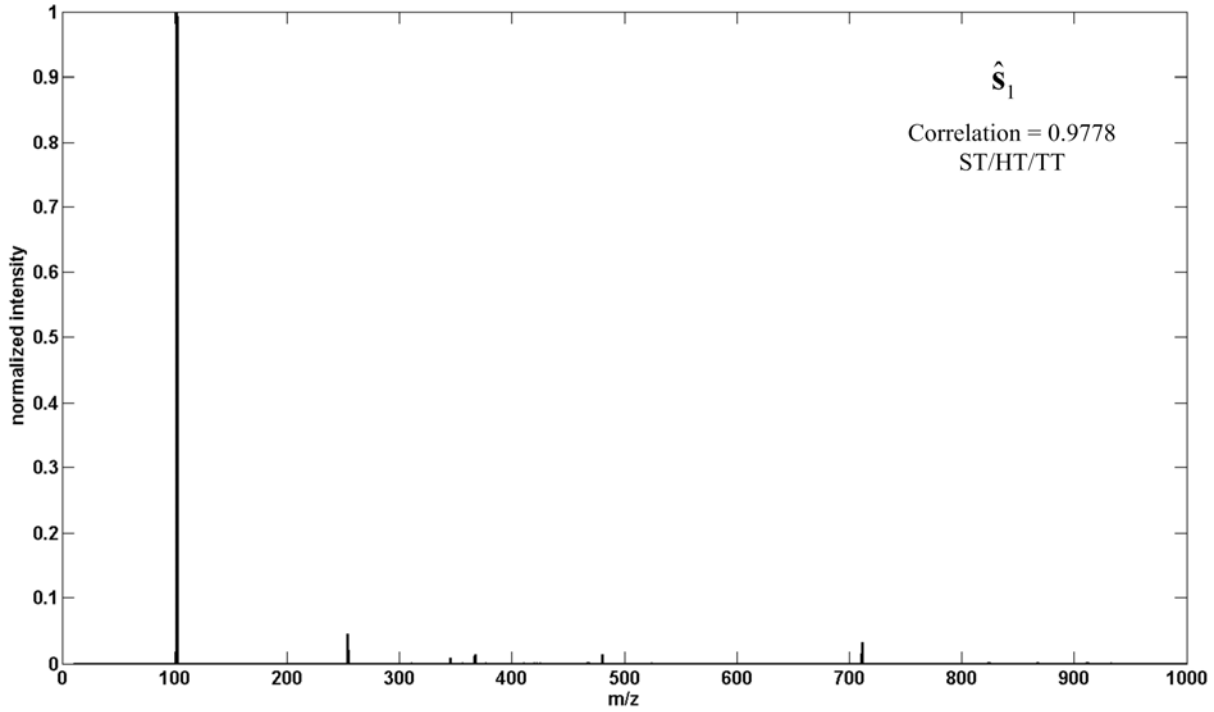
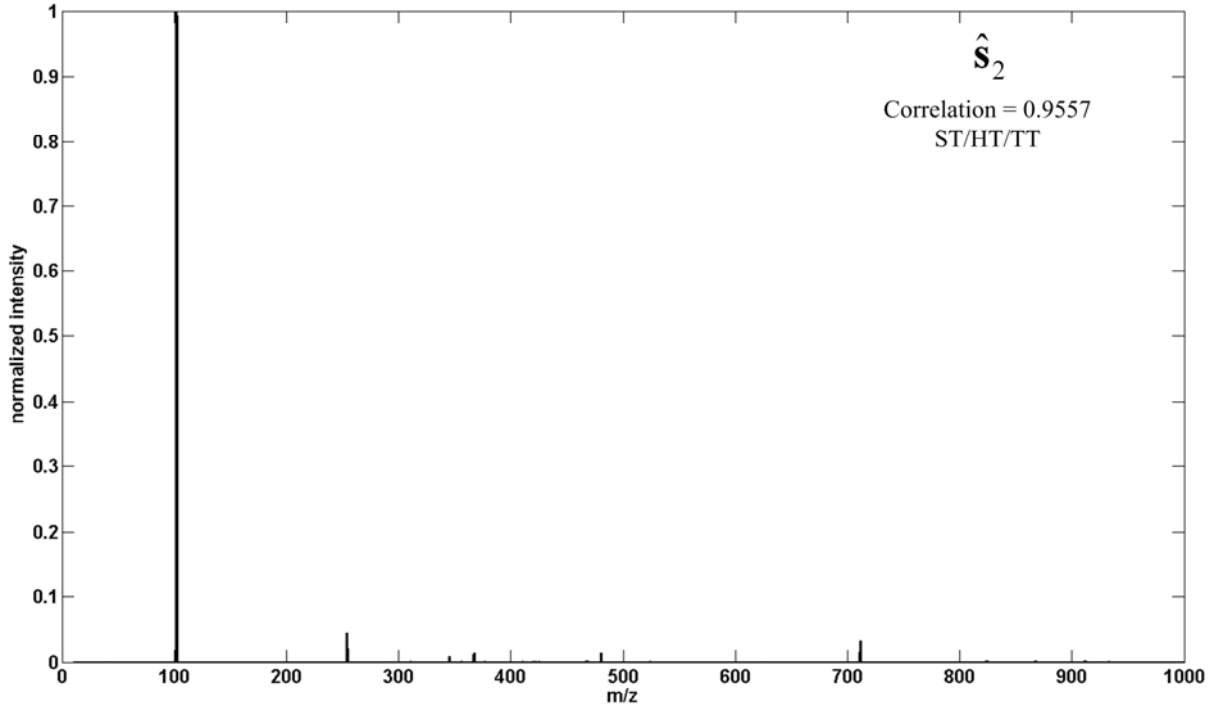
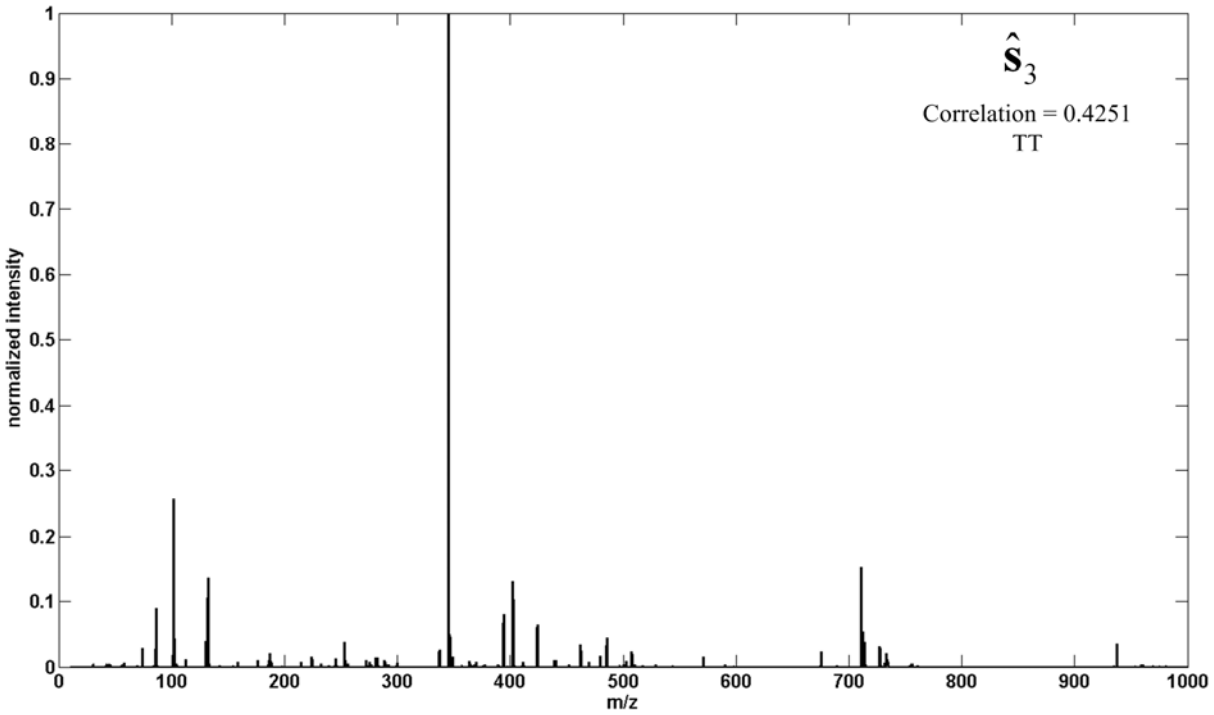
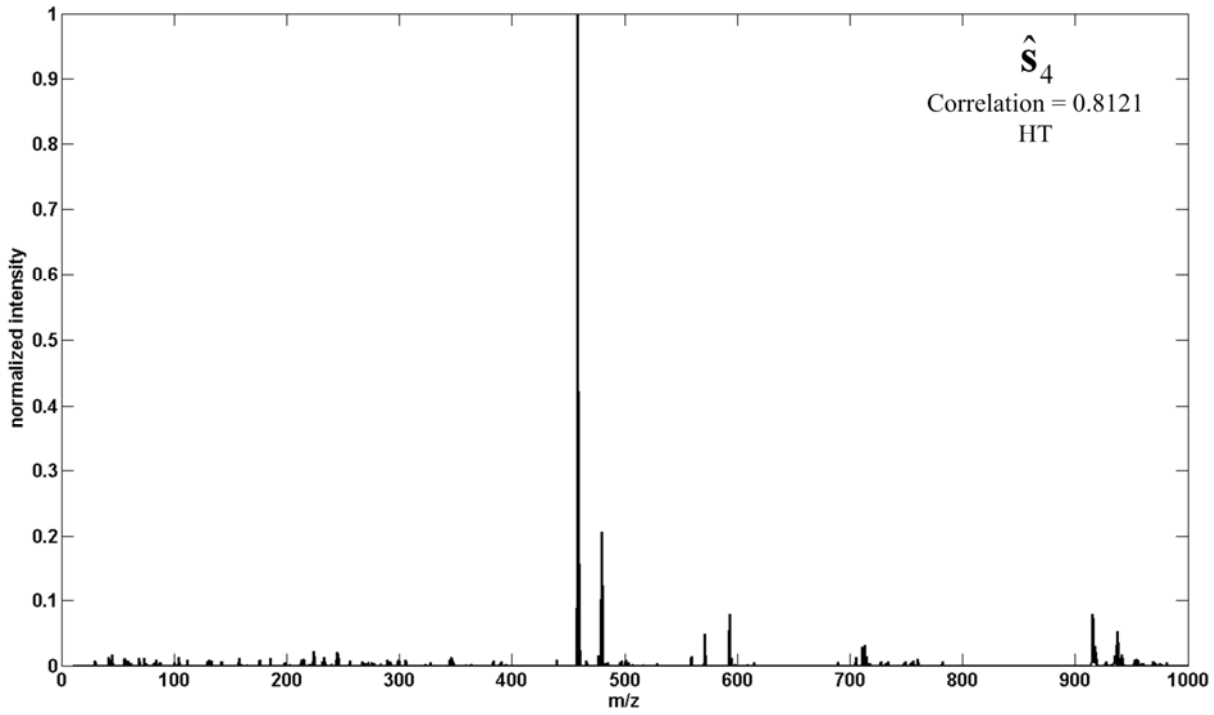

S25
Estimated histogram
Fitted exponential distribution
probability
amplitude
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1

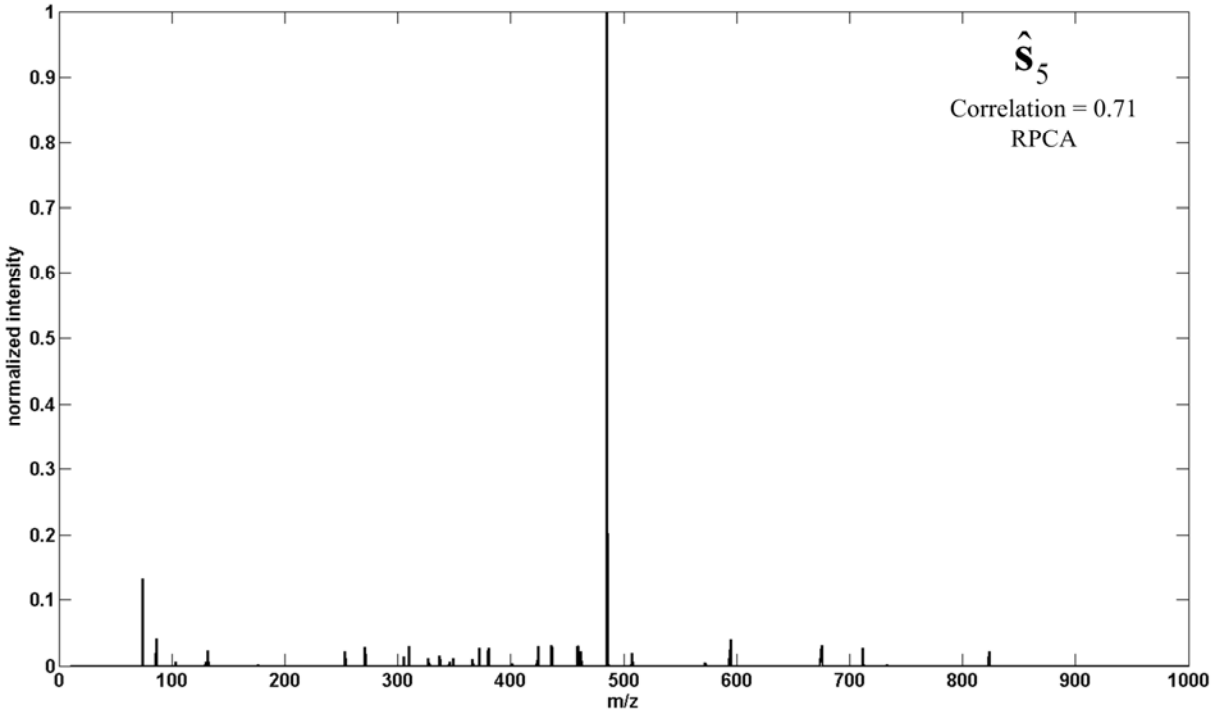
Figure S-8.

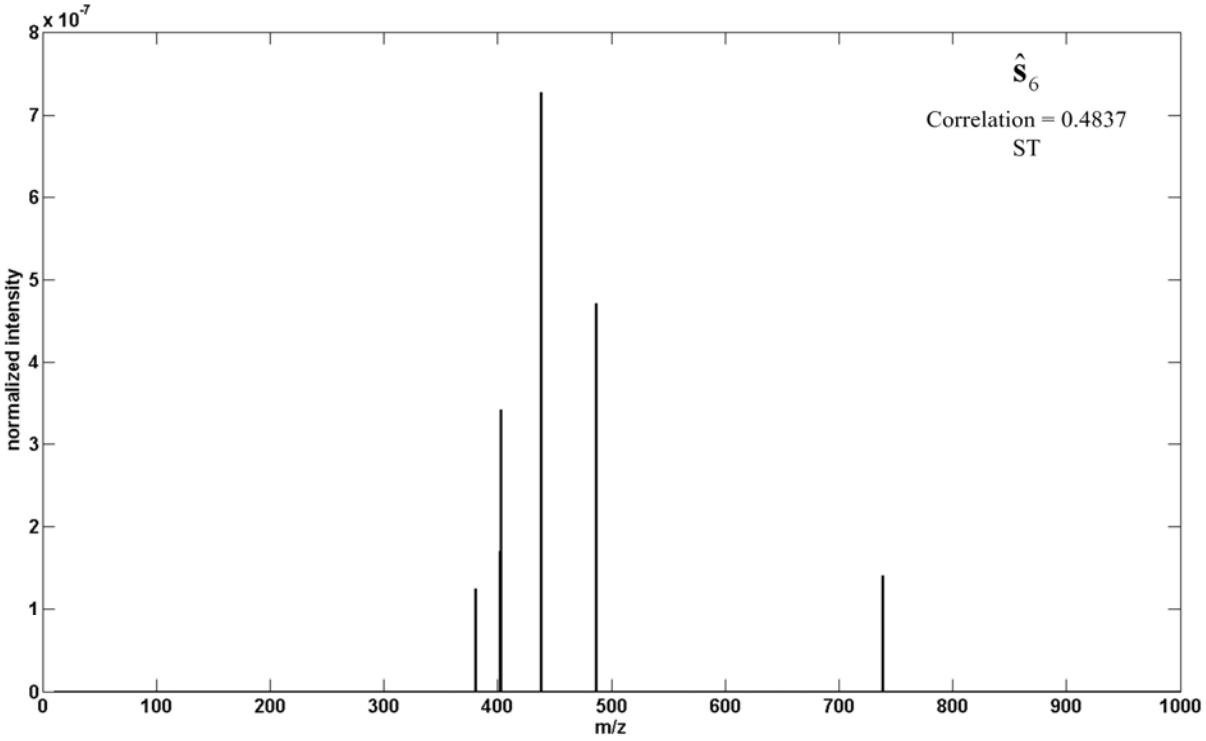

$\hat{\mathbf{s}}_1$
Correlation = 0.9778
ST/HT/TT
normalized intensity
m/z

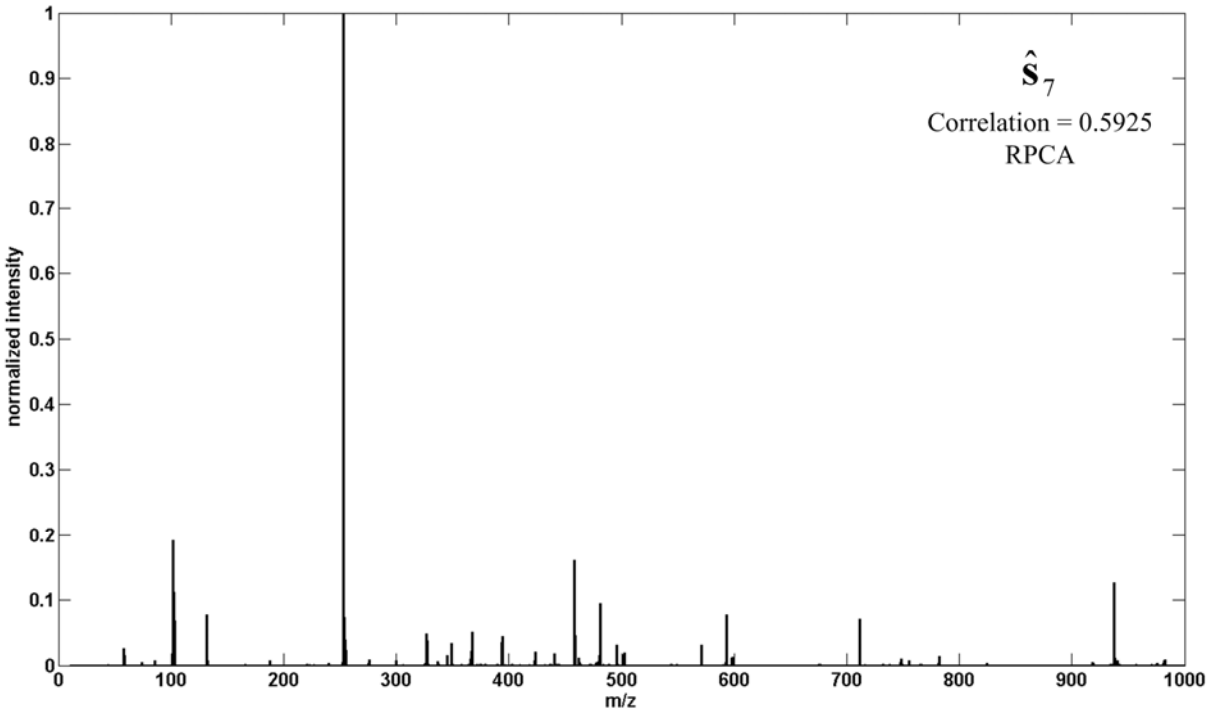

$\hat{\mathbf{s}}_2$
Correlation = 0.9557
ST/HT/TT
normalized intensity
m/z

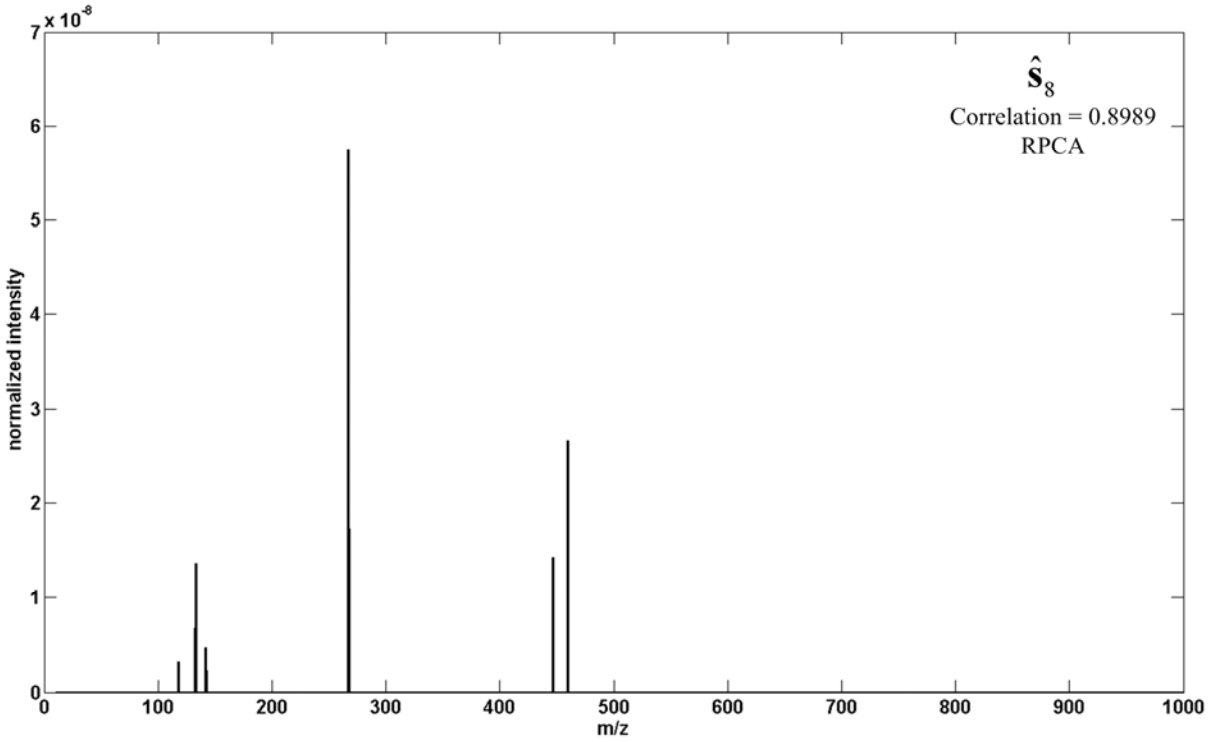

$\hat{\mathbf{s}}_3$
Correlation = 0.4251
TT
normalized intensity
m/z

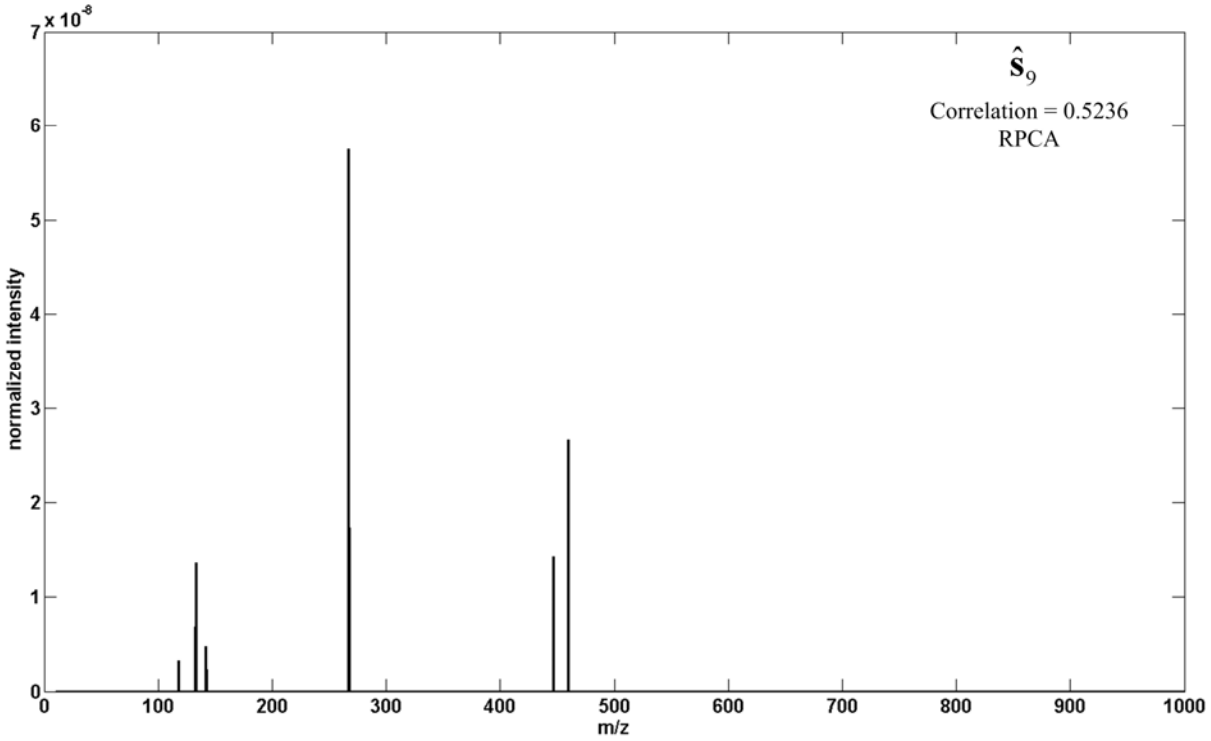

$\hat{\mathbf{s}}_4$
Correlation = 0.8121
HT
normalized intensity
m/z

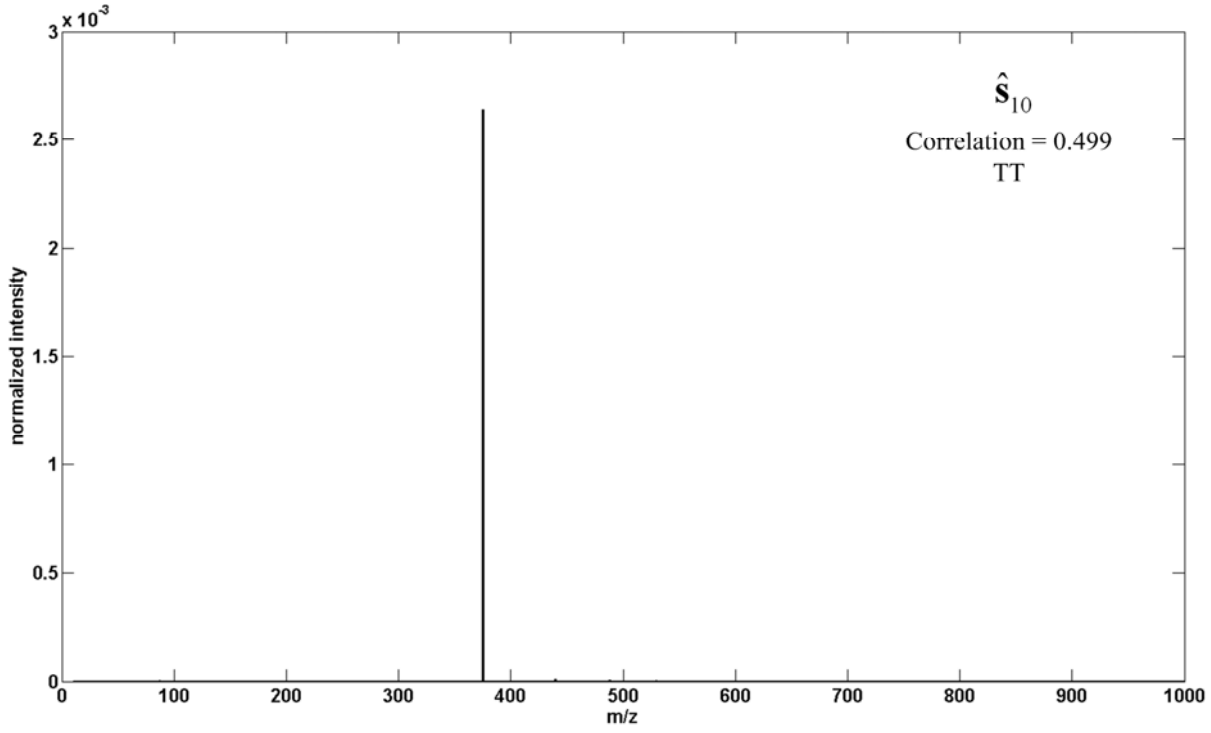

$\hat{\mathbf{s}}_5$
Correlation = 0.71
RPCA
normalized intensity
m/z

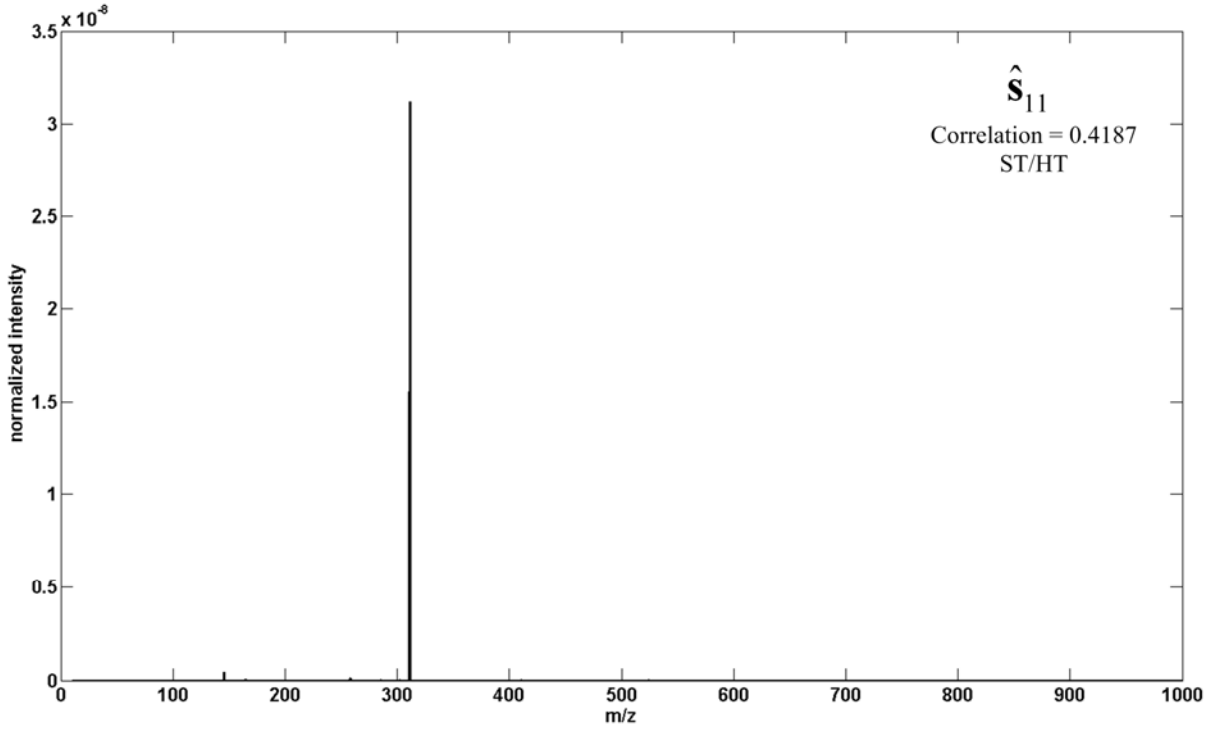

x 10^-7
$\hat{\mathbf{s}}_6$
Correlation = 0.4837
ST
normalized intensity
m/z

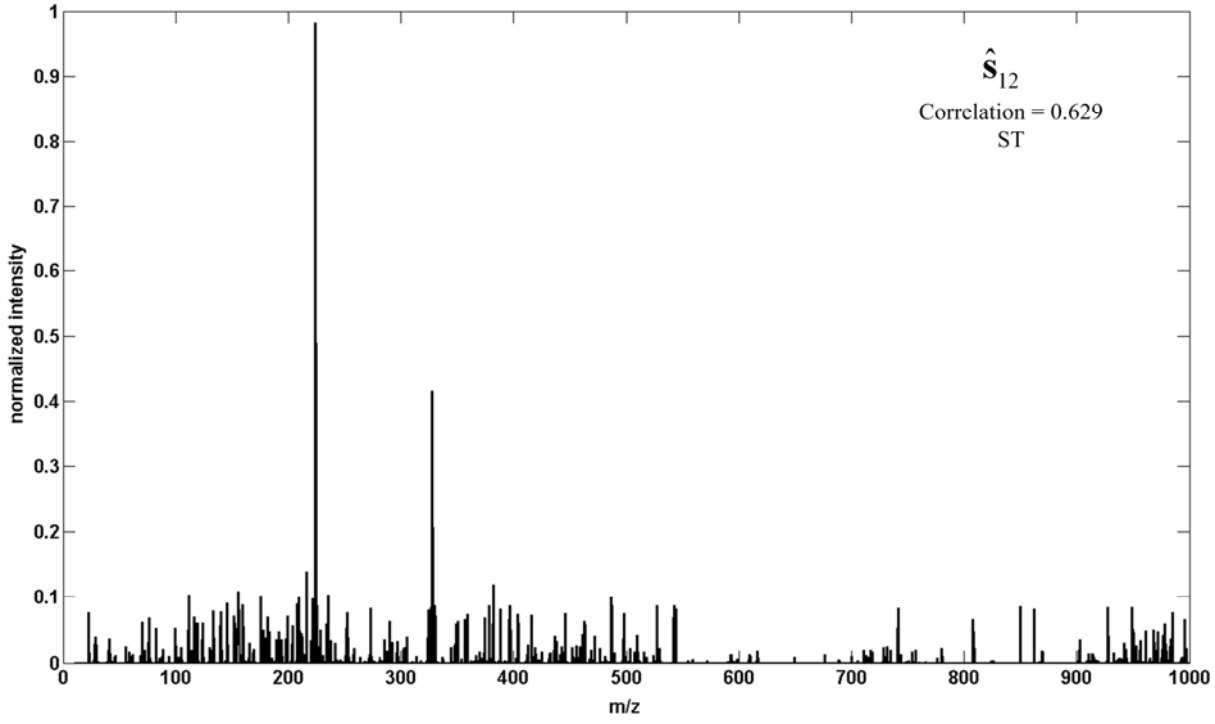

$\hat{\mathbf{s}}_7$
Correlation = 0.5925
RPCA
normalized intensity
m/z

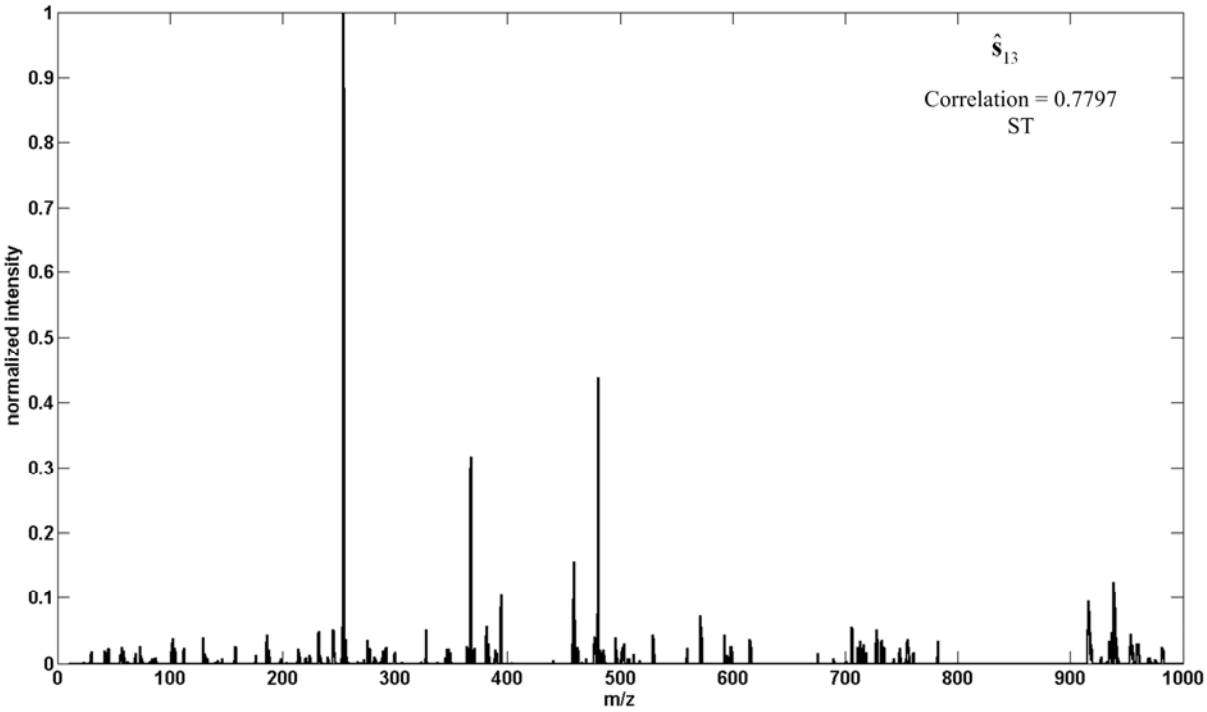

x 10^-8
$\hat{\mathbf{s}}_8$
Correlation = 0.8989
RPCA
normalized intensity
m/z

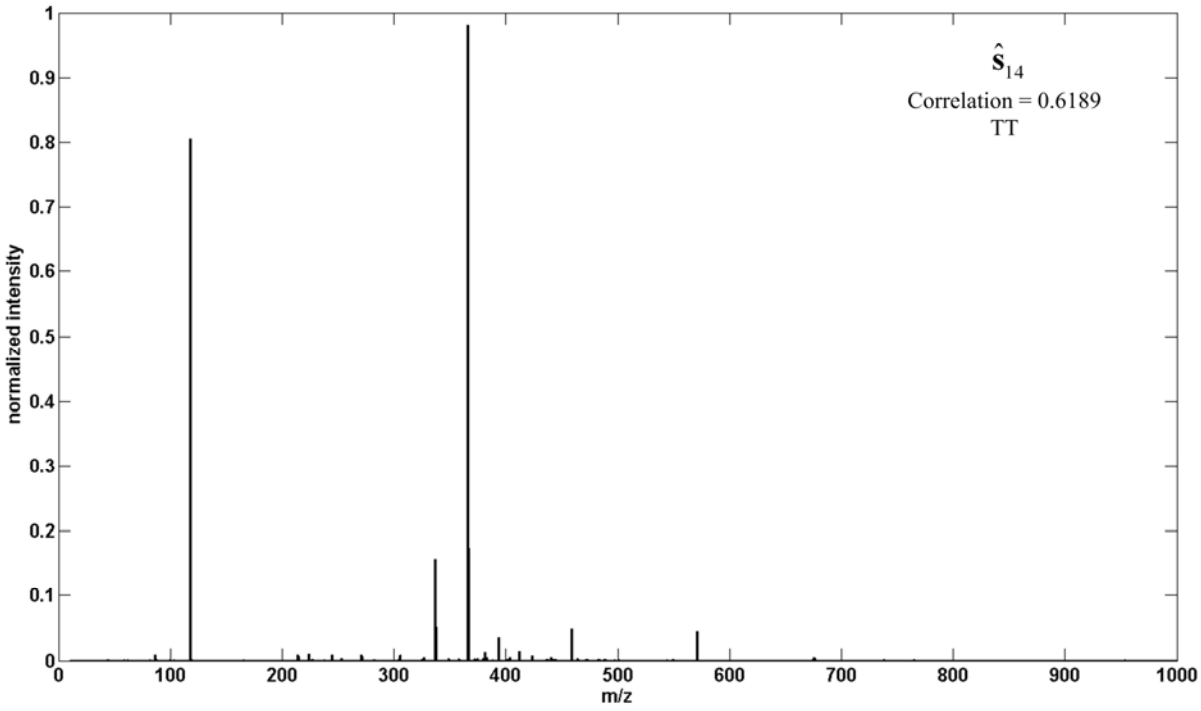

x 10^-8
7
6
5
4
3
2
1
0
normalized intensity
0
100
200
300
400
500
600
700
800
900
1000
m/z
$\hat{\mathbf{s}}_9$
Correlation = 0.5236
RPCA

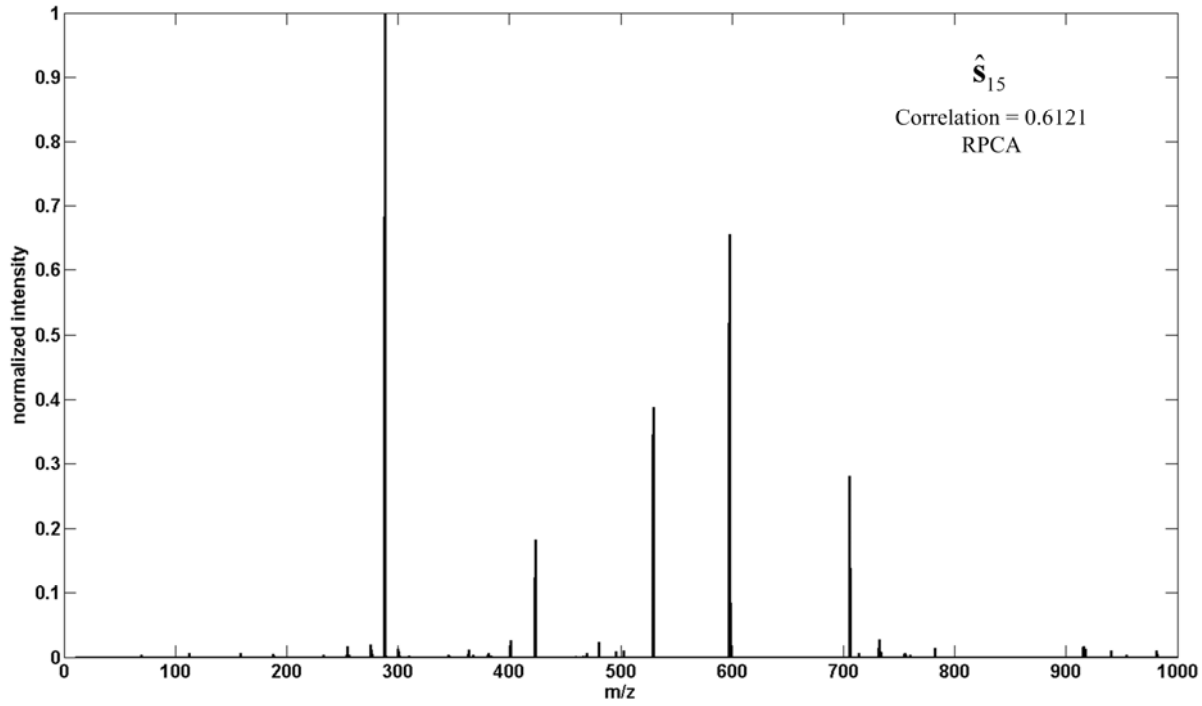

x 10^-3
3
2.5
2
1.5
1
0.5
0
normalized intensity
0
100
200
300
400
500
600
700
800
900
1000
m/z
$\hat{\mathbf{s}}_{10}$
Correlation = 0.499
TT

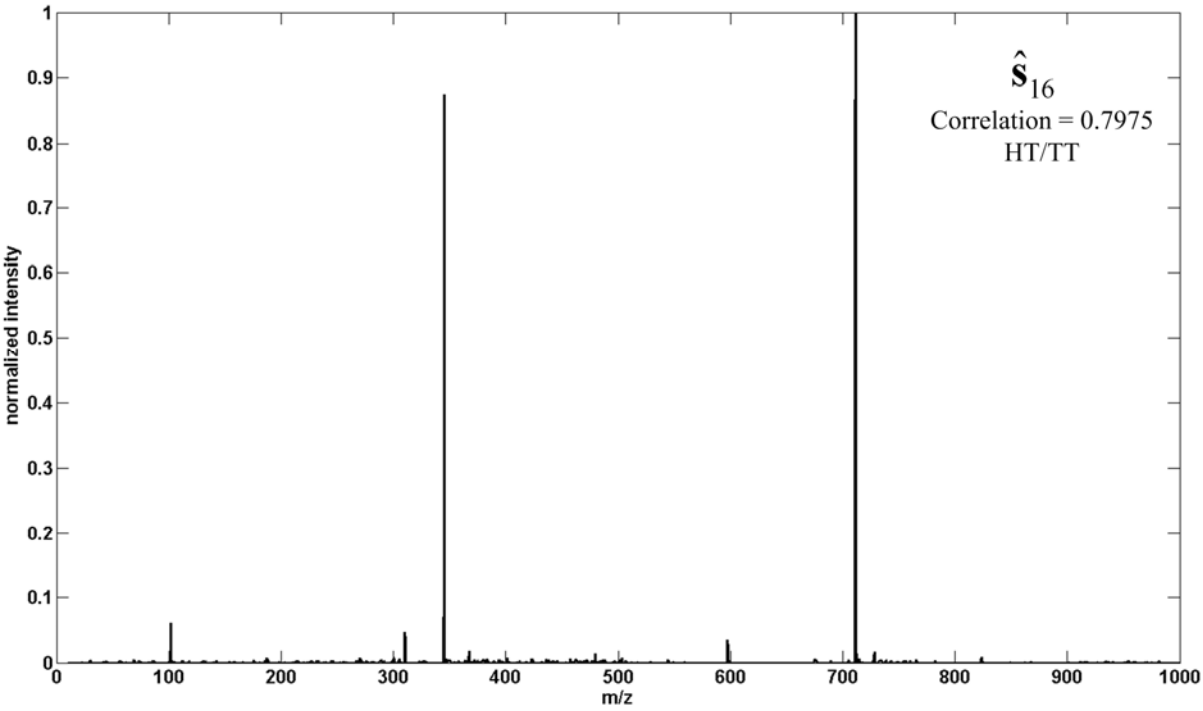

x 10^-8
3.5
3
2.5
2
1.5
1
0.5
0
normalized intensity
0
100
200
300
400
500
600
700
800
900
1000
m/z
ŝ11
Correlation = 0.4187
ST/HT

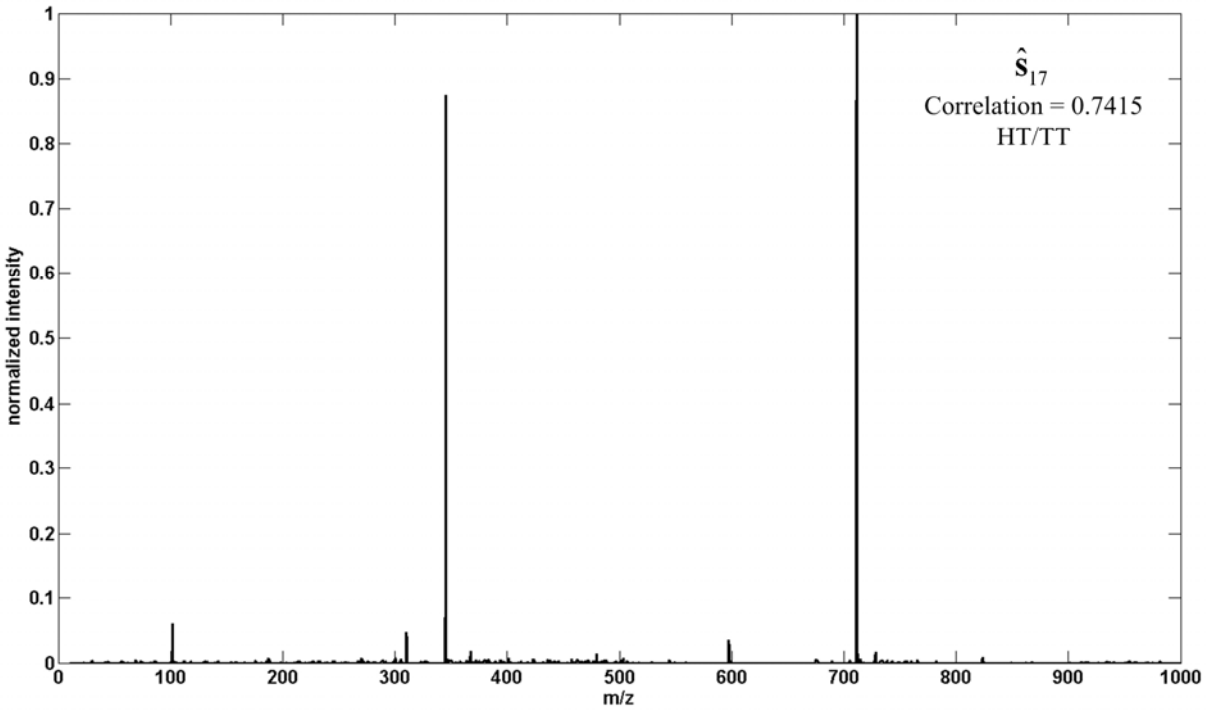

1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
normalized intensity
0
100
200
300
400
500
600
700
800
900
1000
m/z
ŝ12
Correlation = 0.629
ST

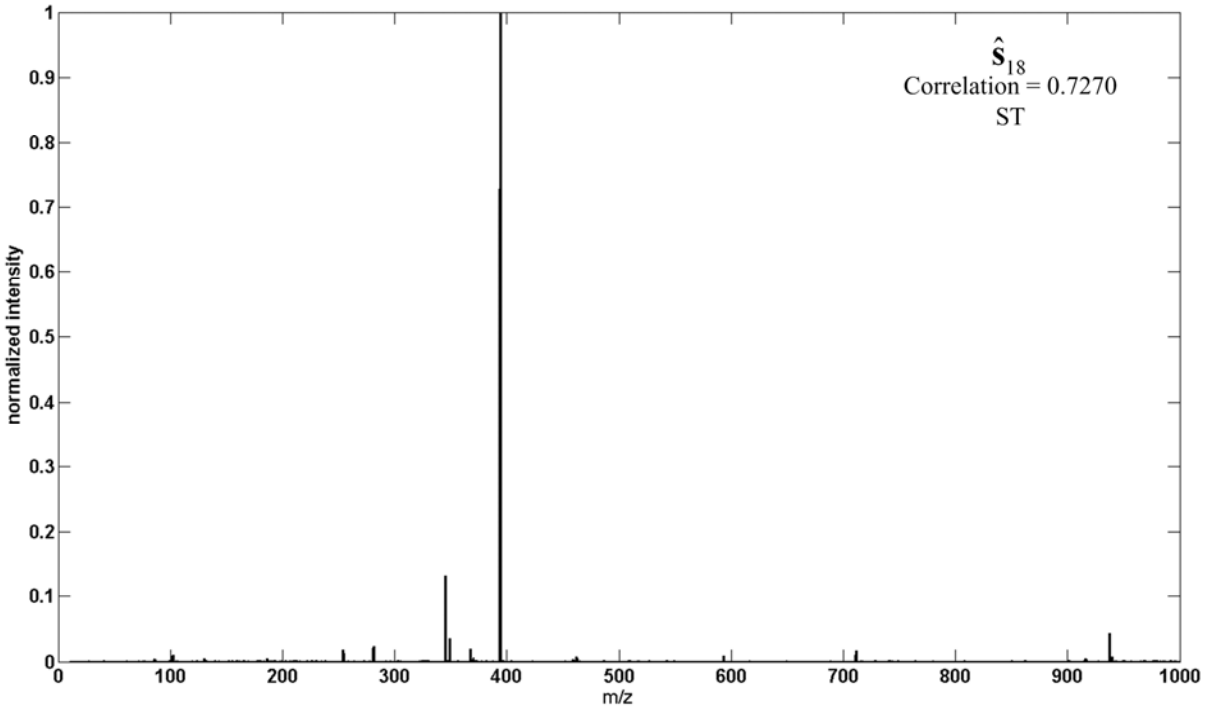

$\hat{\mathbf{s}}_{13}$
Correlation = 0.7797
ST
normalized intensity
m/z

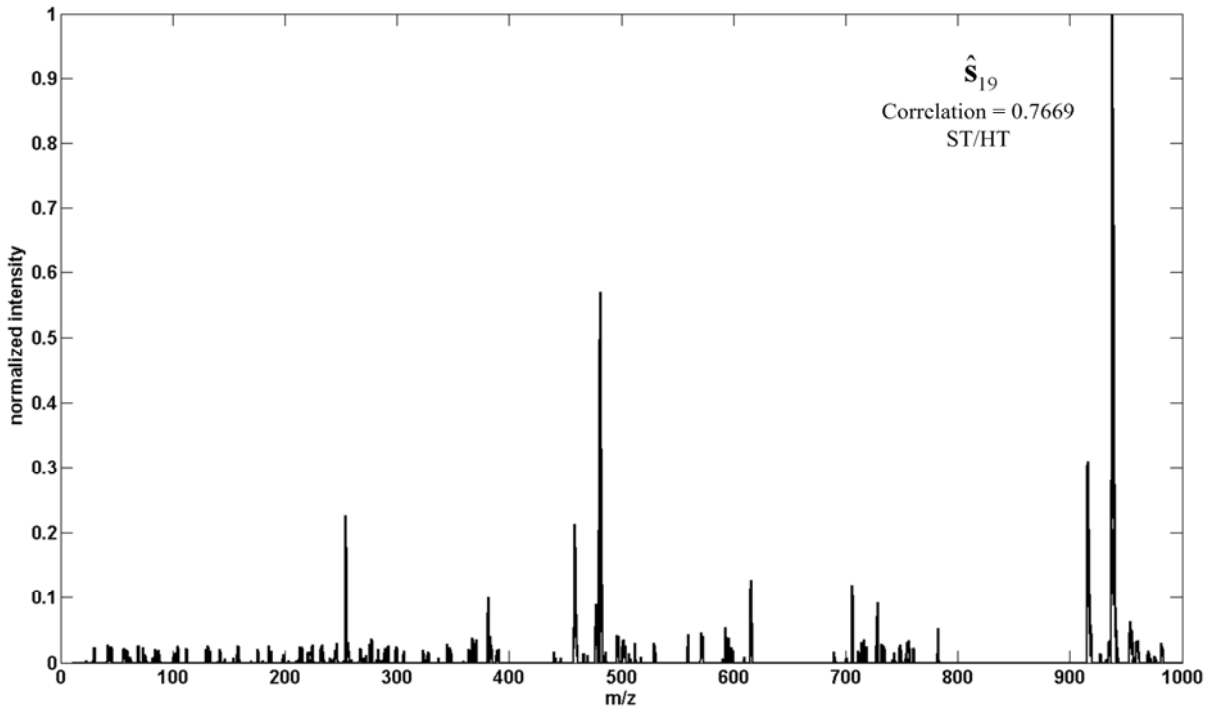

$\hat{\mathbf{s}}_{14}$
Correlation = 0.6189
TT
normalized intensity
m/z

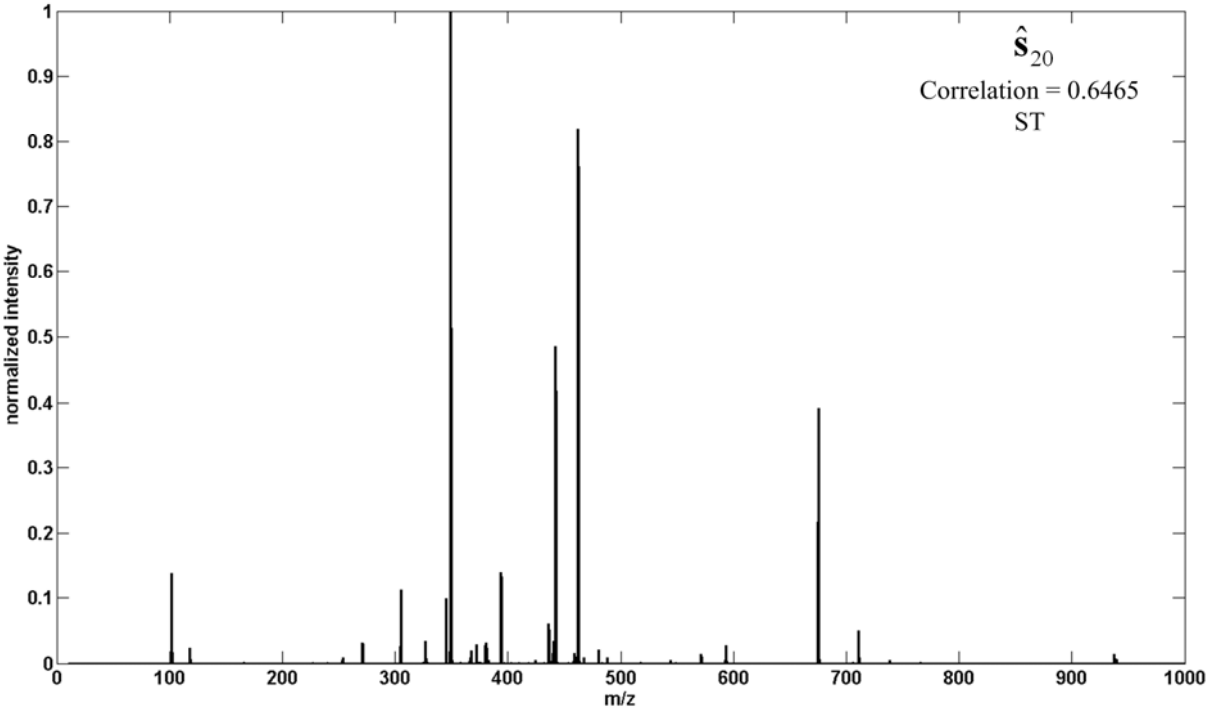

$\hat{\mathbf{s}}_{15}$
Correlation = 0.6121
RPCA
normalized intensity
m/z

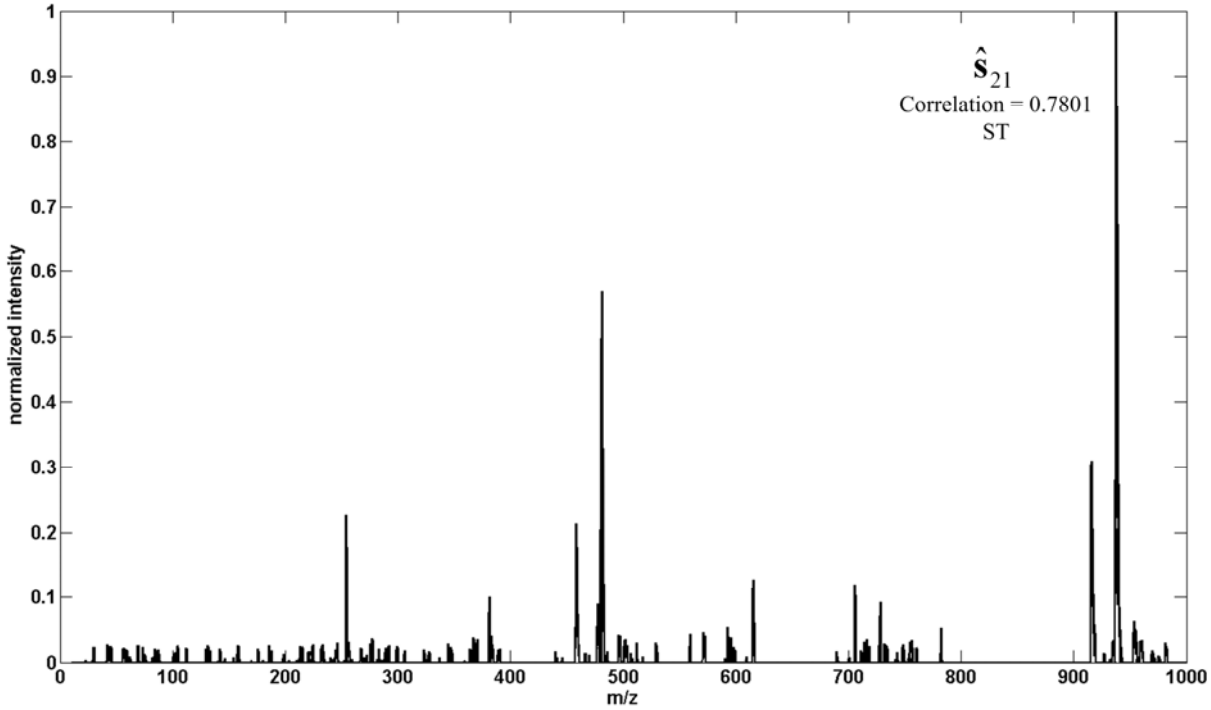

$\hat{\mathbf{s}}_{16}$
Correlation = 0.7975
HT/TT
normalized intensity
m/z

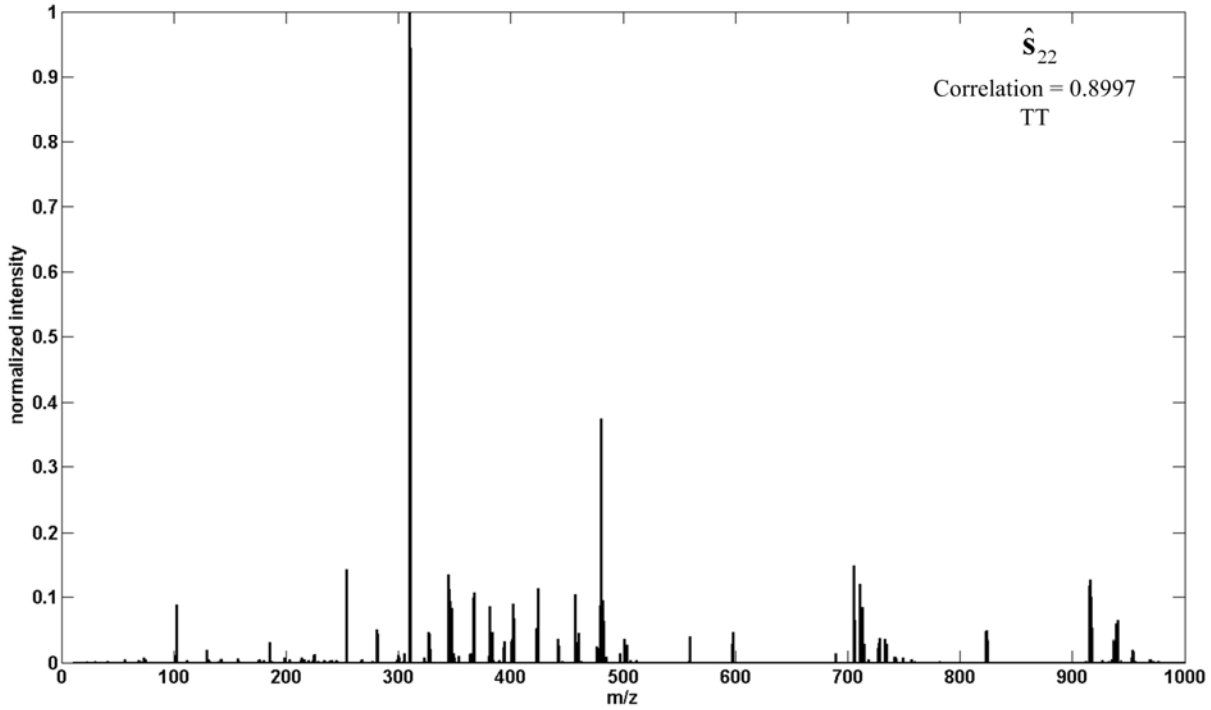

$\hat{\mathbf{s}}_{17}$
Correlation = 0.7415
HT/TT
normalized intensity
m/z

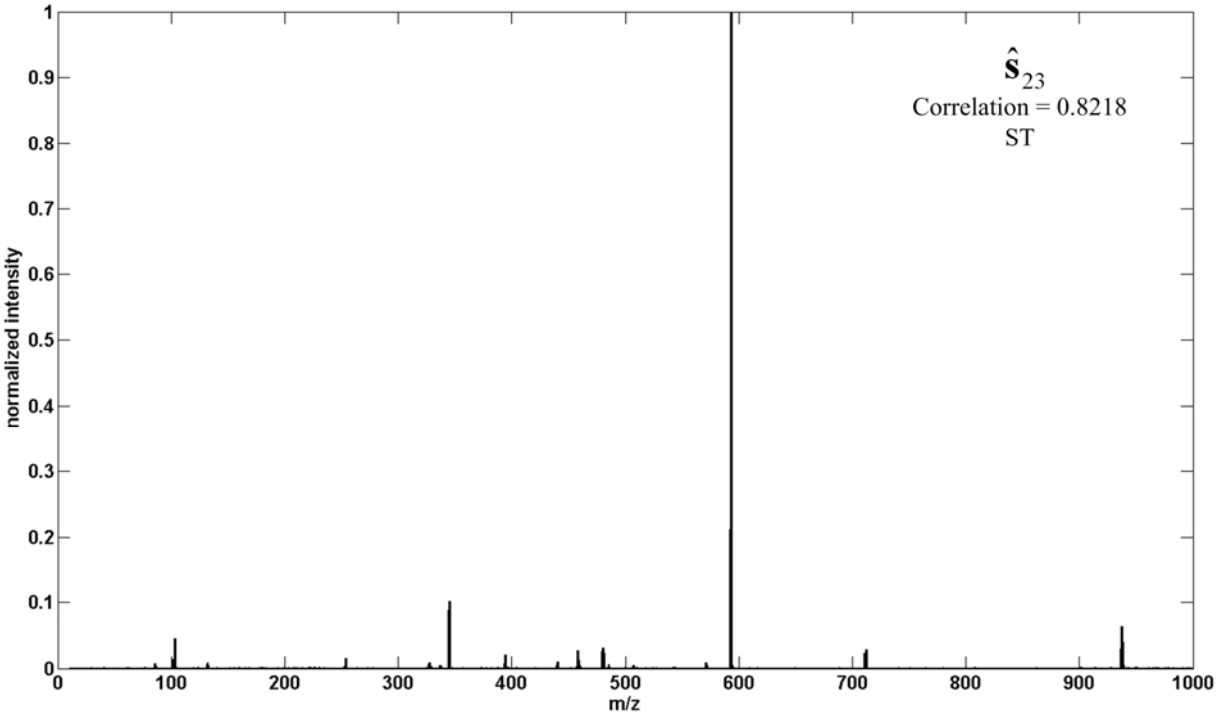

$\hat{\mathbf{s}}_{18}$
Correlation = 0.7270
ST
normalized intensity
m/z

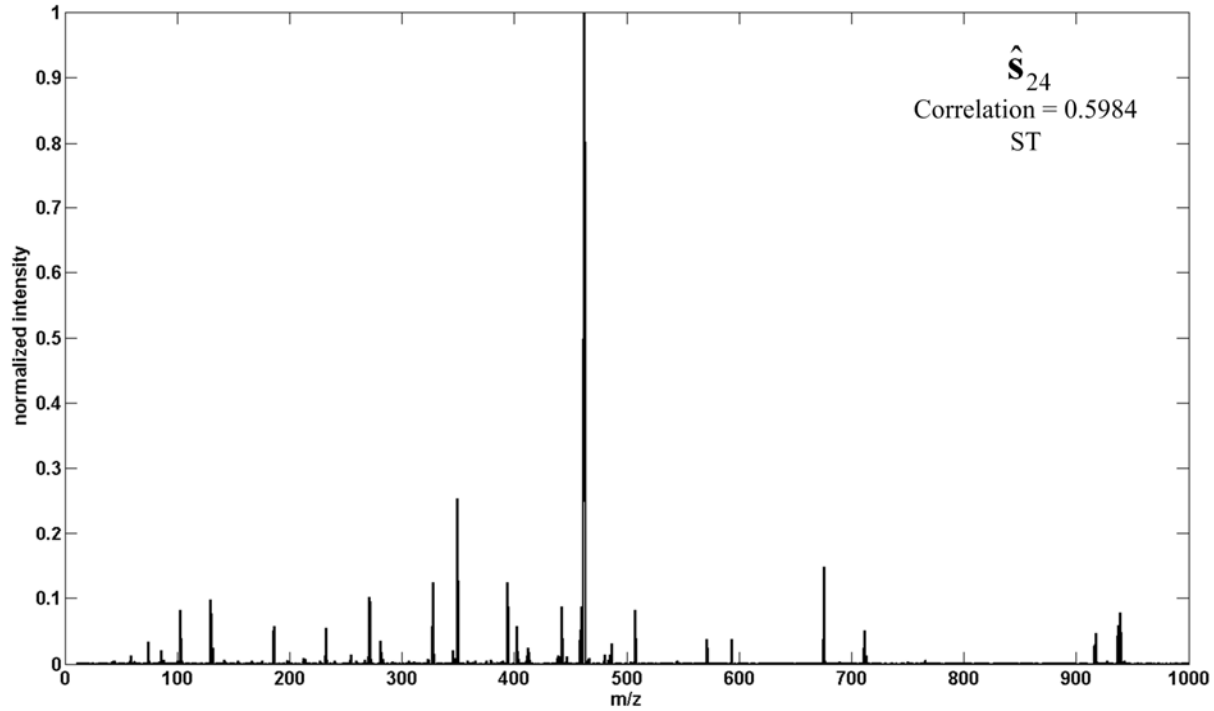

$\hat{\mathbf{s}}_{19}$
Correlation = 0.7669
ST/HT
normalized intensity
m/z


$\hat{\mathbf{s}}_{20}$
Correlation = 0.6465
ST
normalized intensity
m/z


$\hat{\mathbf{s}}_{21}$
Correlation = 0.7801
ST
normalized intensity
m/z


$\hat{\mathbf{s}}_{22}$
Correlation = 0.8997
TT
normalized intensity
m/z


$\hat{\mathbf{s}}_{23}$
Correlation = 0.8218
ST
normalized intensity
m/z


$\hat{\mathbf{s}}_{24}$
Correlation = 0.5984
ST
normalized intensity
m/z

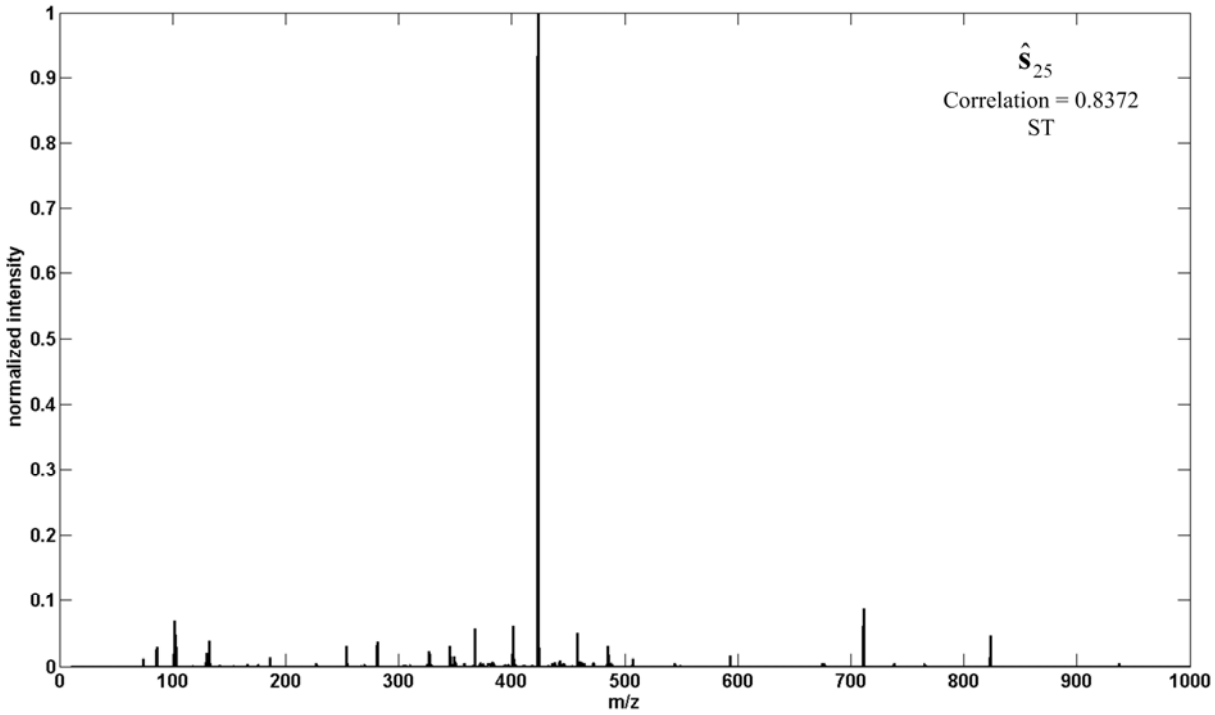

$\hat{\mathbf{s}}_{25}$
Correlation = 0.8372
ST
normalized intensity
m/z
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300
400
500
600
700
800
900
1000

Figure S-9.