Supporting Information

Discovery and identification of arsenolipids using a precursor-finder strategy and data-independent mass spectrometry

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**Table S1.** The information of arsenolipids that have been reported in previous literatures.

| Species | Name      | Molecular Mass | Formula                                      | Structure | Reference |
|---------|-----------|----------------|----------------------------------------------|-----------|-----------|
|         | AsSugPL692 | 693.26         | C_{27}H_{54}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL706 | 707.27         | C_{28}H_{56}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL720 | 721.29         | C_{29}H_{58}AsO_{13}P                         | ![Structure](image) | [9],[11]  |
|         | AsSugPL734 | 735.31         | C_{30}H_{60}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL742 | 743.27         | C_{31}H_{62}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL746 | 747.31         | C_{31}H_{60}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL748 | 749.32         | C_{31}H_{62}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL776 | 777.35         | C_{31}H_{60}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL930 | 931.49         | C_{33}H_{66}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL944 | 945.50         | C_{34}H_{68}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL956 | 957.50         | C_{34}H_{68}AsO_{13}P                         | ![Structure](image) | [11]      |
|         | AsSugPL958 | 959.52         | C_{34}H_{70}AsO_{13}P                         | ![Structure](image) | [3],[9],[11],[17],[27] |
|         | AsSugPL972 | 973.54         | C_{35}H_{72}AsO_{13}P                         | ![Structure](image) | [17]      |
|         | AsSugPL982 | 983.52         | C_{35}H_{72}AsO_{13}P                         | ![Structure](image) | [13],[17],[27] |
|         | AsSugPL984 | 985.54         | C_{35}H_{72}AsO_{13}P                         | ![Structure](image) | [13],[17],[27] |
|         | AsSugPL986 | 987.55         | C_{35}H_{72}AsO_{13}P                         | ![Structure](image) | [3],[9],[17] |
|         | AsSugPL100 | 1001.57        | C_{36}H_{74}AsO_{13}P                         | ![Structure](image) | [17]      |
|         | AsSugPL101 | 1013.57        | C_{37}H_{76}AsO_{13}P                         | ![Structure](image) | [33]      |
|         | AsSugPL101 | 1015.58        | C_{38}H_{76}AsO_{13}P                         | ![Structure](image) | [3],[9],[17] |
|         | AsSugPL104 | 1043.61        | C_{39}H_{78}AsO_{13}P                         | ![Structure](image) | [3],[9],[33] |
|         | AsSugPL107 | 1071.64        | C_{40}H_{80}AsO_{13}P                         | ![Structure](image) | [3],[9],[33] |
| AsHCs   | AsHC164    | 165.03         | C_{17}H_{35}AsO                              | ![Structure](image) | [8]       |
|         | AsHC305    | 305.18         | C_{18}H_{35}AsO                              | ![Structure](image) | [5],[32]  |
|         | AsHC331    | 331.20         | C_{19}H_{35}AsO                              | ![Structure](image) | [5],[22]  |
| Compound   | Mass   | Formula   | References |
|------------|--------|-----------|------------|
| AsHC332    | 333.21 | C_{17}H_{37}AsO | [1],[2],[3], [4],[5],[6],[7], [8],[9],[10],[11],[14],[15],[24] |
| AsHC347    | 347.23 | C_{18}H_{39}AsO | [5],[22] |
| AsHC358    | 359.23 | C_{19}H_{41}AsO | [5],[9] |
| AsHC360    | 361.24 | C_{19}H_{41}AsO | [1],[3],[4],[5],[6],[8],[9],[10],[14],[15],[26],[30] |
| Thio-AsHC360 | 377.22 | C_{19}H_{41}AsS | [10],[14], [20] |
| AsHC374    | 375.26 | C_{20}H_{40}AsO | [17] |
| AsHC386    | 387.26 | C_{21}H_{40}AsO | [24] |
| AsHC388    | 389.28 | C_{21}H_{40}AsO | [9],[17] |
| Thio-AsHC388 | 397.29 | C_{21}H_{40}AsS | [20] |
| AsHC402    | 403.20 | C_{23}H_{50}AsO | [28] |
| AsHC404    | 405.21 | C_{23}H_{50}AsO | [5],[6],[8],[15],[17],[20],[21],[29] |
| AsHC440    | 441.31 | C_{25}H_{52}AsO | [20] |
| AsHC442    | 442.32 | C_{25}H_{52}AsO | [20] |
| AsHC444    | 445.34 | C_{25}H_{52}AsO | [4],[14],[18],[25] |
| Thio-AsHC444 | 453.35 | C_{25}H_{52}AsS | [14],[20] |
| AsHC542    | 543.35 | C_{33}H_{83}AsO | [20] |
| AsFA264    | 265.08 | C_{10}H_{22}AsO | [28],[30] |
| AsFA276    | 277.08 | C_{11}H_{22}AsO | [28],[30] |
| AsFA278    | 279.09 | C_{11}H_{22}AsO | [7],[28] |
| AsFA278    | 279.09 | C_{11}H_{22}AsO | [7],[28] |
| thio-AsFA278 | 295.06 | C_{11}H_{22}AsO | [7] |
| AsFA302    | 303.09 | C_{13}H_{24}AsO | [28],[30] |
| AsFA304    | 305.11 | C_{13}H_{24}AsO | [30] |
| AsFA306    | 307.12 | C_{13}H_{24}AsO | [7] |
| thio-AsFA306 | 323.10 | C_{13}H_{24}AsO | [7] |
| Compound |分子式 | 分子量 |参考文献 |
|----------|--------|--------|---------|
| AsFA316 | C_{14}H_{26}AsO_{3} | [28],[30] |
| AsFA328 | C_{14}H_{26}AsO_{3} | [28],[30] |
| AsFA334 | C_{15}H_{31}AsO_{3} | [11],[17],[8],[10],[28],[32] |
| thio-AsFA334 | C_{14}H_{26}AsO_{3}S | [7] |
| AsFA342 | C_{16}H_{32}AsO_{3} | [28] |
| AsFA348 | C_{15}H_{30}AsO_{3} | [8],[24] |
| AsFA356 | C_{15}H_{32}AsO_{3} | [16],[30] |
| AsFA362 | C_{15}H_{32}AsO_{3} | [11],[4],[5],[6],[7],[8],[9],[10],[14],[16],[19] |
| thio-AsFA362 | C_{15}H_{32}AsO_{3}S | [7] |
| AsFA374 | C_{16}H_{32}AsO_{3} | [4],[14] |
| AsFA376 | C_{16}H_{32}AsO_{3} | [8],[24] |
| AsFA382 | C_{16}H_{32}AsO_{3} | [28] |
| AsFA386 | C_{16}H_{32}AsO_{3} | [22],[24] |
| AsFA388 | C_{16}H_{32}AsO_{3} | [4],[13],[18],[20],[21],[31] |
| AsFA390 | C_{16}H_{32}AsO_{3} | [11],[18],[9],[12],[16],[17],[20],[24] |
| AsFA404 | C_{20}H_{32}AsO_{3} | [8],[24],[30] |
| AsFA408 | C_{17}H_{34}AsO_{3} | [1] |
| AsFA418 | C_{21}H_{44}AsO_{3} | [4],[8],[14],[24] |
| AsFA422 | C_{22}H_{56}AsO_{3} | [28] |
| Compound | Mass  | Formula          | References |
|----------|-------|------------------|------------|
| AsFA436  | 437.20| C$_{23}$H$_{40}$AsO$_3$ | [1],[5],[6],[8],[12],[16],[20] |
| AsFA446  | 447.28| C$_{23}$H$_{40}$AsO$_3$ | [24] |
| AsFA448  | 449.20| C$_{23}$H$_{40}$AsO$_3$ | [1],[5],[6],[12],[21],[30] |
| AsFA462  | 463.22| C$_{23}$H$_{40}$AsO$_3$ | [28] |
| AsFA474  | 475.31| C$_{23}$H$_{40}$AsO$_3$ | [24] |
| AsFA486  | 487.31| C$_{23}$H$_{40}$AsO$_3$ | [24] |
| AsFA502  | 503.34| C$_{23}$H$_{40}$AsO$_3$ | [24] |
| AsFA528  | 529.27| C$_{23}$H$_{40}$AsO$_3$ | [15] |
| AsPC885  | 886.50| C$_{45}$H$_{81}$O$_9$NAsP | [15] |
| AsPC911  | 912.51| C$_{47}$H$_{83}$O$_9$NAsP | [15] |
| AsPC939  | 940.54| C$_{49}$H$_{87}$O$_9$NAsP | [15] |
| AsPC985  | 986.52| C$_{53}$H$_{85}$O$_9$NAsP | [11],[15] |
| AsPC997  | 998.52| C$_{54}$H$_{85}$O$_9$NAsP | [15] |
| AsPE1035 | 1036.54| C$_{57}$H$_{87}$O$_9$NAsP | [11],[15] |
| TMAsFOH37 | 375.26| C$_{20}$H$_{44}$AsO | [5] |
| TMAsFOH41 | 419.23| C$_{24}$H$_{46}$AsO | [5],[21],[23] |
| AsOH375  |       |                  | [13] |
Table S2. The instrument parameters of ICP-MS $^a$.

| Parameter                              | Value            |
|----------------------------------------|------------------|
| Radio frequency power                  | 1500 W           |
| Sample cone depth                      | 7.0 mm           |
| Carrier gas                            | 0.9-1.0 L/min argon |
| Makeup gas                             | 0 L/min argon    |
| Spray chamber temperature (TEM)        | -5 °C            |
| Reaction cell He gas                   | 3.0 mL/min       |

$^a$The same krill oil sample was analyzed by HPLC-ICPMS and HPLC-ESIMS twice. We used the same chromatographic system and same chromatographic conditions. Dimethylarsinic acid (DMA, naturally present in krill oil sample) was used as an internal standard. We adjusted the length of tubing between the HPLC and the mass spectrometers to ascertain that the retention times of DMA were identical in the two runs.
**Table S3.** The candidate precursor ions calculated by hierarchical clustering and wavelet coherence methods in each SWATH window in the untargeted analysis of 5 arsenic standards including iAs$^V$, MMA, NAHAA, 3AHPAA, Rox in water using reversed phase chromatographic separation and SWATH acquisition in tripleToF MS. MS1 of SWATH acquisition covered ions with m/z from 100 to 300. The corresponding peak (peaks shown in Figure S1A) of each candidate precursor ion and the wavelet coherence of each candidate precursor ion with the summed signal of product ions (m/z 106.9120 and m/z 122.9069) were also present in this table.

| Candidate precursor ions | SWATH window (m/z) | Hierarchical clustering (1) | Wavelet coherence(0.7) | Hierarchical clustering(1) | Wavelet coherence(0.7) |
|--------------------------|--------------------|----------------------------|-----------------------|----------------------------|-----------------------|
|                          | 100-150            | m/z | peak no. (coherence) | m/z | peak no. (coherence) | m/z | peak no. (coherence) |
| 106.913                  | 1                  | 106.913 | 1(0.76) | 204.967 | 3 | 213.951 | 3(0.87) |
| 120.929                  | 1                  | 120.929 | 1(0.76) | 213.951 | 3 | 216.917 | 3(0.80) |
| 122.908                  | 1                  | 122.908 | 1(0.81) | 231.961 | 3 | 222.892 | 3(0.77) |
| 123.034                  | 1                  | 123.034 | 1(0.73) | 234.979 | 3 | 231.961 | 3(0.86) |
| 138.939                  | 1                  | 123.916 | 1(0.76) | 218.897 | 4 | 213.951 | 4(0.78) |
| 106.913                  | 2                  | 138.939 | 1(0.76) | 224.872 | 4 | 216.917 | 4(0.74) |
| 122.908                  | 2                  | 140.919 | 1(0.77) | 230.889 | 4 | 222.892 | 4(0.71) |
| 140.919                  | 2                  | 122.908 | 2(0.89) | 234.979 | 4 | 231.961 | 4(0.77) |
| 122.908                  | 2                  | 140.919 | 2(0.88) | 243.925 | 5 | 215.094 | 5(0.77) |
| 123.916                  | 2                  |        |            |        |            | 216.917 | 5(0.75) |
| 138.939                  | 2                  |        |            |        |            | 228.950 | 5(0.70) |
|                          | 200-250            | m/z | peak no. (coherence) | m/z | peak no. (coherence) | m/z | peak no. (coherence) |
| 255.961                  | 6                  | 261.935 | 6(0.99) |        | |        | |
| 256.957                  | 6                  | 262.937 | 6(0.99) |        | |        | |
| 273.972                  | 6                  | 275.951 | 6(0.99) |        | |        | |
| 275.977                  | 6                  | 257.105 | 7(0.70) |        | |        | |
| 287.988                  | 6                  | 261.935 | 7(1.00) |        | |        | |
| 295.954                  | 6                  | 262.937 | 7(0.99) |        | |        | |
| 261.935                  | 7                  | 275.951 | 7(1.00) |        | |        | |
| 262.937                  | 7                  | 283.918 | 7(0.83) |        | |        | |
| 275.951                  | 7                  |        |            |        |            |        | |
| 283.918                  | 7                  |        |            |        |            |        | |
| 287.988                  | 7                  |        |            |        |            |        | |
| 295.954                  | 7                  |        |            |        |            |        | |
|                          | 250-300            | m/z | peak no. (coherence) | m/z | peak no. (coherence) | m/z | peak no. (coherence) |
|                          | m/z | peak no. (coherence) | m/z | peak no. (coherence) | m/z | peak no. (coherence) |
|                          | 255.961            | 6 | 261.935 | 6(0.99) | | |
|                          | 256.957            | 6 | 262.937 | 6(0.99) | | |
|                          | 273.972            | 6 | 275.951 | 6(0.99) | | |
|                          | 275.977            | 6 | 257.105 | 7(0.70) | | |
|                          | 287.988            | 6 | 261.935 | 7(1.00) | | |
|                          | 295.954            | 6 | 262.937 | 7(0.99) | | |
|                          | 261.935            | 7 | 275.951 | 7(1.00) | | |
|                          | 262.937            | 7 | 283.918 | 7(0.83) | | |
|                          | 275.951            | 7 |        |            | | |
|                          | 283.918            | 7 |        |            | | |
|                          | 287.988            | 7 |        |            | | |
|                          | 295.954            | 7 |        |            | | |
Table S4. The candidate precursor ions calculated by hierarchical clustering and wavelet coherence methods in each SWATH window in the untargeted analysis of arsenolipids in a krill oil sample using reversed phase chromatographic separation and SWATH acquisition in tripleToF MS. MS1 of SWATH acquisition covered ions with m/z from 270 to 500. The corresponding peak (peaks shown in Figure 2A) of each candidate precursor ion and the wavelet coherence of each candidate precursor ion with the summed signal of product ions (m/z 104.9685 and m/z 102.9529) were also present in this table.

| SWATH window (m/z) | 270-300 | 299-325 |
|--------------------|---------|---------|
| **Hierarchical clustering (1.5)** | Wavelet coherence(0.6) | **Hierarchical clustering (1.5)** | Wavelet coherence(0.6) |
| m/z | peak no. | m/z | peak no. (coherence) | m/z | peak no. | m/z | peak no. (coherence) |
| 284.238 | 1 | 276.124 | 1(0.70) | 301.643 | 3 | 301.643 | 3(0.98) |
| 297.160 | 1 | 280.179 | 1(0.97) | 302.146 | 3 | 302.146 | 3(0.63) |
| 271.170 | 2 | 280.680 | 1(0.68) | 306.222 | 3 | 306.222 | 3(0.96) |
| 277.180 | 2 | 284.238 | 1(0.60) | 310.180 | 3 | 310.180 | 3(0.77) |

| Candidate precursor ions |
|--------------------------|
| 286.253 | 1(0.61) | 316.250 | 3(0.90) |
| 291.171 | 1(0.68) | 318.242 | 3(0.77) |
| 295.140 | 1(0.81) | 322.240 | 3(0.66) |
| 297.160 | 1(0.86) | 301.643 | 4(0.90) |
| 280.179 | 2(0.96) | 302.146 | 4(0.66) |
| 280.680 | 2(0.62) | 306.222 | 4(0.97) |
| 291.171 | 2(0.66) | 310.180 | 4(0.70) |
| 294.221 | 2(0.65) | 316.250 | 4(0.84) |
| 295.140 | 2(0.74) | 318.240 | 4(0.68) |
| 297.160 | 2(0.82) | |

| SWATH window (m/z) | 350-375 | 374-400 |
|--------------------|---------|---------|
| **Hierarchical clustering (1.5)** | Wavelet coherence(0.6) | **Hierarchical clustering (1.5)** | Wavelet coherence(0.6) |
| m/z | peak no. | m/z | peak no. (coherence) | m/z | peak no. | m/z | peak no. (coherence) |
| 358.252 | 5 | 351.217 | 5(0.64) | 391.219 | 6 | 384.311 | 6(0.80) |
| 363.188 | 5 | 358.252 | 5(0.67) | 394.259 | 6 | 385.317 | 6(0.96) |
| 373.099 | 5 | 362.196 | 5(0.94) | 396.251 | 6 | 388.320 | 6(0.73) |

| Candidate precursor ions |
|--------------------------|
| 363.188 | 5 | 362.196 | 5(0.94) | 396.310 | 6 | 391.219 | 6(0.96) |
| 363.265 | 5(0.96) | 397.317 | 6 | 392.279 | 6(0.65) |
| 367.152 | 5(0.94) | 399.214 | 6 | 394.259 | 6(0.62) |

| SWATH window (m/z) | 399-425 | 424-450 |
|--------------------|---------|---------|
| Candidate precursor ions | Hierarchical clustering(1.5) | Wavelet coherence(0.6) | Hierarchical clustering(1.5) | Wavelet coherence(0.6) |
|--------------------------|-------------------------------|------------------------|-------------------------------|------------------------|
| m/z peak no.             | m/z peak no. (coherence)     | m/z peak no.           | m/z peak no. (coherence)     |
| 404.254 7               | 404.254 7(0.63)              | 437.224 9              | 425.168 9(0.63)              |
| 399.214 8               | 408.214 7(0.73)              | 440.250 9              | 434.290 9(0.66)              |
| 414.262 8               | 410.330 7(0.62)              | 440.336 9              | 437.224 9(0.75)              |
| 415.212 8               | 418.295 7(0.63)              | 437.202 10             | 440.336 9(0.86)              |
| 420.310 8               | 419.298 7(0.65)              | 439.208 10             | 445.341 9(0.83)              |
| 422.320 8               | 420.310 7(0.85)              | 449.336 10             | 424.268 10(0.84)             |
| 421.313 7               | 425.272 10(0.65)             | 433.316 10             | 10(0.79)                     |
| 399.214 8               | 408.214 7(0.73)              | 440.250 9              | 434.290 9(0.66)              |
| 409.316 8               | 410.330 7(0.62)              | 440.336 9              | 437.224 9(0.75)              |
| 412.246 8               | 418.295 7(0.63)              | 437.202 10             | 440.336 9(0.86)              |
| 415.212 8               | 419.298 7(0.65)              | 439.208 10             | 445.341 9(0.83)              |
| 420.310 8               | 421.313 7(0.76)              | 425.272 10             | 433.316 10(0.79)             |
| 422.320 8               | 422.320 7(0.60)              | 436.301 10             | 437.202 10(0.80)             |
| 409.316 8               | 412.246 8(0.72)              | 439.208 10             | 440.336 10(0.75)             |
| 415.212 8               | 415.232 8(0.75)              | 445.341 10             | 448.245 10(0.76)             |
| 418.315 8               | 420.310 8(0.93)              | 449.203 10             | 449.336 10(0.76)             |
| 421.305 8               | 424.270 8(0.70)              | 449.336 10             | 10(0.76)                     |
| 424.270 8               | 492.331 11(0.75)             | 492.331 11             | 499.180 11(0.89)             |
| 497.342 11              | 499.180 11(0.89)             | 485.190 12             | 486.194 12(0.61)             |
| 499.180 11              | 485.190 12(0.61)             | 487.196 12             | 492.331 12(0.68)             |

**SWATH window (m/z)** 485-500
**Table S5.** The candidate precursors ion summarized from Table S4 after being rounded up to one decimal place. All these candidate precursor ions were subjected to product ion scan for further confirmation. The underlined ions were confirmed to produce arsenic fragments.

| SWATH window (m/z) | 270-300 | 299-325 | 350-375 | 374-400 | 399-425 | 424-450 | 485-500 |
|--------------------|---------|---------|---------|---------|---------|---------|---------|
| 271.2              | 301.2   | 351.2   | 384.3   | 404.3   | 429.1   | 487.2   |
| 276.1              | 301.6   | 358.3   | 385.3   | 408.2   | 433.3   | 492.3   |
| 277.2              | 302.1   | 362.2   | 388.3   | 409.3   | 434.3   | 497.3   |
| 280.2              | 302.2   | 363.2   | 391.2   | 412.2   | 436.3   | 499.2   |
| 280.7              | 306.2   | 363.3   | 392.3   | 414.3   | 437.2   |         |
| 281.2              | 307.2   | 367.2   | 394.3   | 415.2   | 439.2   |         |

Candidate precursor ions

| 282.2 | 310.2 | 373.1 | 396.3 | 418.3 | 440.3 |
| 284.2 | 311.2 |       | 397.3 | 419.3 | 440.4 |
| 286.3 | 315.1 | 399.2 | 420.3 | 445.3 |       |
| 287.2 | 318.2 |       | 421.3 | 446.3 |       |
| 291.2 | 319.2 |       |       | 448.2 |       |
| 294.2 | 319.3 |       |       | 449.2 |       |
| 297.2 | 322.2 |       |       |       | 449.3 |
**Table S6.** The candidate precursor ions calculated by hierarchical clustering and wavelet coherence methods in each SWATH window in the untargeted analysis of arsenolipids in tuna extracts using reversed phase chromatographic separation and SWATH acquisition in tripleToF MS. MS1 of SWATH acquisition covered ions with m/z from 200 to 600. The corresponding peak (Figure S4A) of each candidate precursor ion and the wavelet coherence of each candidate precursor ion with the summed signal of product ions (m/z 104.9685 and m/z 102.9529) were also present in this table.

| SWATH window (m/z) | 324-350 | 349-375 |
|-------------------|---------|---------|
| Hierarchical clustering (1.5) | Wavelet coherence (0.6) | Hierarchical clustering (1.5) | Wavelet coherence (0.6) |
| m/z peak no. | m/z peak no. (coherence) | m/z peak no. | m/z peak no. (coherence) |
| 333.214 1 | 325.178 1(0.74) | 363.183 2 | 359.221 2(0.61) |
| 327.308 1(0.66) | 305.663 3 | 365.247 4 | 374.203 2(0.67) |
| 328.197 1(0.75) | 370.387 4 | 374.203 2(0.67) | |
| 332.293 1(0.63) | 332.293 1(0.63) | 352.322 4(0.62) | |
| 333.146 1(0.87) | 364.695 4(0.63) | 369.807 4(0.70) | |
| 333.214 1(1.00) | 364.695 4(0.63) | 373.326 4(0.64) | |
| 337.236 1(0.68) | 339.194 1(0.61) | 340.208 1(0.63) | |
| 339.253 1(0.77) | 341.209 1(0.88) | 341.305 1(0.67) | |
| 340.208 1(0.63) | 349.214 1(0.61) | 349.215 1(0.61) | |
| 377.230 5(0.89) | 378.234 5(0.79) | 381.204 5(0.86) | |
| 384.105 5(0.86) | 384.223 5(0.65) | 384.223 5(0.65) | |
| 388.342 5(0.79) | 389.194 5(0.82) | 389.230 5(0.68) | |

| SWATH window (m/z) | 374-400 | 400-425 |
|-------------------|---------|---------|
| Hierarchical clustering (1.5) | Wavelet coherence (0.6) | Hierarchical clustering (1.5) | Wavelet coherence (0.4) |
| m/z peak no. | m/z peak no. (coherence) | m/z peak no. | m/z peak no. (coherence) |
| 391.210 5 | 374.203 5(0.89) | 419.242 7 | 401.229 6(0.50) |
| 392.212 5 | 376.220 5(0.88) | 423.214 7 | 403.235 6(0.42) |
| 397.230 5(0.88) | 424.218 7 | 404.206 6(0.41) | |
| 378.234 5(0.89) | 403.233 8 | 405.226 6(0.48) | |
| 381.204 5(0.86) | 403.284 8 | 407.075 6(0.47) | |
| 384.223 5(0.65) | 419.276 8 | 408.178 6(0.44) | |
| 388.342 5(0.79) | 409.163 6(0.45) | 409.163 6(0.45) | |
| 389.194 5(0.82) | 389.230 5(0.68) | 410.254 6(0.43) | |
| m/z   | peak no. | (coherence) | m/z   | peak no. | (coherence) |
|-------|----------|-------------|-------|----------|-------------|
| 390.197 | 5         | 0.66        | 411.171 | 6         | 0.48        |
| 391.210 | 5         | 0.85        | 413.266 | 6         | 0.42        |
| 392.291 | 5         | 0.67        | 417.225 | 6         | 0.57        |
| 396.311 | 5         | 0.92        | 420.244 | 6         | 0.42        |
| 397.198 | 5         | 0.90        | 421.190 | 6         | 0.48        |
| 398.202 | 5         | 0.90        | 400.342 | 7         | 0.87        |
| 398.342 | 5         | 0.65        | 401.327 | 7         | 0.95        |
| 399.214 | 5         | 0.90        | 405.214 | 7         | 0.85        |
| 405.261 |           |             | 407.277 | 7         | 0.66        |
| 407.313 |           |             | 407.313 | 7         | 0.65        |
| 419.242 |           |             | 419.276 | 7         | 0.62        |
| 420.280 |           |             | 420.280 | 7         | 0.61        |
| 421.329 |           |             | 423.214 | 7         | 0.67        |
| 424.218 |           |             | 424.218 | 7         | 0.69        |
| 400.342 |           |             | 401.286 | 8         | 0.89        |
| 401.326 |           |             | 401.326 | 8         | 0.95        |
| 405.214 |           |             | 405.214 | 8         | 0.89        |
| 405.261 |           |             | 405.261 | 8         | 0.67        |
| 407.277 |           |             | 407.277 | 8         | 0.73        |
| 407.313 |           |             | 407.313 | 8         | 0.67        |
| 419.276 |           |             | 419.276 | 8         | 0.61        |
| 420.280 |           |             | 420.280 | 8         | 0.60        |
| 421.329 |           |             | 421.329 | 8         | 0.72        |
| 423.310 |           |             | 423.310 | 8         | 0.70        |

**SWATH window (m/z)**

| m/z   | peak no. | (coherence) | m/z   | peak no. | (coherence) |
|-------|----------|-------------|-------|----------|-------------|
| 449.205 | 9         | 0.65        | 449.205 | 11        | 0.65        |
| 472.407 | 11        | 0.88        | 471.237 | 11        | 0.65        |
| 471.237 | 11        | 0.65        | 471.237 | 11        | 0.65        |
| 471.405 | 11        | 0.87        | 471.405 | 11        | 0.87        |
| 471.405 | 11        | 0.87        | 471.405 | 11        | 0.87        |

**Candidate precursor ions**

| m/z   | peak no. | (coherence) | m/z   | peak no. | (coherence) |
|-------|----------|-------------|-------|----------|-------------|
| 449.205 | 9         | 0.65        | 449.205 | 11        | 0.65        |
| 472.407 | 11        | 0.88        | 471.237 | 11        | 0.65        |
| 471.237 | 11        | 0.65        | 471.237 | 11        | 0.65        |
| 471.405 | 11        | 0.87        | 471.405 | 11        | 0.87        |
| 471.405 | 11        | 0.87        | 471.405 | 11        | 0.87        |
| 471.405 | 11        | 0.87        | 471.405 | 11        | 0.87        |
| SWATH window (m/z) | Hierarchical clustering (1.5) | Wavelet coherence(0.6) |
|-------------------|-------------------------------|-----------------------|
| m/z               | peak no.                      | m/z                   | peak no. (coherence) |
| 529.319           | 12                             | 524.275               | 12(0.71)             |
| 533.367           | 12                             | 527.359               | 12(0.87)             |
| 534.370           | 12                             | 529.267               | 12(0.84)             |
| 537.344           | 12                             | 529.327               | 12(0.60)             |
|                   |                                | 531.362               | 12(0.61)             |
|                   |                                | 533.367               | 12(0.71)             |
|                   |                                | 538.291               | 12(0.80)             |
|                   |                                | 538.351               | 12(0.66)             |
|                   |                                | 539.354               | 12(0.71)             |
|                   |                                | 540.307               | 12(0.69)             |
Table S7. The candidate precursors ion summarized from Table S6 after being rounded up to one decimal place. All these candidate precursor ions were subjected to product ion scan for further confirmation. The underlined ions were confirmed to produce arsenic fragments.

| SWATH window (m/z) | 324-350 | 349-375 | 374-400 | 400-425 | 425-450 | 449-475 | 524-550 |
|--------------------|---------|---------|---------|---------|---------|---------|---------|
| 325.2              | 305.7   | 374.2   | 400.3   | 425.3   | 449.2   | 524.3   |         |
| 327.3              | 352.3   | 376.2   | 401.3   | 426.4   | 471.2   | 527.4   |         |
| 328.2              | 359.2   | 377.2   | 403.2   | 427.4   | 471.4   | 529.3   |         |
| 332.3              | 363.2   | 378.2   | 403.3   | 437.2   | 472.4   | 531.4   |         |
| 333.1              | 364.7   | 381.2   | 405.2   | 442.3   | 474.3   | 533.4   |         |
| 333.2              | 365.3   | 384.2   | 405.3   | 445.3   | 534.4   |         |         |
| 337.2              | 369.8   | 388.3   | 407.3   | 445.4   | 537.3   |         |         |
| 339.2              | 370.4   | 389.2   | 419.2   | 449.2   | 538.3   |         |         |
| 339.3              | 373.3   | 390.2   | 419.3   | 449.3   | 538.4   |         |         |
| 340.2              | 374.2   | 391.2   | 420.3   |         |         | 539.4   |         |
| 341.2              | 392.2   | 421.2   |         |         |         | 540.3   |         |
| 341.3              | 392.3   | 423.2   |         |         |         |         |         |
| 349.2              | 396.3   | 423.3   |         | 397.2   | 424.2   |         |         |
|                    | 398.2   |         |         |         |         |         |         |
|                    | 398.3   |         |         |         |         |         |         |
|                    | 399.2   |         |         |         |         |         |         |
Table S8. The candidate precursor ions calculated by hierarchical clustering and wavelet coherence methods in each SWATH window in the untargeted analysis of arsenolipids in extracts of hairtail heads using reversed phase chromatographic separation and SWATH acquisition in tripleToF MS. MS1 of SWATH acquisition covered ions with m/z from 100 to 550. The corresponding peak (Figure S6A) of each candidate precursor ion and the wavelet coherence of each candidate precursor ion with the summed signal of product ions (m/z 104.9685 and m/z 102.9529) were also present in this table.

| SWATH window (m/z) | 149-175 | 324-350 |
|-------------------|---------|---------|
|                   | Hierarchical clustering(1.5) | wavelet coherence(0.6) | Hierarchical clustering(1.5) | wavelet coherence(0.6) |
| m/z               | peak no. | m/z | coherence | peak no. | m/z | coherence | peak no. |
| 151.075           | 1        | 150.057 | 0.73 | 1       | 333.213 | 2       | 324.259 | 0.64 | 2       |
| 152.971           | 1        | 150.112 | 0.83 | 1       | 347.228 | 2       | 331.263 | 0.78 | 2       |
|                   |          | 151.144 | 0.79 | 1       | 341.224 | 2       | 332.267 | 0.75 | 2       |
|                   |          | 151.148 | 0.70 | 1       | 333.213 | 2       | 333.213 | 0.83 | 2       |
|                   |          | 152.056 | 0.94 | 1       | 341.194 | 2       | 341.194 | 0.67 | 2       |
|                   |          | 152.147 | 0.79 | 1       | 332.267 | 2       | 341.224 | 0.61 | 2       |
|                   |          | 152.971 | 0.99 | 1       | 342.373 | 2       | 331.263 | 0.78 | 2       |
|                   |          | 153.040 | 0.88 | 1       | 346.274 | 2       | 347.228 | 0.87 | 2       |
|                   |          | 153.149 | 0.69 | 1       | 347.277 | 2       | 347.277 | 0.87 | 2       |
|                   |          | 154.059 | 0.76 | 1       | 347.277 | 2       | 347.277 | 0.87 | 2       |
|                   |          | 156.032 | 0.74 | 1       | 348.261 | 2       | 348.261 | 0.64 | 2       |
|                   |          | 156.102 | 0.84 | 1       |          |         |         |       |         |
|                   |          | 156.138 | 0.84 | 1       |          |         |         |       |         |
|                   |          | 159.028 | 0.94 | 1       |          |         |         |       |         |
|                   |          | 161.029 | 0.85 | 1       |          |         |         |       |         |
| Candidate precursor ions |       | 162.112 | 0.72 | 1       |          |         |         |       |         |
|                   |          | 163.974 | 0.63 | 1       |          |         |         |       |         |
|                   |          | 164.092 | 0.71 | 1       |          |         |         |       |         |
|                   |          | 164.985 | 0.87 | 1       |          |         |         |       |         |
|                   |          | 165.054 | 0.74 | 1       |          |         |         |       |         |
|                   |          | 166.002 | 0.63 | 1       |          |         |         |       |         |
|                   |          | 166.072 | 0.95 | 1       |          |         |         |       |         |
|                   |          | 167.091 | 0.66 | 1       |          |         |         |       |         |
|                   |          | 168.065 | 0.68 | 1       |          |         |         |       |         |
|                   |          | 168.174 | 0.94 | 1       |          |         |         |       |         |
|                   |          | 169.034 | 0.75 | 1       |          |         |         |       |         |
|                   |          | 169.986 | 0.63 | 1       |          |         |         |       |         |
|                   |          | 170.033 | 0.78 | 1       |          |         |         |       |         |
|                   |          | 170.190 | 0.91 | 1       |          |         |         |       |         |
|                   |          | 171.026 | 0.94 | 1       |          |         |         |       |         |
|                   |          | 171.062 | 0.84 | 1       |          |         |         |       |         |
|                   |          | 171.125 | 0.83 | 1       |          |         |         |       |         |
|                   |          | 173.042 | 0.74 | 1       |          |         |         |       |         |
| Candidate precursor ions | m/z | peak no. | m/z | coherence | peak no. | m/z | peak no. | m/z | coherence | peak no. |
|--------------------------|-----|----------|-----|-----------|----------|-----|----------|-----|-----------|----------|
| 351.287                  | 4   | 363.186  | 0.66| 3         | 391.219  | 5   | 384.223  | 0.68| 5         |
| 360.311                  | 4   | 364.190  | 0.64| 3         | 394.224  | 5   | 388.306  | 0.88| 5         |
| 361.245                  | 4   | 358.274  | 0.92| 4         | 391.219  | 0.97| 5         |
| 362.246                  | 4   | 358.368  | 0.97| 4         | 391.245  | 0.89| 5         |
| 371.262                  | 4   | 360.375  | 0.60| 4         | 391.340  | 0.74| 5         |
| 361.245                  | 4   | 358.274  | 0.98| 4         | 392.280  | 0.96| 5         |
| 362.246                  | 4   | 358.368  | 0.65| 4         | 394.224  | 0.78| 5         |
| 362.297                  | 4   | 360.375  | 0.89| 4         | 397.201  | 0.65| 5         |
| 363.230                  | 4   | 399.216  | 0.92| 4         | 399.216  | 0.72| 5         |
| 363.291                  | 4   | 360.375  | 0.64| 4         | 391.340  | 0.74| 5         |

| Candidate precursor ions | m/z | peak no. | m/z | coherence | peak no. | m/z | peak no. | m/z | coherence | peak no. |
|--------------------------|-----|----------|-----|-----------|----------|-----|----------|-----|-----------|----------|
| 403.198                  | 7   | 410.327  | 0.45| 7         | 435.176  | 13  | 429.227  | 0.85| 11        |
| 410.394                  | 7   | 413.381  | 0.46| 7         | 433.178  | 13  | 425.149  | 0.73| 12        |
| 415.151                  | 7   | 415.321  | 0.51| 7         | 433.178  | 14  | 436.306  | 0.87| 12        |
| 407.221                  | 8   | 416.324  | 0.48| 7         | 435.176  | 14  | 437.196  | 0.65| 12        |
| 414.335                  | 8   | 418.389  | 0.64| 7         | 441.235  | 14  | 439.215  | 0.92| 12        |
| 419.229                  | 8   | 399.337  | 0.66| 8         | 443.327  | 15  | 449.203  | 0.94| 12        |
| 424.342                  | 8   | 407.221  | 0.96| 8         | 444.406  | 15  | 427.306  | 0.61| 13        |
| 407.221                  | 9   | 412.342  | 0.91| 8         | 435.176  | 15  | 433.178  | 0.72| 13        |
| 407.229                  | 9   | 414.333  | 0.88| 8         | 441.235  | 15  | 434.384  | 0.76| 13        |
| 416.352                  | 9   | 414.333  | 0.87| 8         | 435.176  | 15  | 437.194  | 0.69| 13        |
| 424.342                  | 9   | 415.321  | 0.51| 8         | 437.194  | 0.69| 13        |
| 424.342                  | 10  | 416.324  | 0.46| 8         | 439.201  | 0.67| 13        |
| 410.251                  | 10  | 419.229  | 0.62| 8         | 439.247  | 0.69| 13        |
| 416.352                  | 10  | 419.275  | 0.88| 8         | 441.235  | 0.67| 13        |
| 424.342                  | 10  | 421.282  | 0.57| 8         | 432.240  | 0.67| 15        |
|                          |     | 422.317  | 0.58| 8         | 447.279  | 0.64| 15        |
|                          |     | 422.399  | 0.51| 8         | 424.342  | 0.69| 8         |
| m/z   | peak no. | m/z   | coherence | peak no. |
|-------|----------|-------|-----------|----------|
| 526.447 | 16       | 524.274 | 0.68      | 16       |
| 529.266 | 16       | 524.314 | 0.76      | 16       |

| Candidate precursor ions |
|--------------------------|
| 526.447 0.98 16 |
| 529.266 0.98 16 |
| 540.305 0.82 16 |
| 542.322 0.85 16 |
| 543.327 0.85 16 |
| 545.333 0.86 16 |

**SWATH window (m/z)**

524-550
**Table S9.** The candidate precursors ion summarized from Table S8 after being rounded up to one decimal place. All these candidate precursor ions were subjected to product ion scan for further confirmation. The underlined ions were confirmed to produce arsenic fragments.

| SWATH window(m/z) | 149-175 | 324-350 | 349-375 | 374-400 | 399-425 | 424-450 | 524-550 |
|-------------------|---------|---------|---------|---------|---------|---------|---------|
| 151.1             | 324.3   | 351.3   | 384.2   | 399.2   | **425.2** | 524.3   |
| 150.1             | 331.3   | 358.3   | 388.3   | 400.3   | 427.3   | 526.4   |
| 151.1             | 332.3   | 360.3   | **391.2** | 401.3   | 429.3   | **529.3** |
| 152.1             | 333.2   | **361.2** | 392.3   | 402.3   | **431.3** | 540.3   |
| 153.0             | 341.2   | 362.2   | 394.2   | 403.2   | 432.2   | 542.3   |
| 154.1             | 342.4   | **363.2** | 397.2   | **405.2** | 433.2   | 543.3   |
| 156.0             | 346.3   | 364.2   | 399.2   | **407.2** | 434.2   | 545.3   |
| 159.0             | 347.2   | 371.3   | 408.2   | 435.2   |         |         |
| 161.0             | 348.3   |         |         | 409.2   | 436.3   |         |
| 162.1             |         |         |         | 410.3   | 437.2   |         |
| 164.0             |         |         |         | 411.2   | 439.2   |         |
| 165.0             |         |         |         | 412.3   | **441.2** |         |
| 166.0             |         |         |         | 413.4   | 443.3   |         |
| 167.1             |         |         |         | 414.3   | 444.4   |         |
| 168.1             |         |         |         | 415.2   | **447.3** |         |
| 169.0             |         |         |         | 416.3   | **449.2** |         |
| 170.0             |         |         |         |         |         |         |
| 171.0             |         |         |         |         |         |         |
| 173.0             |         |         |         |         |         |         |
| 174.0             |         |         |         |         |         |         |

Candidate precursor ions
Table S10. The candidate precursor ions calculated by hierarchical clustering and wavelet coherence methods in each SWATH window in the untargeted analysis of arsenolipids in extracts of kelp using reversed phase chromatographic separation and SWATH acquisition in tripleToF MS. MS1 of SWATH acquisition covered ions with m/z from 100 to 550. The corresponding peak (Figure S10A) of each candidate precursor ion and the wavelet coherence of each candidate precursor ion with the summed signal of product ions (m/z 104.9685 and m/z 102.9529) were also present in this table.

| SWATH window(m/z) | 349-375 | 399-425 |
|-------------------|---------|---------|
| **Candidate precursor ions** |         |         |
| **Hierarchical clustering(1.5)** | **wavelet coherence(0.6)** | **Hierarchical clustering(1.5)** | **wavelet coherence(0.6)** |
| m/z | peak no. | m/z | coherence | peak no. | m/z | peak no. | m/z | coherence | peak no. |
| 357.218 | 1 | 351.289 | 0.91 | 1 | 402.337 | 2 | 399.216 | 0.61 | 2 |
| 361.243 | 1 | 353.295 | 0.90 | 1 | 406.331 | 2 | 400.218 | 0.71 | 2 |
| 355.224 | 0.62 | 1 | 413.197 | 3 | 404.316 | 0.70 | 2 |
| 357.263 | 0.62 | 1 | 421.198 | 3 | 405.320 | 0.84 | 2 |
| 357.264 | 0.62 | 1 | 425.203 | 3 | 406.353 | 0.89 | 2 |
| 359.197 | 0.71 | 1 | 413.197 | 4 | 417.219 | 0.63 | 2 |
| 359.198 | 0.75 | 1 | 421.198 | 4 | 423.187 | 0.90 | 2 |
| 361.357 | 0.92 | 1 | 425.203 | 4 | 401.098 | 0.81 | 3 |
| 362.305 | 0.70 | 1 | 402.337 | 0.78 | 3 |
| 365.245 | 0.69 | 1 | 404.316 | 0.70 | 3 |
| 365.302 | 0.66 | 1 | 405.320 | 0.84 | 3 |
| 367.322 | 0.90 | 1 | 406.331 | 0.62 | 3 |
| 367.326 | 0.64 | 1 | 419.108 | 0.76 | 3 |
| 370.295 | 0.67 | 1 | 423.188 | 0.90 | 3 |
| 371.326 | 0.68 | 1 | 425.203 | 0.99 | 3 |
| 374.275 | 0.62 | 1 | 425.268 | 0.78 | 3 |
|                      |         |                      | 403.113 | 0.65 | 4 |
|                      |         |                      | 404.315 | 0.64 | 4 |
|                      |         |                      | 406.331 | 0.62 | 4 |
|                      |         |                      | 411.190 | 0.70 | 4 |
|                      |         |                      | 414.227 | 0.61 | 4 |
|                      |         |                      | 419.183 | 0.65 | 4 |
|                      |         |                      | 423.188 | 0.90 | 4 |
|                      |         |                      | 425.269 | 0.74 | 4 |

| SWATH window(m/z) | 424-450 | 474-500 |
|-------------------|---------|---------|
| **Candidate precursor ions** |         |         |
| **Hierarchical clustering(1.5)** | **wavelet coherence(0.6)** | **Hierarchical clustering(1.5)** | **wavelet coherence(0.6)** |
| m/z | peak no. | m/z | coherence | peak no. | m/z | peak no. | m/z | coherence | peak no. |
| 425.203 | 5 | 425.203 | 0.99 | 5 | 476.265 | 7 | 474.285 | 0.83 | 7 |
| 425.268 | 5 | 425.268 | 0.78 | 5 | 479.262 | 7 | 476.265 | 0.85 | 7 |
| 426.207 | 5 | 427.114 | 0.66 | 5 | 481.220 | 7 | 479.262 | 0.80 | 7 |
| 437.194 | 5 | 432.297 | 0.78 | 5 | 486.306 | 7 | 481.220 | 0.92 | 7 |
| 439.201 | 5 | 437.194 | 0.67 | 5 | 496.340 | 7 | 486.306 | 0.87 | 7 |
| 425.203 | 6 | 439.201 | 0.67 | 5 | 499.240 | 7 | 494.253 | 0.86 | 7 |
|    |      |      |      |      |      |      |
|----|------|------|------|------|------|------|
| 425.268 | 6  | 442.331 | 0.70 | 5 | 499.240 | 0.96 | 7 |
| 425.269 | 6 | 445.124 | 0.62 | 5 | 499.240 | 0.96 |    |
| 426.207 | 6 | 425.269 | 0.74 | 6 |      |      |    |
| 442.288 | 6 | 426.207 | 0.97 | 6 |      |      |    |
|    |     | 433.177 | 0.64 | 6 |      |      |    |
Table S11. The candidate precursors ion summarized from Table S10 after being rounded up to one decimal place. All these candidate precursor ions were subjected to product ion scan for further confirmation. The underlined ions were confirmed to produce arsenic fragments.

| SWATH window(m/z) | 345-375 | 399-425 | 424-450 | 474-500 |
|-------------------|---------|---------|---------|---------|
| 351.3             | 399.2   | **425.2** | 474.3   |
| 353.3             | 400.2   | 426.2   | 476.3   |
| 355.2             | 401.1   | 427.1   | 479.3   |
| 357.3             | 402.3   | 432.3   | 481.2   |
| 359.2             | 403.1   | 433.2   | 486.3   |
| **361.2**         | 404.3   | 437.2   | 494.3   |
| 362.3             | 405.3   | 439.2   | 496.3   |
| 365.2             | 406.3   | **442.3** | **499.2** |
| 367.3             | 411.2   | 445.1   |         |
| 370.3             | 413.2   |         |         |
| 371.3             | 414.2   |         |         |
| 374.3             | 417.2   |         |         |
| 419.1             |         |         |         |
| 421.2             |         |         |         |
| **423.2**         |         |         |         |
Figure S1. (A) The XICs of AsO$_2^-$ and AsO$_3^-$ in 3 SWATH windows obtained from RP HPLC-SWATH of 50 µg/L arsenic standards (iAs$^V$, MMA, NAHAA, 3AHPAA, and Rox) in water. Seven peaks were detected in the windows of m/z 100-150, 200-250, and 250-300. (B) The XICs of iAs$^V$, MMA, NAHAA, 3AHPAA, and Rox in the same SWATH analysis.
Figure S2. MS/MS spectrum and proposed structure of (a) AsFA362, (b) AsFA390, (c) AsFA436, and (d) AsFA 448 in krill oil.
**Figure S3.** The measured isotope patterns of (a) doubly-charged species (m/z 301.64) and (b) singly-charged species (m/z 499.18) of AslysoPC601.
Figure S4. (A) The XICs of As(CH$_2$)$_2^+$, As(CH$_3$)$_2^+$, and As(CH$_3$)$_2$OH$_2^+$ in 7 SWATH windows obtained from RP HPLC-SWATH of a tuna lipid extract. Twelve peaks were detected in the windows of m/z 324-350, 349-375, 374-400, 400-425, 424-450, 449-475, and 524-550. (B) The XICs of As(CH$_2$)$_2^+$ and As(CH$_3$)$_2^+$ in the product ion scan of precursor ions at m/z 333.214, 363.183, 359.221, 391.210, 405.214, 419.242, 421.329, 449.205, and 529.267.
Figure S5. MS/MS spectrum and proposed structure of (a) AsHC332, (b) AsFA362, (c) AsFA390, (d) AsHC404, (e) AsFA418, (f) AsFA448, and (g) AsFA528 in tuna fillets extracts.
Figure S6. (A) Sixteen peaks of As(CH$_2$)$_2^+$, As(CH$_3$)$_2^+$, and As(CH$_3$)$_2$OH$_2^+$ were detected in 7 SWATH windows obtained from the RP HPLC-SWATH of the lipid extract of hairtail heads. (B) The XICs of As(CH$_2$)$_2^+$ and As(CH$_3$)$_2^+$ in the product ion scan of precursor ions at m/z 152.971, 333.213, 361.245, 363.186, 391.219, 405.213, 407.229, 409.245, 419.250, 425.203, 431.250, 441.235, 447.279, 449.203, and 529.266.
Figure S7. MS/MS spectra and proposed structures of (A) AsHC332, (B) AsHC360, and (C) AsHC362 in the lipid extract of hairtail heads.
Figure S8. MS/MS spectra and proposed structures of (A) AsFA390, (B) AsHC404, and (C) AsFA418 in the lipid extract of hairtail heads.
Figure S9. MS/MS spectra and proposed structures of (A) AsFA446, (B) AsFA448, and (C) AsFA528 in the lipid extract of hairtail heads.
Figure S10. (A) Seven peaks of As(CH$_2$)$_2^+$, As(CH$_3$)$_2^+$, and As(CH$_3$)$_2$OH$_2^+$ were detected in 4 SWATH windows obtained from the RP HPLC-SWATH of the kelp lipid extract. (B) The XICs of As(CH$_2$)$_2^+$ and As(CH$_3$)$_2^+$ in the product ion scan of precursor ions at m/z 361.243, 423.187, 425.203, and 499.240.
Figure S11. MS/MS spectra and proposed structures of (A) AsHC360 and (B) AsFA422 in the lipid extract of kelp.
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User manual for Precursorfinder v1.0

1. Overview

Precursorfinder v1.0 is used for the analysis of data generated by SWATH (Sequential Windowed Acquisition of all Theoretical fragment ions) technology on high-resolution mass spectrometry. Because there is no relationship between parent ions and product ions in SWATH data, it is inconvenient to find the parent ions of specific product ions. Precursorfinder v1.0 provides a solution for finding the parent ions for the product ions in SWATH data. The software has six functions including "reading in SWATH data", "detecting the peaks of specific product ion", "generating possible precursor ions", "calculating wavelet coherence to filter candidate parent ions", "calculating hierarchical clustering to filter candidate parent ions", and "generating wavelet coherence plot and XICs of specific product ion and precursor ion". Users can adjust the parameters in each function to optimize the results. Graphical user interface (GUI) is used to make the software visually acceptable and convenient to use.

2. Developing and operating environment

2.1. Developing environment

Platform: MATLAB R2018a
Language: MATLAB

2.2. Operating environment

Operating system: Windows Server 2003 R2/ Windows Server 2008 R2 Service Pack 1/ Windows Server 2008 Service Pack 2/ Windows Server 2012/ Windows XP x64 Edition Service Pack 2/ Windows XP Service Pack 3/ Windows Vista Service Pack 2/ Windows 7 Service Pack 1/ Windows 8/ Windows 8.1/ Windows 10.

Memory: > 4 GB
Hard disk space: > 10 GB (3-4 GB for installation of MATLAB)
Software needed: 1) MATLAB R2018a

2) ProteoWizard MS Converter 3.0.10702 or later version

3) AB Sciex OS or AB Sciex Peakview (From AB Sciex)

3. Software design

3.1. Software structure

1. Data input and peak detection
   1) Importing SWATH data
   2) Importing the list of all precursor ions
   3) Detecting peaks of product ions

2. Selecting possible precursors based on retention time proximity with product ions

3. Selecting candidate precursors.
   1) Hierarchical clustering
   2) Wavelet coherence calculation

4. Plotting extracted ion chromatogram and wavelet coherence heat map

3.2. Graphic user interface (GUI)

GUI is shown as Figure 1. The functions of the four sections correspond to the four sections shown in 3.1.
4. Operation introduction

4.1. Preparation of data

There are two sets of data need to be prepared beforehand:

(1) The SWATH data after format conversion. Please download and install ProteoWizard MS Converter (http://proteowizard.sourceforge.net/downloads.shtml) suitable for your computer. After installation, import the source data (suffix .wiff or .wiff2) generated on the AB Sciex time-of-flight (TOF) mass spectrometer into MS Converter for format conversion. In the GUI of MS Converter, select: "Output format" as "text"; "Binary encoding precision" as "64bit" or "32bit"; "Filters" as "Peak picking", "Vender" and "MS Levels: 1-". Refer to Figure 2 for the selection of parameters.
(2) A list of all MS1 ions. Use the "Analytics" function on AB Sciex OS. Create a "Results" session and import the wiff2 data. When creating the processing method, select "Exhaustive" under "Non-targeted peaks". This step can find out all MS1 ions and their retention time. Or, use the "XIC Manager" function on AB Sciex Peakview. Create a new session, import the wiff data, select "Non-targeted Peak Finding" under "options", set "minimum intensity in counts", "approximate LC peak width", and "chemical noise intensity multiplier", and then click "run" to generate a list of all MS1 ions.

4.2. The installation of Precursorfinder v1.0

We put the source code of Precursorfinder v1.0 in the folder with the same name. Install MATLAB R2018a. Check Wavelet Toolbox and Bioinformatics Toolbox when installing. In the default path where the MATLAB workspace (suffix .mat) and methods (suffix .m) are stored, add in all source code files.

Enter "run precursorfinder5" or "precursorfinder5" in the MATLAB command window. The GUI (Figure 1) will appear. Use Section 1 to Section 4 in sequence. The
parameters of each section can be adjusted individually.

4.3. **SWATH data read-in**

Enter the parameters in the Section 1 to import SWATH data. This step converts the txt file obtained in 4.1 (1) into a specific format in MATLAB for subsequent use. The parameter "directory_data" is the storage path of the txt file. For example, if the txt file is stored at "D:\matlab\test\krillhex.txt", then enter "D:\matlab\test\krillhex" in "directory_data". The parameter "directory_output" is the storage path of the result. For example, if the result file is "D:\matlab\test\result.txt", then enter "D:\matlab\test\result" in "directory_output".

The parameter "swath window" is the SWATH window setting. For example, when sampling, the TOF acquisition range of MS1 is 350-450, and the range of MS/MS is m/z 350-375, 374 -400, 399-425, and 424-450. Then we need to enter the following sentence in "swath window":

```
[350 450;350 375;374 400;399 425;424 450]
```

Please keep this format, otherwise the program cannot be read.

The parameter "mz1rangemin" is the minimum m/z value of MS1. The parameter "mznrangemin" is the minimum m/z value of MS/MS.

One or two product ions can be set in the GUI in "first product ion" and "second product ion". Using two characteristic product ions to determine a parent ion can improve accuracy. Please refer to Figure 3 for all parameter settings.

![Figure 3. Section 1 parameter setting](image-url)
After entering the parameters, click the button "step1-wiff2 data read-in". The result will be displayed in the MATLAB workspace (Figure 4). The variables "Ionpool", "Timepool", "mainproductiondata", "secondproductiondata", and "sumiondata" are the results generated in this step. "Ionpool" contains all information of the original SWATH data. "Timepool" contains the time point information of each MS and MS/MS experiment. "Mainproductiondata" and "secondproductiondata" contain the data of the first and second product ions in all MS/MS experiments.

| directory_data           | 'F:\wavelet project202 |
|-------------------------|-------------------------|
| directory_output        | 'F:\wavelet project202 |
| Ionpool                 | 1x4 cell                |
| mainproduction          | 106.9120                |
| mainproductiondata      | 1x4 cell                |
| mzlrangeemin            | 100                     |
| mzrangeimin             | 50                      |
| secondproduction        | 122.9069                |
| secondproductiondata    | 1x4 cell                |
| sumiondata              | 1x4 cell                |
| swathnumber             | 3                       |
| swathwindow             | [100, 300; 100, 150, 200, 2 |
| Timepool                | 1x4 cell                |

Figure 4. Section 1 output.

4.4. Import the list of all precursor ions

In "step2", use the "Import" function of MATLAB to import the list generated in 4.1 (2). Import the numeric data in the two columns named "Retention time" and "Found at mass" to the MATLAB workspace in a numeric matrix format, and change the variable name to "PrecursorALL" (Note: Must change the variable name!).

4.5. Detecting the peaks of specified product ion

Click the button "Step-3 product ion peak detection" to perform peak detection. The program will automatically determine the number of incoming signals and call different functions. If two signals (two product ions) are imported, the user is required to select the detection mode: co-located peak "co" or superimposed peak "sup" (Figure
The co-located peaks are the peaks that the first and second product ions both have, while the superimposed peaks are the sum of all peaks of the first and second product ions.

Figure 5. Dialogue of detecting co-located peaks or superimposed peaks.

The extracted ion chromatograms (XICs) of the one/two product ion(s) in each SWATH window will be plotted, and the detected peaks will be marked on the XICs.

We provide users 9 peak detection modes. The 7 modes are composed of different value combinations of the following four parameters: number_of_decomposition, denoising_coefficient, peak_check_range, minpeakdistance.

number_of_decomposition: The number of wavelet decomposition layers. If the input signal is complex and has many peaks, it is recommended to use 6-layer wavelet decomposition. If the input signal is relatively simple and has fewer peaks, it is recommended to use 3-layer wavelet decomposition.

denoising_coefficient: If the input signal does not have a peak that deviates far from the other peaks (that is, the distribution of all peaks is relatively concentrated), it is recommended to use 0.1. If there are two peaks that are far apart from other peaks, it is recommended to use 0.01.

peak_check_range: Since the smoothed peak is often not at the same location as the original peak, it is generally necessary to set maximum detection range from the vicinity of the smoothed peak. peak_check_range is the size of the detection range.

minpeakdistance: The minimum distance between two local maximum.
Mode 1-3 are more suitable for simple spectra whereas Mode 4-8 are more suitable for complex spectra. In mode 9, users can type in values for the 4 parameters.

If users find one mode is not suitable for the peak detection when observing the XICs with naked eyes, please enter "0" in the dialogue (Figure 6), and the program will automatically go back to the previous step and ask the users to re-select the detection mode (Figure 7). Users can try different detection mode until they feel the peak detection is okay. The height of the superimposed peaks or the co-located peaks should be greater than a threshold, e.g., no less than 5% of the highest peak along the XIC. The threshold can be adjusted by the user. Users can also specifically pick out the peaks of interest from the superimposed peaks or the co-located peaks.

When okay, enter "1" to go to the next SWATH window.

Figure 6. Dialogue of determining if the peaks are correctly detected.

Figure 7. Dialogue of selecting peak detection mode.

After all SWATH window are processed, the MATLAB workspace displays the newly generated variables: "productpeaks". The retention time (unit: min) and peak width (unit: min) of peaks in each SWATH window are shown in each column of
'productpeaks". And *Precursorfinder* draws a figure containing the XICs of user-set product ions in all SWATH windows.

Figure 8. The XICs of two product ions in each SWATH window generated by Step 3 in Section 1.

4.6. Generation of possible precursor ions

Click on the "possible precursor pool" in Section 2 to filter possible precursor ions and generate their chromatographic data.

4.7. Calculate hierarchical clustering to screen candidate precursor ions

In Section 3, enter a cluster threshold in the parameter "cluster threshold", such as 1.5, and click the button "candidate generation1" to start the calculation. Under this clustering condition, the parent ion clustered with the product ion will be recorded in the variable "Candidate_cluster" in the MATLAB workspace. "Candidate_cluster" contains all candidate precursor ions for each peak in each SWATH window.

This threshold can be set as high as possible at the beginning to avoid underreport. If the threshold exceeds the range of the hierarchical clustering, MATLAB
will report an error in the command window. In this case, users can adjust the threshold to a lower value.

4.8. Calculate wavelet correlation to screen candidate precursor ions

Section 3 also calculates the wavelet coherence. Enter a correlation threshold (0 < coherence threshold < 1) in the parameter "coherence threshold", such as 0.7, and click the button "candidate generation2" to start the calculation. If the wavelet coherence of the possible precursor ion is greater than 0.7, this ion will be output to the "Candidate_coh" variable. The user can make the threshold as low as possible at the beginning to avoid underreport and adjust the threshold gradually to optimize the result.

The Candidate_coh variable contains the candidate precursor ions for each peak in each SWATH window. For each peak, the first column is the serial number of the precursor ion in the variable "PrecursorALL", the second column is the m/z of the precursor ion, the third column is the retention time of the peak (unit: min), and the fourth column is the wavelet coherence between the precursor ion and the product ion (Figure 9).

| Candidate_coh[1, 2][1, 1] |
|---------------------------|
| 1 | 2 | 3 | 4 |
|---|---|---|---|
| 1 | 1 | 106.9128 | 2.7659 | 0.7755 |
| 2 | 2 | 106.9130 | 2.7659 | 0.7755 |
| 3 | 15 | 120.9288 | 2.7659 | 0.7698 |
| 4 | 16 | 122.9079 | 2.7659 | 0.8101 |
| 5 | 17 | 122.9081 | 2.7659 | 0.8101 |
| 6 | 18 | 123.9159 | 2.7659 | 0.7731 |
| 7 | 24 | 138.9392 | 2.7659 | 0.7719 |
| 8 | 26 | 140.9187 | 2.7659 | 0.7607 |

Figure 9. Candidate_coh data structure.
4.9. Plot XICs and wavelet coherence coefficient

In order to make the results intuitive, Section 4 provides a function of drawing the XICs of parent ions, XICs of product ions, and their wavelet coherence coefficient heat map. Enter the serial number of the precursor ion in the parameter "precursor ion No.,” and click "precursor and product ion XIC & wavelet coherence plot” to draw the graph (Figure ). The left plot is the XIC of the parent ion, the middle plot is the XIC of the summed signal of first and second product ions, and the right plot is the heat map of the wavelet coherence between them. From the wavelet coherence plot, we can see that throughout the whole analysis time, the parent ion has a high correlation with the product ion in the low frequency region. They also have a high correlation in the higher frequency region around peak retention time. However, the un-selected precursor ion will not have a continuous high wavelet coherence from low frequency to high frequency (not shown in this guide).

![Figure 10. The XIC of precursor ion(left), the XIC of product ions(middle), and the wavelet coherence coefficient heat map(right).](image-url)