

Supplementary Figure S1. Peptide maps of CAM-PPL3A $(\alpha+\alpha), 3 B(\alpha+\beta)$, and $3 C(\beta+\beta)$ digested by Achromobacter protease I (A) and Staphylococcus aureus V8 protease (B). Peptides were separated by reversed-phase HPLC on COSMOSIL ${ }^{\oplus}$ Protein-R colomn ( $\varnothing 4.6 \times 250 \mathrm{~mm}$ ) using a linear gradient of acetonitrile in $0.1 \%$ trifluoroacetic acid. Common peaks between $\alpha$ and $\beta$ subunits were marked by magenta and green-colored arrow heads, respectively.


Supplementary Figure S2. Nucleotide sequences of cDNAs and the corresponding amino acid sequences of PPL3 subunits. Nucleotide residues and amino acid residues different from PPL3 $\alpha$ and $\beta$ subunits are indicated by bold characters, respectively. Peptide fragments generated by digestion with Achromobacter protease I (L) and S. aureus V8 protease (V), respectively, are indicated by lines. Italic letter with underline indicates the signal peptide region. Asterisk indicates the stop codon.


Supplementary Figure S3. Separation of CAM-PPL4 $\alpha$ and $\beta$ subunits by reversed-phase HPLC. Separation of CAM-PPL4 subunits was conducted by HPLC on a CAPCELL PAK (C8) colomn ( $\varnothing .6$ $\times 150 \mathrm{~mm}$ ) using graded linear gradient of acetonitrile in $0.1 \%$ TFA.


Supplementary Figure S4. Peptide maps of CAM-PPL4 $\alpha$ (A) and $\beta$ (B) subunits digested by Achromobacter protease I. Peptides were separated by reversed-phase HPLC on COSMOSIL ${ }^{\otimes}$ ProteinR colomn ( $\varnothing 4.6 \times 250 \mathrm{~mm}$ ) using a linear gradient of acetonitrile in $0.1 \%$ trifluoroacetic acid. Common peaks between $\alpha$ and $\beta$ subunits were marked by magenta arrow heads.

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(A) }1\mathrm{ ATGGGTGTCTATGTGTACATTGTTCTTCTAGTACCATGTCTAATGGCAATACAAGCAGAT 60
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    61 GCAAGTTGCGGAGCCCTATCAGAATCATATGGGGGTCCAGGTGGTTTA.A.CCGTTTTGAC 120
    A S C G A L S S E S Y Y G G P F G G L L N R F F D
                            L9
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    E K A L L V K N G D I K E E I E E L L L C G R R
181 GThacGGCAMTARGATTAAGATATGGCACAGTGTGGGGTACACTTCATGGTTGGAMATCC 240
    V T A A I I R I L R R Y G G T V N W G T T I H H G N K K
2 4 1 ~ C C A C C A G G A R A R A G T T G C G C A R G R G A T T G G G A T G T C G G C A G C A R A G T C A T T T A T A C A C T G ~ 3 0 0 ~
    P P G K S C A R D N D V G S K K V V I Y T L L
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    |H! P N E Y V K
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    T L K
421 RAATCAGTCGATGGCCGGCGATTARAGTATATA.ACCGGARA.CTCTGGATGTATTCTTGAC 480
    K S V D G R R L K Y I T G N S G C I L D
1 aghatachgttttactggccattgtggtak 510
    R I I Q F
(B)
```



Supplementary Figure S5. Nucleotide sequences of cDNAs and the corresponding amino acid sequences of PPL4 $\alpha$ (A) and $\beta$ (B) subunits. Peptide fragments generated by Achromobacter protease I (L) digestion are indicated by lines. Italic letter with underline indicates the signal peptide region. Asterisk indicates the stop codon.


Supplementary Figure S6. Schematic representation of oligosaccharide structures. Note that the reducing terminal is pyridylaminated for FAC analysis. Symbols used to represent pyranose rings of monosaccharides are shown in the box at the bottom. Anomeric carbon, i.e. position 1, is placed at the right side, and 2, 3, 4 are placed clockwise. Thin and thick bars represent $\alpha$-linkage and $\beta$-linkage, respectively.

Supplementary Table S1. The amino acid sequences and masses of the peptides generated by cleavage of the CAM-PPL3B with Achromobacter protease I (A) and S. aureus V8 protease (B).
(A) Achromobacter protease I

| Fragment number | Amino acid sequences |  |  | Molecular mass ( $\mathrm{m} / \mathrm{z}$ ) |  |
| :---: | :--- | :--- | :---: | :---: | :---: |
|  |  |  | Calculated | Observed |  |
| L4 | YVQSITFK |  | 984.55 | 987.10 |  |
| L5 | EIASEYLGGPGGDAFDDK | [N terminus] | 1839.84 | 1844.19 |  |
| L8 | YISGRWGCRIDGLRFHAK |  | 2192.18 | 2196.43 |  |
| L9 | VDSRQWGWANENCIQWSK |  | 2264.09 | 2269.28 |  |
| L10 | AVAQNGDITRIEMQCTDVATYIK | 2597.33 | 2601.46 |  |  |
| L11 | ALAQNGDITRIEMQCTDVATYIK | 2611.34 | 2616.13 |  |  |

(B) S.aureus V8 protease

| Fragment number |  | Molecular mass $(\mathrm{m} / \mathrm{z})$ |  |
| :---: | :--- | :---: | :---: |
|  |  | Calculated | Observed |
| V7 | NCIQWSKKGE | 1250.64 | 1250.01 |
| V8 | NCIQWSKKGEKVVHE | 1842.97 | 1842.29 |
| V9 | ITFKTNKRTLPRCGTSATE | 2182.18 | 2181.85 |
| V10 | NCIQWSKKGVKVVHE | 1813.00 | 1812.06 |
| V14 | YLGGPGGDAFDDKAVAQNGDITRIE | 2579.24 | 2579.64 |
| V16 | YLGGPGGDAFDDKALAQNGDITRIE | 2593.25 | 2593.96 |
| V17 | VATYIKLRYGKVDSRQWGWANE | 2640.37 | 2640.23 |
| V19 | MQCTDVATYIKLRYGKVDSRQWGWANE | 3276.61 | 3276.47 |
| V20 | KSVTVLIPGGLKYISGRWGCRIDGLRFHAKC | 3546.99 | 3545.02 |
| V21 | LSSGEYITSAIVTYGKYVQSITFKTNKRTLPRCGTSATE | 4329.26 | 4325.63 |

Common peptides between PPL3A $(\alpha+\alpha)$ and PPL3B $(\alpha+\beta)$, and between PPL3B $(\alpha+\beta)$ and PPL3C $(\beta+\beta)$ are indicated by magenta and green boxes, respectively.

Supplementary Table S2. The amino acid sequences and masses of the peptides generated by cleavage of the CAM-PPL4 $\alpha(\mathrm{A})$ and $\beta$ (B) subunits with Achromobacter protease I.

## (A) PPL4 $\alpha$

| Fragment number |  | Amino acid sequences |  | Molecular mass $(\mathrm{m} / \mathrm{z})$ |  |
| :---: | :--- | :--- | ---: | :---: | :---: |
|  |  | Calculated | Observed |  |  |
| L3 | SVDGRRLK | 929.56 | 932.82 |  |  |
| L4 | PNEYVK | 748.40 | 773.46 |  |  |
| L5 | TNMRELPK | 987.54 | 991.19 |  |  |
| L6 | SCARDWDVGSK | 1280.62 | 1283.11 |  |  |
| L8 | VIYTLK |  | 735.48 | 760.08 |  |
| L9 | SCGALSESYGGPGGLNRFDEK | [N terminus] | 2201.05 | 2203.56 |  |
| L10 | GATITYDRFVNSLTLK |  | 1797.99 | 1802.24 |  |
| L11 | EIELLCGRRVTAIRLRYGTVWGTLHGWK | 3340.88 | 3344.89 |  |  |
| L12 | YITGNSGCILDRIQFYWPLW | 2502.28 | 2506.58 |  |  |

(B) PPL4 $\beta$

| Fragment number | Amino acid sequences | Molecular mass $(\mathrm{m} / \mathrm{z})$ |  |  |
| :---: | :--- | ---: | ---: | ---: |
|  |  | Calculated | Observed |  |
| L4 | TNMRELPK | 987.54 | 990.42 |  |
| L6 | SCARDWDVGVK | 1292.66 | 1295.40 |  |
| L10 | VLYTLQPNEYVK | 1465.81 | 1469.29 |  |
| L11 | VCTALSESYGGFGGLNRFDENALAK | [N terminus] | 2626.31 | 2630.86 |
| L14 | GATITYDRFVNSLTLK | 1797.99 | 1802.24 |  |
| L15 | EIELLCGRRVTAIRLRYGSVWGTLHGWK | 3326.86 | 3331.88 |  |
| L18 | YITGNSGCILDRIQFYWPSW | 2476.23 | 2478.06 |  |

Common peptides between PPL4 $\alpha$ and $\beta$ subunits are indicated by magenta boxes.

Supplementary Table S3. Properties of lectin-immobilized columns used for FAC analysis.

|  | Amount of Immobilized <br> Lectin name $(\mathbf{m g} / \mathrm{ml}$ gel $)$ | $\mathbf{B t}(\mathbf{n m o l})$ | $\boldsymbol{K d} \mathbf{( M )}$ | $\mathbf{R}^{2 \boldsymbol{a}}$ | Used carbohydrate |
| :---: | :---: | :---: | :---: | :---: | :---: |
| PPL2A | 0.05 | 0.02 | $2.0 \times 10^{-7}$ | 0.996 | 1M2M-5NC-Asn Fmoc |
| PPL3 | 0.5 | 0.63 | $3.01 \times 10^{-5}$ | 0.985 | 1M2M-5NC-Asn Fmoc |
| PPL4 | 1.0 | 0.98 | $2.0 \times 10^{-5}$ | 0.996 | ManapNP |

${ }^{\text {a }}$ the coefficient of determination quantified the degree of linear correlation obtained from a Woolf-Hofstee-type plot in each concentration-dependent analysis. $B_{t}$ and $K_{d}$ values were calculated from those determined by concentration-dependent analysis.

