Table S1 Primers used for cloning components of *Ct*Xyl5A and for the mutagenesis of CtCBM62

Protein	Primers	Sequence (5'-3')
*Ct*CBM62	Forward	CTCGCTAGCTATCCTAAACTTACGG
	Reverse	CACCTCGAGATGCACATCATCATTC
CBM6-CBM13-Fn3-*Ct*CBMX	Forward	CTCGCTAGCACGGATTCGGTGAATG
	Reverse	CACCTCGAGATGCACATCATCATTC
W754A	Forward	GGAACCCAAGGTTCGGCGAATAACATTGGG
	Reverse	CCCAATGTTATTCGCCGAACCTTGGGTTCC
F772A	Forward	GGTGACCTGAACACGGCTTTTGACGGTCCT
	Reverse	AGGACCGTCAAAAGCCGTGTTCAGGTCACC
D774A	Forward	GAACACGTTTTTTGCCGGGCCCACAGCAAACGG
	Reverse	CCGTTTGCTGTGGGCCCGGCAAAAAATGTGTTC
W780A	Forward	CAGCAAACGGCTGCGCGCTGGGACTGGATTTTG
	Reverse	CAAAATCCAGTCCCAGCGCGCAGCCGTTTGCTG
D786A	Forward	TGGCTGGGACTGGCTTTTGGGGAAGGTGTG
	Reverse	CACACCTTCCCCAAAAGCCAGTCCCAGCCA
E789A	Forward	GGACTGGATTTTGGGGCAGGTGTGAGGAATGTC
	Reverse	GACATTCCTCACACCTGCCCCAAAATCCAGTCC
R803A	Forward	ATTAAATTCTGCCCGGCGTCCGGCTATGAACAG
	Reverse	CTGTTCATAGCCGGACGCCGGGCAGAATTTAAT
Y806A	Forward	GCCCGCGTTCCGGCTATGAACAGCGCATGATAG
	Reverse	CTATCATGCGCTGTTCATAGCCGGAACGCGGGC
R809A	Forward	CCGGCTATGAACAGCGCATGATAGGGGGAATT
	Reverse	AATTCCCCCTGTCATGCGCTGTTCATAGCCGG
Q816A	Forward	GGGGGAATTTTTGCGGGGGCAAATAAAG
	Reverse	CTTTATTTGCCCCCGCAAAAATTCCCCC
E85A	Forward	GGGGCAAATAAAGCAGAATTCAGCGATGC
	Reverse	GCATCGCTGAATTCTGCTTTATTTGCCCC
F821A	Forward	GCAAATAAAGAAGATGCTAGCGATGCAGTG
	Reverse	CACTGCATCGCTGGCATCTTCTTTATTTGC
R853A	Forward	TTCCGCTATGTCGCGTATTTGTCCCCGGAC
	Reverse	GTCCGGGGACAAATACGCGACATAGCGGAA
N866A	Forward	CCGGACGGCAGTGCTGGAAATATTGCA
	Reverse	TGCAATATTTCCAGCACTGCCGTCCGG

Table S2 **Crystal and structure statistics of *Ct*CBM62**

	***Ct*CBM62 SeMet**	***Ct*CBM62 Native**	***Ct*CBM62/ Xyloglucan oligosaccharide**	***Ct*CBM62/GM3**	***Ct*CBM62/arabinose**
Data collection					
Space group	$F2_3$	$F2_3$	$F2_3$	$F2_3$	$F2_3$
cell dimensions					
a, *b*, *c* (Å)	*a* = *b* = *c* = 191.39	*a* = *b* = *c* = 192.39	*a* = *b* = *c* = 191.68	*a* = *b* = *c* = 191.35	*a* = *b* = *c* = 191.79
Resolution (Å)	42.80 - 2.49 (2.62-2.49)	22.22-1.70 (1.79-1.70)	110.43-1.75 (1.80-1.75)	39.07-1.70 (1.79-1.70)	30.29-2.00 (2.10-2.00)
R_{merge}	0.159 (0.613)	0.182 (0.528)	0.140 (0.851)	0.146 (0.895)	0.121 (0.736)
$I / \sigma I$	33.9 (5.6)	10.7 (2.1)	13.2 (1.9)	14.0 (1.8)	16.9 (1.8)
completeness (%)	99.8 (98.6)	100.0 (100.0)	100.0 (100.0)	100.0 (100.0)	99.8 (99.4)
Redundancy	27.5 (22.8)	8.3 (8.2)	7.3 (7.0)	7.4 (7.2)	5.9 (5.0)
Anomalous completeness (%)	99.7 (98.0)				
Anomalous redundancy	14.0 (11.5)				
Refinement					
No. reflections	-	64559 (9370)	55596 (4087)	63488 (9198)	39402 (5705)
R_{work} / R_{free}	-	0.189/0.204	0.181/0.199	0.176/0.194	0.226/0.243
No. Atoms					
Protein	-	2254	2199	2165	2118
Ligand / Ion	-	0/2	52/2	37/2	20/2
Water	-	480	416	503	135
B-factor					
Protein	-	9	13	15	18
Ligand / Ion	-	7	36	35	33
Water	-	25	35	36	38
R.m.s.[b] deviations					
Bond lengths (Å)		0.012	0.012	0.011	0.017
Bond angles (°)		1.49	1.34	1.23	1.49

Table S3 Affinity of variants of *Ct*CBM62 for xyloglucan

Variants of *Ct*CBM62	**Mutation(s) introduced**	**Affinity (K_A x 10^4 M^{-1}) for xyloglucan**	
		5 mM calcium	**10 mM EDTA**
Wild type	No mutation	177	0.71
Mutants in the calcium binding site	D130	126	0.81
	D766A	151	1.10
	D766A/T771A	232	0.58
	D766A/T771A/E869A	102	1.2
	E869A	168	0.30
Mutants of residues at the interface of the two molecules in the asymmetric unit	K799A	91	0.54
	T840A	241	1.7
	Q871A	106	0.91
	N770A	117	0.41
Mutants of Aspartates and and Glutamates on the surface of the protein	D825A	192	3.2
	D861A	81	0.73
	D822A	100	0.81
	E821A	158	0.64
	D768A	252	0.81
	D774A	103	0.21
	D786A	79	0.45
	E805A	157	1.6
	D845A	123	0.17
	E789A	145	0.21

The affinity of the mutants for xyloglucan was determined by ITC

Figure S1 Structure of *Ct*CBM62 in complex with arabinose

Panel A displays the interactions between *Ct*CBM62 and arabinose, *Panel B* provides a surface representation of the ligand binding site occupied by arabinose, while *Panel C* is a schematic representation of the interaction of arabinose with the protein. Arabinose is coloured cyan while in *Panel A* the amino acids that interact with the ligand are shown in stick format with carbons coloured green, while in *Panel B*, the surface of these residues are also coloured green.

Figure S2 Sequence alignment of *Ct*CBM62 with related protein modules

BLAST analysis of *Ct*CBM62, corresponding to residues 739 to 885 of *Ct*Xyl5A identified five proteins (Panel A) that displayed >42 % sequence identity, which are identified by their GenBank accession numbers. ABR45148.1 (Panel B) is an example of a group of 40 proteins that display non-contiguous similarity to *Ct*CBM62. Extensive co-linear identity (~40 %) to *Ct*CBM62, which includes the conservation of the key ligand binding residues, is evident after EFI14249 has been subject to a circular permutation event (Panel C). Residues that are identical in all 10 proteins are indicted by *, while those that play a direct role in ligand recognition in *Ct*CBM62 are highlighted in green.

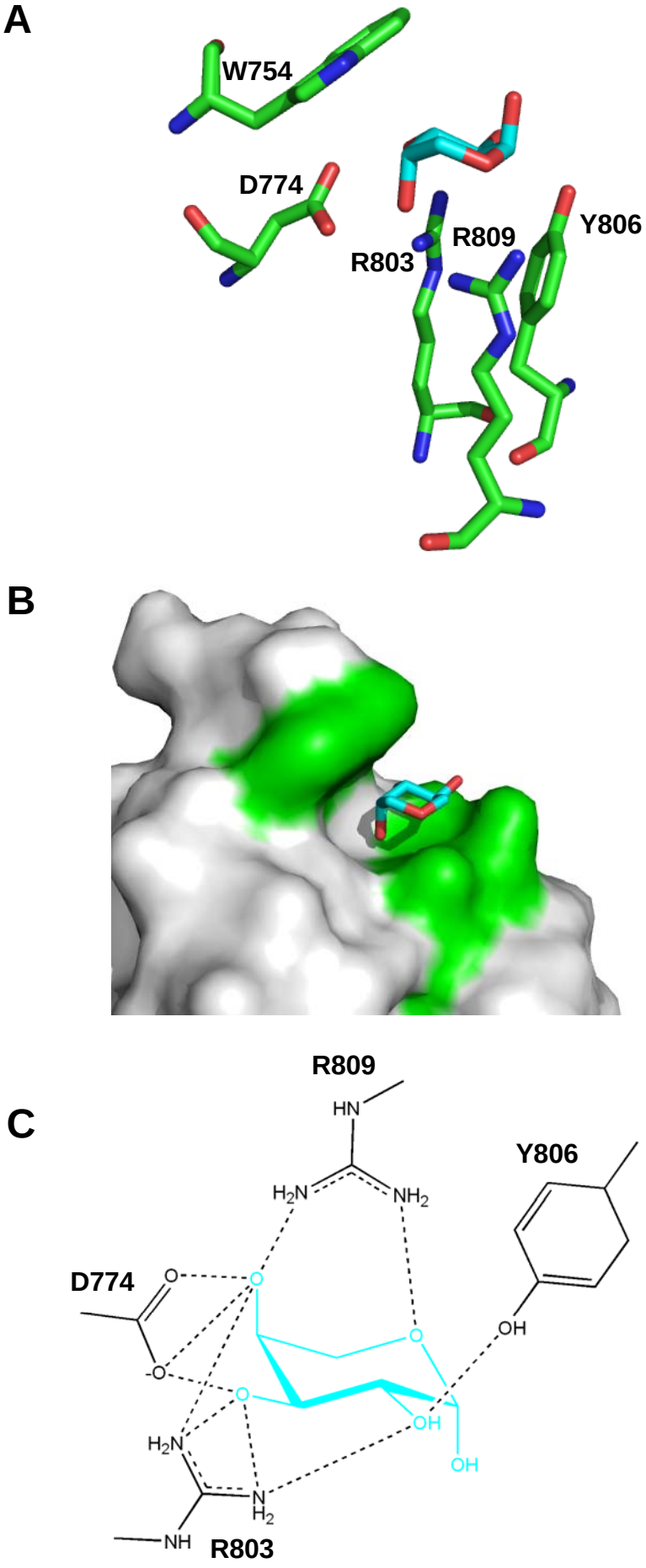
A
W754
D774
R809
R803
Y806
B
C
R809
HN
H2N
NH2
Y806
D774
O
O
O
O
OH
OH
OH
H2N
N
H2
NH
R803

Figure S1

A

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CtCBM62              YPKLTGTVIGTQGSWNN-IGNTIHKAFDGDLNTFFDGPTANGCWLGLDFGEGVRNVITQI 798
CBK73885             YPKLTGTVIGTTGSWNN-SGNTIAKIFDNDLNSYFDAAAGSGCWAGLDFGVGVSNVVMQI 793
ZP_06143050          ---LTGTIIGTSGSWNN-SGNTKEKAMDGSLTTFFDAPTGNGDWVGLDLGTNVSKVISQV 109
ABG59655             --KVTGTQFGTSGSFGNDSSTTSDKATDGNINTWSDSANANGAYTGIDLGAGNEKRIYKI 503
EEU61164             --KLQGTVIGTPGSWNG-SGNTREKAFDGNINTYFDAS-VDVAWTGLDLSATYK--VTTI 778
ABR45148             ----TARIIGTPGSWGG-MGNTCDKAMDGNIGTYFDSDVDTNAWVGLDLGSNMRATVSRI 491
                             ** **       *  *  *       *         * *

CtCBM62              KFCPRSGYEQRMIGGIFQGANKEDFSDAVTLFTITSLPGSGTLTSV-DVDNPTGFRYVRY 857
CBK73885             SYCPRPNFSQRMVGGIFQGANLADFSDAVTLFTITAQPATGVLTSA-NVTNTNTFRYVRY 852
ZP_06143050          RYCPRSSNEARMVGGKFQGANAADFSGAVDLYTITAQPVTGTLTTQ-SISVTNAFRYVRY 168
ABG59655             RYYPRNGWASRMLGGRFQGSNTSQTAGYVDLHTITTMPASGIFTEA-TVSDTTQYRYLRY 562
EEU61164             RYVPRAGLEGRMIGGKFQGSNAEDFSTYTDLATVVAKP-SFTWNCF-DVSSTASFKYLRY 836
ABR45148             GYAPRSGYASRLYGGCFQLADNKDFIDPVTFYCIDVYDTEYYVVSHREVDINKAYRYMRY 552
                        **     *  ** **                                      * **

CtCBM62              LSP-DGSNGNIAELQFFGTPAG 878
CBK73885             LSP-DGSYGNVAEIQFLGAP-- 869
ZP_06143050          LSP-DGGFGNVAEVQFY----- 184
ABG59655             VSP-DGGYCNVAELEFY----- 578
EEU61164             IGP-EGGVGNIAEIEFYGTP-- 855
ABR45148             LSSGTKSNCNISEVEFWGYP-- 571
                             *  *  *
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B

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CtCBM62    740 -YPKLTGTVIGTQGSWNNIGNT----IHKAFDGDLNTFFDGPTANGCWLGLDFG-EGVRN 793
ABR45148   466 FRDRMIGGVFEGS-NRTDFGEKDTLFIIQSRPDRLNTTVKSWSDKEYRYIRYVGPPNAHC 524
                     * *          *      *       ***                *

CtCBM62    793 VITQIKFCPRSGYEQRMIGGIFQGANKEDFSDAVTLFTITSLPGSGTLTSVDVDNPTGFR 853
ABR45148   525 NVSEIAFYEKN--DTAALSGKIIGTPGCWEHDGTHEYTN-AFDG-RTWTSFDYSEPTGGW 579
                    * *            *   *       *     *     *  * ** *   ***

CtCBM62    854 YVRYLSPDGSNGNIAELQFFGTPAG 878
ABR45148   580 TGLDLG-----RKIRIDRIVYTPRN 600
                    *        *       **
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C

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CtCBM62    740 -YPKLTGTVIGTQGSWNNIG-NTIHKAFDGDLNTFFDGPTANGCWLGLDFGEGVRNVITQIKFCPRSG 797
ABR45148   536 DTAALSGKIIGTPGCWEHDGTHEYTNAFDGRTWTSFDYSEPTGGWTGLDLGRKIR--IDR IVYTPRN-593
                    * *  *** * *   *      ****   * **     * * *** *   *  *

CtCBM62    798 YEQRMIGGIFQGANKEDFSDAVTLFTITSLPGSGTLTSVDVDNPTGFRYVRY 849
ABR45148   466 FRDRMIGGVFEGSNRTDFGEKDTLFIIQSRPDR-LNTTVKSWSDKEYRYIRY 516
                  ***** * * *  **    *** * * *     * *        ** **

CtCBM62    850 LSPDGSNGNIAELQFFGTPAG 878
ABR45148   517 VGPPNAHCNVSEIAFYEKN-- 535
                 *     *  *  *
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Figure 2S