Table S1. Listing of Proteins Identified in CMT3 Purifications Filtered at 1% of CMT3, Related to Figure 1 and Table 1

Proteins present in both CMT3 and wild-type samples were filtered at 10% of CMT3 after subtraction of the protein amounts present in WT. [1]Unique peptides map to the group of these genes.

Protein	AGI Code	Spectra	Unique Peptides	%Coverage	NSAFe5	% CMT3
CMT3	At1g69770	756	131	63.4	7240	100
H1	At1g06760	19	11	31.8	557	7.7
H1	At3g18035	29	17	22.3	485	6.7
H1/H5	At1g48620	28	12	17.3	470	6.5
H1.2	At2g30620	46	16	47.3	1353	18.7
HTA13	At3g20670	50	7	21.2	3043	42.0
HTA6	At5g59870	18	4	17.3	964	13.3
HTA9	At1g52740	15	4	30.6	899	12.4
HTA7	At5g27670	15	3	8.0	803	11.1
HTA12	At5g02560	5	2	7.8	262	3.6
HTA8	At2g38810	15	4	32.4	251	3.5
H2B[1]	At5g02570 At3g09480	59	13	32.6	3675	50.8
H2B	At2g37470	62	15	46.4	3610	49.9
H2B	At3g53650	32	8	30.4	1863	25.7
H2B[1]	At5g59910 At1g07790 At5g22880 At3g45980 At3g46030	79.6	16.6	42.2	4327	59.8
H3[1]	At5g10400 At1g09200 At5g10390 At5g65360 At3g27360	63	21	52.9	3674	50.7
HTR12	At1g01370	2	2	25.3	90.3	1.2
H4[1]	At1g07820 At3g3730 At2g28740 At5g59970 At3g46320 At3g45930 At5g59690 At1g07660	26	13	62.1	2028	28.0
Ku80	At1g48050	24	19	33.8	284	3.9
Ku70	At1g16970	10	8	16.7	129	1.8

	At5g25980	204	50	62	3205	44.3
	At5g62390	67	34	43.7	1207	16.7
	At4g25210	51	26	36.4	1114	15.4
RPL32	AtCg01020	3	2	30.8	463	6.4
60S[1]	At1g15250 At1g52300 At3g16080	4	2	17.9	338	4.7
60S[1]	At2g25210 At4g31985	2	2	25.4	338	4.7
RPS18[1]	AtCg00650	4	2	20.8	318	4.4
	At2g27402	2	2	32.7	309	4.3
40S[1]	At3g53890 At5g27700	2	2	19.2	192	2.7
RPS15	AtCg01120	2	2	25	183	2.5
40S[1]	At5g02960 At3g09680	3	3	21.1	170	2.3
	At4g29410	3	3	15.4	169	2.3
CIP4	At5g37190	18	2	3.2	165	2.3
	At1g02813	3	2	16.1	162	2.2
	At2g38270	6	4	10.7	161	2.2
	At2g38270	5	2	6.5	137	1.9
	At2g19740 At5g56710	2	2	16.8	135	1.9
	At5g1590	7	5	19.8	134	1.9
	At1g31310	6	4	14.6	126	1.7
	At3g12390	3	2	13.8	119	1.6
	At2g32650 At2g32180	2	2	14.4	116	1.6
	At5g06910	4	4	21.8	113	1.6
	At5g67510	2	2	14.4	110	1.5
	At3g54090	6	5	20.0	102	1.4
	At4g20130	6	5	15.7	100	1.4
	Atcg00190	13	8	11.5	97	1.3
	At3g07050	7	2	4.5	96	1.3
	At5g40950	2	2	14.1	81	1.1
	At1g69200	6	6	11.7	78	1.1
	At5g42960	2	2	6.6	75	1.0

Table S2. Summary of Proteins Associated with CMT3 after Benzonase Treatment (Top) and Partial Proteins Associated with Chromo Mutant CMT3chr3 (Bottom) Identified by Mass Spectrometric Analyses, Related to Figures 1 and 6

The percentage of CMT3 (% CMT3) column indicates the approximate stoichiometry of each co-purifying protein as a function of the Normalized Spectral Abundance Factor (NSAF). Listing of proteins were filtered at 1% of CMT3.

CMT3 Purification after Benzonase Treatment

Name	AGI Code	Spectra	Unique Peptides	%Coverage	NSAFe5	%CMT3
CMT3	At1g69770	508	57	39.5	3957	100
H1.2	At2g30620	4	3	16.1	95	2.4

CMT3chr3 Purification

Name	AGI Code	Spectra	Unique Peptides	%Coverage	NSAFe5	%CMT3
CMT3	At1g69770	277	54	41.8	1472	100

Table S3. List of Primers Used for Cloning CMT3, Bisulfite Sequencing, and Methyltransferase Activity Assay, Related to Figure 3

	Primer numbers	Primer sequences from 5’ to 3’
Full length genomic cloning into pENTR/D	JP5048 JP5049	CACCATGCATACATATAGCTTCACAAATACCCG TGCAAGCTCGGAAGGAAGAGTTGGCTTCATGAATGC
Point mutation C460A	JP4026 JP4027	GTGGGCCACCAGCCCAAGGAATCAGTGGTC GACCACTGATTCCTTGGGCTGGTGGCCCAC
Point mutation F382A	JP2061 JP2062	GATGATGGAGAAGTTGCGACTGTTGACAAGATTGTTG CAACAATCTTGTCAACAGTCGCAACTTCTCCATCATC
Point mutation W409L	JP2073 JP2074	CTTTATTTGAAGGTAAGGCTGCTGAATTATGATGATTCTCATGAT ATCATGAGAATCATCATAATTCAGCAGCCTTACCTTCAAATAAAG
Point mutation Y412A	JP2069 JP2070	GGTAAGGTGGCTGAATGCGGATGATTCTCATG CATGAGAATCATCCGCATTCAGCCACCTTACC
Full length cDNA cloning into pVIC	JP3946 JP4286	CCGGGGTACCGAATTCTGCAAGCTCGGAAGGAAGA CCGGCTCGAGGAATTCTGCAAGCTCGGAAGGAAGA
Full length cDNA cloning to pET-SUMO	JP6093 JP6094	ATGGCGCCGAAGCGAAAGAGACC ACAGTGTTCATGCAAGCTCGGAAG
Ta3 bisulfite sequencing	JP1274 JP1269	CCACTRATTCCTRAAACACAACATTTCTRCTRATA GAGAATYAGGTTAATAAGAAAGTGAAGTGTT
Ta3 hairpin bisulfite sequencing	JP3200 JP1615	AATCCAATTCCRATTTAATATTTTCTAATAATATTCRATTTAA CGGTTTAATGTTGGTTTAGTGATATTYGGTTTAGT
unmethylated oligo	JP3010 JP3011	AACGCAGCATGCGCTGCTAGCGCAGCTAGCGCTGCATG AACATGCAGCGCTAGCTGCGCTAGCAGCGCATGCTGCG
hemimethylated oligo	JP3010 JP3018	AACGCAGCATGCGCTGCTAGCGCAGCTAGCGCTGCATG AAMGMAGMATGMGMTGMTAGMGMAGMTAGMGMTGMATG
Fully-methylated oligo	JP3018 JP3019	AAMGMAGMATGMGMTGMTAGMGMAGMTAGMGMTGMATG AAMATGMAGMGMTAGMTGMGMTAGMAGMGMATGMTGMG
CHG methylated oligo	JP3014 JP3015	AACGMAGCATGCGMTGCTAGCGMAGCTAGCGMTGCATG AACATGMAGCGCTAGMTGCGCTAGMAGCGCATGMTGCG

Table S4. Summary of Arabidopsis Histone H3 Proteins Copurified with CMT3, Related to Figure 3

CMT3 specifically pulls down centromeric histone and histone H3.1, but not H3.3. [1]Unique peptides map to the group of these genes.

AGI Code	H3 type	# of unique peptides associated with CMT3
At1g01370	HTR12	2
At1g09200	H3.1	21^1
At3g27360	H3.1	
At5g10390	H3.1	
At5g10400	H3.1	
At5g65360	H3.1	
At5g65350	H3.1-like	0
At4g40030	H3.3	0^1
At4g40040	H3.3	
At5g10980	H3.3	
At1g13370	H3.3-like	
At1g19890	H3.3-like	
At1g75600	H3.3-like	
At1g75610	H3.3-like	
At5g12910	H3.3-like	

Table S5. Summary of X-Ray Diffraction Data and Structure Refinement Statistics, Related to Figures 4 and 5

Summary of Diffraction Data

Crystal	Se-ZMET2 + SAH	ZMET2 + SAH + H3(1-15)K9me2	ZMET2 + SAH + H3(1-32)K9me2
Beamline	APS-24ID-E	APS-24ID-E	APS-24ID-E
PDB code	4FSX	4FT2	4FT4
Wavelength (Å)	0.9792	0.9792	0.9792
Space group	*P*1	*P*1	$P2_1$
Cell parameters			
a (Å)	64.9	64.9	64.6
b (Å)	89.0	88.7	111.6
c (Å)	113.5	113.4	151.5
α (°)	93.5	93.2	90
β (°)	95.5	95.7	102.0
γ (°)	110.4	110.7	90
Resolution (Å)	50.0-3.2 (3.31-3.20) [a]	50.0-3.2 (3.31-3.20)	50.0-2.7 (2.80-2.70)
R_{merge} (%)	17.5 (59.5)	11.8 (50.3)	8.8 (50.7)
Observed reflections	143,259	133,229	210,454
Unique reflections	38,702	38,805	57,221
Redundancy	3.7 (3.6)	3.4 (3.4)	3.7 (3.7)
Average $I/\sigma(I)$	7.2 (1.6)	12.6 (2.2)	21.2 (2.4)
Completeness (%)	99.2 (98.8)	99.0 (98.8)	98.6 (98.4)
Refinement and Structure Model			
R factor / Free *R* factor (%)	24.0 / 26.3	23.5 / 27.7	20.6 / 24.8
Number of non-H atoms	10,631	10,657	11,265
Protein / Peptide	10,579 / -	10,549 / 56	10,957 / 96
Ligand	52	52	52
Water	-	-	160
Average B factor (Å^2)	70.7	76.2	73.6
Protien / Peptide	70.8 / -	76.2 / 92.3	73.6 / 122.5
Ligand	57.3	61.5	50.7
Water	-	-	49.7
RMS deviations			
Bond lengths (Å)	0.013	0.009	0.009
Bond angles (°)	1.819	1.575	1.275

[a] Values in parentheses are for highest-resolution shell.