

**S2 Table. (a) Optimal parameter settings and corresponding calibration statistics for matching each pair of datasets.**

Matched datasets	Matching method	Parameter settings					# of matched signals	Shared known matching calibration statistics					
		Adduct ion	Correlation	Correlation cutoff	Partition	Match type		# of matches	# correct	# $r^2 > 0.8$	% correct	% $r^2 > 0.8$	% $r^2 > 0.8$ for incorrect
OE-MCDS	IMP	Adduct	All correlation	None	Within method	Multiple	4432 <sup>a,c</sup>	166	97	151	58.4	91.0	78.3
						Reciprocal	1573 <sup>a,b,c</sup>	83	58	76	69.9	91.6	72.0
	RT	Combined	N/A	N/A	Within method	Multiple	4432	166	94	151	56.6	91.0	79.2
						Reciprocal	1983	152	81	138	53.3	90.8	80.3
OE-BioAge	IMP	Combined	Dataset 1 correlation	None	Within method	Multiple	7654 <sup>d,e</sup>	252	216	237	85.7	94.0	58.3
						Reciprocal	3879	241	211	231	87.6	95.9	66.7
	RT	Combined	N/A	N/A	Within method	Multiple	7654	252	215	224	85.3	88.9	24.3
						Reciprocal	3824	234	206	213	88.0	91.0	25.0
MCDS-BioAge	IMP	Combined	All correlation	None	Within method	Multiple	4818 <sup>e</sup>	178	155	171	87.1	96.1	69.6
						Reciprocal	2954	169	148	163	87.6	96.4	71.4
	RT	Combined	N/A	N/A	Within method	Multiple	4818	178	163	168	91.6	94.4	33.3
						Reciprocal	3025	173	159	163	91.9	94.2	28.6

**S2 Table. (b) Optimal parameter settings and corresponding calibration statistics for cross-method matching.**

Matched datasets	Matching method	Parameter settings					# of matched signals	Shared known matching calibration statistics					
		Adduct ion	Correlation	Correlation cutoff	Partition	Match type		# of matches	# correct	# $r^2 > 0.8$	% correct	% $r^2 > 0.8$	% $r^2 > 0.8$ for incorrect
OE-MCDS	IMP	Adduct	All correlation	None	Across method	Multiple	7911	170	94	155	58.9	88.4	71.8
						Reciprocal	1875	76	54	69	71.1	90.8	68.2
	RT	Combined	N/A	N/A	Across method	Multiple	7911	170	75	142	44.1	83.5	70.5
						Reciprocal	2385	135	55	115	40.7	85.2	75.0
OE-BioAge	IMP	Combined	Dataset 1 correlation	None	Across method	Multiple	9579	252	201	235	79.8	93.3	66.7
						Reciprocal	4041	240	195	228	81.3	95.0	73.3
	RT	Combined	N/A	N/A	Across method	Multiple	9579	252	200	210	79.4	83.3	19.2
						Reciprocal	3994	223	187	194	83.9	87.0	19.4
MCDS-BioAge	IMP	Combined	All correlation	None	Across method	Multiple	5251	178	148	170	83.1	95.5	73.3
						Reciprocal	3004	158	137	151	86.7	95.6	66.7
	RT	Combined	N/A	N/A	Across method	Multiple	5251	178	141	146	79.2	82.0	13.5
						Reciprocal	3042	149	122	123	81.9	82.6	3.7

"# of matched signals": number of unknown or unshared known signals matched; "# of matches": number of shared known metabolites matched; "# correct" and "% correct": number and percentage of correct matches; "#  $r^2 > 0.8$ " and "%  $r^2 > 0.8$ ": number and percentage of matches strongly correlated ( $r^2 > 0.8$ ) with the correct known in observed data; "%  $r^2 > 0.8$  for incorrect": percentage of incorrect matches strongly correlated with the correct known. "IMP": m/z and imputation-based matching; "RT": m/z and retention time-based matching; "N/A": not applicable. See Methods for detailed parameter explanations. Analyses that used the matching results: <sup>a</sup> matching validation; <sup>b</sup> GWAS replication and meta-analysis; <sup>c</sup> BMI-associated signal replication; <sup>d</sup> pathway validation; <sup>e</sup> BMI pathway analysis.