



Article

Chemical Characterization and Source Apportionment of PM₁₀ Using Receptor Models over the Himalayan Region of India

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Table S1. S/N of species and MDL used for calculation of uncertainty at different study sites.

Species	Mohal-Kullu		Nainital		Darjeeling		MDL ($\mu\text{g m}^{-3}$)
	S/N	Category	S/N	Category	S/N	Category	
OC	6.05	Strong	5.54	Strong	6.0	Strong	0.87
EC	9.09	Strong	4.27	Strong	4.4	Strong	0.38
WSOC	5.64	Weak	8.00	Weak	7.4	Weak	0.4
Na ⁺	6.54	Weak	1.05	Weak	4.1	Strong	0.34
NH ₄ ⁺	9.63	Strong	6.65	Strong	5.2	Strong	0.27
K ⁺	7.97	Strong	0.84	Weak	1.8	Strong	0.32
Mg ²⁺	-	-	-	-	0.4	Weak	0.18
Ca ²⁺	-	-	-	-	2.0	Strong	0.31
F ⁻	6.52	Strong	0.05	Weak	0.1	Bad	0.10
Cl ⁻	8.85	Strong	0.47	Weak	1.4	Strong	0.26
NO ₃ ⁻	8.33	Strong	6.46	Strong	3.3	Strong	0.37
SO ₄ ²⁻	9.07	Strong	8.60	Strong	8.8	Strong	0.23
B	7.04	Strong	9.67	Strong	9.6	Strong	0.01
Al	8.76	Strong	6.72	Strong	9.7	Strong	0.18
P	6.30	Strong	2.87	Strong	3.7	Strong	0.05
S	8.15	Weak	8.11	Weak	7.4	Weak	0.12
Ti	-	-	4.72	Strong	2.5	Strong	0.03
Cr	0.05	Weak	9.07	Strong	6.1	Strong	0.02

Fe	4.88	Strong	8.60	Strong	7.2	Strong	0.07
Ni	-	-	2.45	Strong	3.2	Strong	0.02
Cu	-	-	7.83	Strong	3.1	Strong	0.01
Zn	6.99	Strong	1.01	Weak	1.7	Strong	0.06
Zr	0.00	Weak	6.24	Bad	3.2	Strong	0.01
Mo	-	-	5.47	Strong			0.01
Mn	-	-	2.67	Strong	-	-	0.02
Br	-	-	4.67	Strong	-	-	0.01
Mg	6.68	Strong	6.46	Strong	-	-	0.03
Ca	8.72	Strong	7.62	Strong	-	-	0.03

Table S2. Model summary and the results of error estimates at Mohal-Kullu PMF.

Model summary and the result of BS, DISP and BS-DISP error estimates										
No. of base runs: 100										
Base random seed: 13										
Number of factors: 4										
Extra modeling uncertainty (%): 5										
Base run summary table:										
Run #	Q(Robust)	Q(True)	Converged	# Steps	Q(true)/Qexp					
1	1124.29	1124.29	Yes	189	2.007					
BS-DISP Diagnostics:										
# of Cases Accepted: 94										
% of Cases Accepted: 94%										
Largest Decrease in Q: -0.41										
%dQ: -0.04										
# of Decreases in Q: 0										
# of Swaps in Best Fit: 0										
# of Swaps in DISP: 6										
Swaps by Factor: 0 0 0 0 0										
DISP Diagnostics:										
Error Code: 0										
Largest Decrease in Q: -0.056										
%dQ: -0.0049										
Swaps by Factor: 0 0 0 0 0										
BS Mapping:										
Base Factor 1 Base Factor 2 Base Factor 3 Base Factor 4 Unmapped										
Boot Factor 1	100	0	0	0	0					
Boot Factor 2	0	98	2	0	0					
Boot Factor 3	0	0	100	0	0					
Boot Factor 4	0	0	0	100	0					

Table S3. Model summary and the results of error estimates in PMF at Nainital.

Table S4. Model summary and the results of error estimates in PMF at Darjeeling.

Model summary and the result of BS, DISP, and BS-DISP error estimates						
Number of base runs:	100					
Base random seed:	73					
Number of factors:	5					
Extra modeling uncertainty (%):	2					
Base run summary table:						
Run #	Q(Robust)	Q(True)	Converged	# Steps	Q(true)/Qexp	
1	2099.47	2099.47	Yes	805	1.1922	
BS-DISP Diagnostics:						
# of Cases Accepted:		99				
% of Cases Accepted:		99%				
Largest Decrease in Q:		-5.781000137				
%dQ:		-0.275361773				
# of Decreases in Q:		0				
# of Swaps in Best Fit:		0				
# of Swaps in DISP:		1				
Swaps by Factor:		0	0	0	0	0
DISP Diagnostics:						
Error Code:		0				
Largest Decrease in Q:		-0.005				
%dQ:		-0.000238161				
Swaps by Factor:		0	0	0	0	0
BS Mapping:						
	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Unmapped
Boot Factor 1	100	0	0	0	0	0
Boot Factor 2	0	100	0	0	0	0
Boot Factor 3	0	0	99	1	0	0
Boot Factor 4	0	0	0	100	0	0
Boot Factor 5	0	0	0	0	100	0

Table S5. Annual and seasonal average concentration of PM₁₀ and its elemental composition at study sites of IHR.

Val- ues are in μg m^{-3}	Mohal-Kullu					Nainital					Darjeeling				
	Annual	Winter (Jan- Feb)	Sum- mer (Mar- May)	Mon- soon (Jun- Sept)	Post- mon- soon (Oct- Dec)	Annual	Winter (Jan- Feb)	Sum- mer (Mar- May)	Mon- soon (Jun- Sept)	Post- mon- soon (Oct- Dec)	Annual	Winter (Jan- Feb)	Sum- mer (Mar- May)	Mon- soon (Jun- Sept)	Post- mon- soon (Oct- Dec)
Samples (n)	n = 76	n = 5	n = 8	n = 34	n = 29	n = 86	n = 17	n = 27	n = 9	n = 33	n = 134	n = 25	n = 32	n = 31	n = 46
PM ₁₀	58 ± 32	51 ± 16	52 ± 15	44 ± 26	76 ± 36	62 ± 39	38 ± 9	100 ± 50	49 ± 17	47 ± 14	54 ± 18	52 ± 18	63 ± 21	47 ± 9	50 ± 15
B	1.73 ± 1.13	1.41 ± 0.24	1.26 ± 0.35	1.54 ± 1.31	2.15 ± 1.03	0.4 ± 0.3	0.22 ± 0.16	0.51 ± 0.38	0.65 ± 0.34	0.31 ± 0.12	0.37 ± 0.23	0.29 ± 0.19	0.52 ± 0.26	0.38 ± 0.26	0.24 ± 0.20
Na	2.64 ± 1.67	2.19 ± 0.37	2.07 ± 0.62	2.38 ± 1.92	3.22 ± 1.55	0.36 ± 0.33	0.53 ± 0.31	0.28 ± 0.19	0.22 ± 0.19	0.35 ± 0.43	0.05 ± 0.04	0.05 ± 0.03	0.08 ± 0.05	0.05 ± 0.05	0.03 ± 0.04
Mg	0.15 ± 0.10	0.13 ± 0.10	0.22 ± 0.10	0.12 ± 0.10	0.17 ± 0.09	0.42 ± 0.49	0.17 ± 0.08	0.83 ± 0.60	0.62 ± 0.55	0.16 ± 0.13	0.14 ± 0.15	0.20 ± 0.27	0.18 ± 0.14	0.09 ± 0.14	0.11 ± 0.08
Al	1.22 ± 0.70	1.08 ± 0.38	1.26 ± 0.38	1.05 ± 0.81	1.44 ± 0.62	1.65 ± 1.73	0.67 ± 0.32	3.07 ± 2.06	2.54 ± 1.91	0.75 ± 0.54	0.85 ± 0.75	0.78 ± 0.54	0.91 ± 0.64	1.06 ± 1.37	0.72 ± 0.27
P	0.01 ± 0.001	0.006 ± 0.005	0.009 ± 0.004	0.005 ± 0.004	0.006 ± 0.004	0.05 ± 0.06	0.01 ± 0.008	0.10 ± 0.07	0.05 ± 0.03	0.02 ± 0.01	0.06 ± 0.30	0.21 ± 0.30	0.03 ± 0.67	0.02 ± 0.02	0.02 ± 0.01
S	1.02 ± 1.01	0.91 ± 0.37	1.20 ± 0.33	1.13 ± 1.43	0.84 ± 0.38	1.36 ± 0.71	1.13 ± 0.57	1.66 ± 0.66	1.80 ± 0.97	1.09 ± 0.60	1.12 ± 0.72	1.27 ± 0.76	1.67 ± 0.75	0.66 ± 0.75	0.97 ± 0.47
Cl	0.44 ± 0.59	0.64 ± 0.22	0.23 ± 0.11	0.20 ± 0.16	0.78 ± 0.84	0.40 ± 0.44	0.58 ± 0.42	0.358 ± 0.40	0.09 ± 0.009	0.43 ± 0.51	0.12 ± 0.13	0.17 ± 0.22	0.07 ± 0.05	0.08 ± 0.07	0.13 ± 0.10
K	0.91 ± 0.55	0.73 ± 0.20	0.84 ± 0.23	0.81 ± 0.65	1.10 ± 0.48	1.30 ± 1.09	0.56 ± 0.26	2.20 ± 1.36	1.62 ± 1.06	0.86 ± 0.39	0.60 ± 0.37	0.71 ± 0.30	0.78 ± 0.44	0.29 ± 0.21	0.62 ± 0.32
Ca	0.49 ± 0.29	0.39 ± 0.14	0.50 ± 0.15	0.42 ± 0.34	0.58 ± 0.26	1.81 ± 1.88	0.88 ± 0.63	3.32 ± 2.14	2.22 ± 1.93	0.95 ± 1.16	0.57 ± 0.38	0.53 ± 0.21	0.65 ± 0.45	0.41 ± 0.25	0.62 ± 0.43
Ti						0.11 ± 0.09	0.05 ± 0.02	0.19 ± 0.11	0.19 ± 0.07	0.05 ± 0.03	0.06 ± 0.02	0.06 ± 0.01	0.07 ± 0.01	0.05 ± 0.03	0.05 ± 0.02
Cr	0.01 ± 0.001	0.004 ± 0.001	0.006 ± 0.001	0.005 ± 0.004	0.007 ± 0.003	0.13 ± 0.01	0.14 ± 0.01	0.11 ± 0.02	0.13 ± 0.008	0.13 ± 0.01	0.15 ± 0.10	0.09 ± 0.22	0.13 ± 0.01	0.13 ± 0.03	0.13 ± 0.02
Fe	0.19 ± 0.13	0.16 ± 0.12	0.28 ± 0.11	0.16 ± 0.12	0.22 ± 0.11	1.42 ± 1.16	0.73 ± 0.29	2.41 ± 1.27	2.01 ± 1.31	0.80 ± 0.49	0.61 ± 0.26	0.62 ± 0.23	0.70 ± 0.39	0.49 ± 0.21	0.60 ± 0.15
Ni	-	-	-	-	-	0.06 ± 0.02	0.03 ± 0.004	0.10 ± 0.36	0.03 ± 0.003	0.03 ± 0.006	0.04 ± 0.01	0.05 ± 0.02	0.04 ± 0.01	0.04 ± 0.01	0.03 ± 0.10
Cu	-	-	-	-	-	0.13 ± 0.01	0.04 ± 0.03	0.23 ± 0.18	0.15 ± 0.07	0.09 ± 0.07	0.03 ± 0.02	-	-	0.04 ± 0.02	0.02 ± 0.003
Zn	0.29 ± 0.18	0.23 ± 0.06	0.26 ± 0.07	0.26 ± 0.21	0.34 ± 0.14	0.08 ± 0.07	0.10 ± 0.07	0.07 ± 0.08	0.03 ± 0.009	0.09 ± 0.06	0.09 ± 0.03	0.09 ± 0.03	0.08 ± 0.03	0.08 ± 0.02	0.09 ± 0.03
Zr	0.005 ± 0.004	0.001 ± 0.001	0.001 ± 0.001	0.001 ± 0.001	0.001 ± 0.001	0.0001 ± 0.0001	0.001 ± 0.001	0.11 ± 0.11	0.02 ± 0.03	0.21 ± 0.10	0.17 ± 0.11	0.06 ± 0.09	0.04 ± 0.01	0.05 ± 0.01	0.04 ± 0.009

Mo	-	-	-	-	-	0.05 ± 0.06	0.04 ± 0.01	0.07 ± 0.11	0.03 ± 0.01	0.04 ± 0.01	0.05 ± 0.06	0.06 ± 0.01	0.05 ± 0.01	0.06 ± 0.01	0.05 ± 0.01
Pb	-	-	-	-	-	0.06 ± 0.02	0.05 ± 0.008	0.07 ± 0.04	0.06 ± 0.009	0.05 ± 0.08	0.06 ± 0.02	-	-	0.12 ± 0.13	0.21 ± 0.23
Mn	-	-	-	-	-	0.08 ± 0.14	0.05 ± 0.04	0.11 ± 0.19	0.08 ± 0.05	0.06 ± 0.16	0.08 ± 0.14	0.05 ± 0.01	0.05 ± 0.02	0.04 ± 0.02	0.04 ± 0.02
Ba	1.13 ± 0.69	0.90 ± 0.22	1.03 ± 0.28	1.01 ± 0.83	1.35 ± 0.58	-	-	-	-	-	-	-	-	-	-

average ± Standard deviation, all values are in $\mu\text{g m}^{-3}$.

Table S6. Diagnostic mass ratios of carbonaceous and water soluble inorganic ionic components of PM_{10} at different study sites.

	Mohal-Kullu				Nainital				Darjeeling				Post Mon- soon		
	An- nual	Win- ter	Sum- mer	Mon- soon	Post Mon- soon	An- nual	Win- ter	Sum- mer	Mon- soon	Post Mon- soon	An- nual	Win- ter	Sum- mer	Mon- soon	
OC/EC	2.95	2.61	2.98	2.84	3.14	3.44	2.33	3.84	3.4	3.70	2.60	2.04	1.74	3.04	3.21
WSOC/OC	0.68	0.55	0.42	0.82	0.60	0.72	0.74	0.75	0.7	0.68	0.68	0.74	0.61	0.59	0.76
SOC/OC	0.30	0.14	0.31	0.25	0.34	0.43	0.23	0.50	0.4	0.42	0.43	0.34	0.23	0.42	0.49
nss K ⁺ /OC	0.05	0.03	0.03	0.05	0.05	0.09	0.10	0.09	0.12	0.09	0.10	0.10	0.13	0.08	0.10
nss K ⁺ /EC	0.14	0.08	0.08	0.16	0.15	0.31	0.23	0.31	0.39	0.32	0.26	0.21	0.22	0.22	0.33
Cl ⁻ /Na ⁺	0.76	0.77	0.56	0.77	0.81	0.97	1.23	1.06	0.63	0.85	0.50	0.51	0.61	0.48	0.44
NH ₄ ⁺ / SO ₄ ²⁻	0.68	0.73	0.63	0.63	0.75	0.45	0.45	0.44	0.37	0.49	0.50	0.62	0.58	0.41	0.45
NO ₃ ⁻ / SO ₄ ²⁻	0.97	1.17	1.00	0.92	0.99	0.52	0.53	0.53	0.52	0.51	0.50	0.49	0.51	0.37	0.57
Mg ²⁺ /Ca ²⁺	0.32	0.27	0.10	0.55	0.11	0.59	0.18	1.23	0.94	0.18	0.23	0.20	0.21	0.19	0.28
K ⁺ / NH ₄ ⁺	0.34	0.28	0.35	0.31	0.38	0.33	0.39	0.30	0.40	0.31	0.77	0.40	0.29	0.62	1.39
Cl ⁻ /NO ₃ ⁻	0.80	0.73	0.71	0.77	0.87	0.18	0.23	0.21	0.07	0.17	0.34	0.28	0.32	0.47	0.31
Ca ²⁺ / NO ₃ ⁻	0.66	0.78	0.69	0.59	0.71	0.47	0.41	0.50	0.40	0.48	0.50	0.47	0.51	0.69	0.38
K ⁺ / Ca ²⁺	0.59	0.26	0.36	0.78	0.49	2.94	1.15	6.17	3.08	1.17	1.70	1.76	0.91	0.55	2.99
K ⁺ / SO ₄ ²⁻	0.22	0.21	0.23	0.17	0.28	0.12	0.12	0.13	0.13	0.12	0.22	0.24	0.13	0.11	0.35
K ⁺ /NO ₃ ⁻	0.22	0.18	0.22	0.18	0.27	0.25	0.27	0.24	0.24	0.25	0.51	0.44	0.28	0.34	0.84
K ⁺ / Cl ⁻	0.29	0.25	0.42	0.25	0.31	2.37	1.87	2.08	3.52	2.55	2.98	3.98	1.76	1.01	4.62

Table S7. PCA factor loadings of PM₁₀ at different locations of IHR.

Table S8. Source composition of PM₁₀ sources by UNMIX at different study sites.

	Mohal-Kullu				Nainital					Darjeeling					
	1	2	3	4	1	2	3	4	5	1	2	3	4	5	
OC	-0.18	3.14	4.21	1.47	0.39	0.30	1.18	1.34	1.39	0.36	-0.53	0.88	3.47	0.83	
EC	-0.18	2.03	1.11	0.61	0.28	-0.18	0.16	0.59	0.72	0.12	0.12	0.49	1.18	0.29	
WSOC	0.51	1.80	1.15	0.79	0.56	-0.01	0.58	1.36	0.85	0.43	-0.65	0.39	2.95	0.42	
B	0.16	0.72	0.24	0.43						0.02	0.14	0.21	-0.17	0.13	
Al	0.07	0.40	0.30	0.39	-0.07	0.03	1.12	0.27	0.08	0.08	0.09	0.05	0.13	0.47	
P										0.002	0.003	0.003	0.003	0.02	
S															
Ti							0.003	0.08	0.01	0.01	0.001	0.01	0.01	0.01	0.02
Fe	0.01	0.02	0.08	0.09	-0.001	-0.06	0.80	0.23	0.25	0.04	0.05	0.13	0.11	0.29	
Zn	0.02	0.10	0.06	0.09	0.003	0.03	0.01	-0.04	0.08	-0.002	0.001	0.02	0.03	0.03	
Zr	0.00	0.00	0.00	0.00	0.11	-0.01	0.01	0.01	-0.01	-0.004	0.004	0.03	0.00	0.01	
Cr	0.00	0.00	0.00	0.00						-0.004	-0.01	0.05	0.05	0.03	
Mg					-0.002	0.01	0.34	0.01	0.01	0.02	0.02	0.02	0.01	-0.01	0.08
Ca					-0.22	0.14	1.61	-0.15	0.25	0.08	0.03	0.06	0.09	0.30	
Mo					0.01	0.01	0.002	-0.01	0.02	-0.004	0.002	0.03	0.01	0.02	
Na ⁺	0.16	0.66	0.52	0.51	-0.05	0.37	0.07	0.06	-0.03	0.04	0.07	0.57	0.16	0.65	
NH ₄ ⁺	0.02	0.55	0.16	1.04	0.20	0.03	0.44	0.07	0.92	0.03	2.87	0.07	-0.24	0.29	
K ⁺	0.02	0.51	-0.03	-0.04	0.03	0.02	0.10	0.06	0.21	0.12	-0.03	0.11	0.42	0.02	
Mg ²⁺	0.21	-0.02	0.01	0.01											
Ca ²⁺	0.21	0.98	0.12	-0.13											
F ⁻	0.01	0.05	0.00	0.02											
Cl ⁻	0.11	1.15	0.02	0.22	0.06	0.07	0.03	-0.07	0.12	0.57	-0.02	0.03	-0.02	0.03	
NO ₃ ⁻	0.10	0.96	0.19	0.94	0.17	-0.09	0.65	0.17	1.04	0.33	1.79	0.14	0.02	0.06	
SO ₄ ²⁻	0.13	1.12	0.08	1.21	0.06	0.02	1.37	0.33	1.93	-0.38	5.12	1.24	-1.05	0.71	
% Contribution	4.4	45.1	26.2	24.3	6.7	3.0	37.4	18.6	34.3	6.8	33.2	16.6	26.2	17.3	
Source	SMS	Combustion	SD	SA	VE	SMS	SD	BB	SA	CC	SA	VE	BB	SD	

Table S9. Percent contribution of PM₁₀ sources by PMF at different study sites.

	Mohal-Kullu				Nainital					Darjeeling				
	1	2	3	4	1	2	3	4	5	1	2	3	4	5
OC	41	7	38	14	18	23	13	18	28	10	10	56	12	11
EC	41	9	32	18	0	28	26	14	32	11	0	43	27	19
WSOC	38	14	46	2	14	17	23	16	30	12	13	63	0	12
Na ⁺	39	8	41	12	29	37	3	25	6	3	29	36	28	3
NH ₄ ⁺	0	54	30	15	15	60	11	4	11	0	5	0	10	85
K ⁺	8	6	86	0	11	30	21	19	19	15	10	53	10	13
Mg ²⁺	-	-	-	-	-	-	-	-	-	24	30	23	23	0
Ca ²⁺	-	-	-	-	-	-	-	-	-	9	25	34	20	12
F	39	8	53	0	9	3	16	52	19					
Cl ⁻	31	19	44	6	11	9	8	53	18	13	15	51	14	7
NO ₃ ⁻	12	39	35	14	20	51	7	7	15	14	2	16	10	58
SO ₄ ²⁻	6	48	32	14	21	52	3	8	15	55	7	0	6	32
Mg	17	1	29	53	70	0	13	3	14					
Ca	29	5	32	33	76	1	3	6	14					
B	34	8	42	16	15	5	80	0	0	0	11	0	86	3
Al	32	5	29	34	67	1	13	1	17	1	88	0	8	4
P	10	3	27	60	57	3	11	3	27	0	74	5	17	4
S	23	1	15	61	22	17	35	11	15	5	15	23	44	12
Ti	-	-	-	-	63	2	13	6	16	2	38	29	25	6
Cr	32	9	35	24	4	17	16	51	12	0	9	69	22	0
Fe	19	0	30	51	61	5	10	10	15	1	65	16	15	4
Ni	-	-	-	-	11	19	14	44	11	0	21	44	31	3
Cu	-	-	-	-	16	0	0	0	84	2	11	54	29	4
Zn	33	7	34	26	1	51	0	47	0	3	27	47	17	5
Zr	67	0	9	25	-	-	-	-	-	0	5	52	43	1
Mo	-	-	-	-	0	10	11	63	16					
Mn	-	-	-	-	35	2	15	31	18	-	-	-	-	-
Br	-	-	-	-	6	0	16	62	17	-	-	-	-	-
Ba	69	26	0	5	-	-	-	-	-	-	-	-	-	-
% Contribution	29.5	13.2	34.3	23.0	26.1	17.8	15.2	22.1	18.8	8.2	23.2	32.5	22.6	13.5
Source	VE	SA	BB	SD	SD	SA	BB	FFC	VE	CC	SD	BB	VE	SA

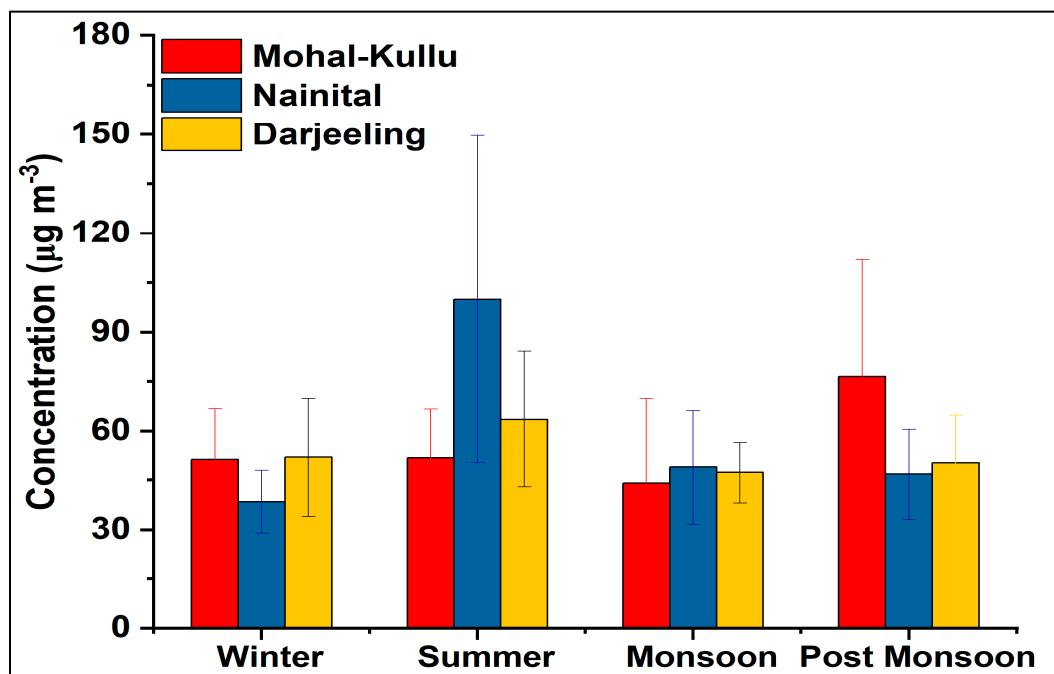


Figure S1. Seasonal variations in the concentration of PM₁₀ at different study sites (in $\mu\text{g m}^{-3}$).

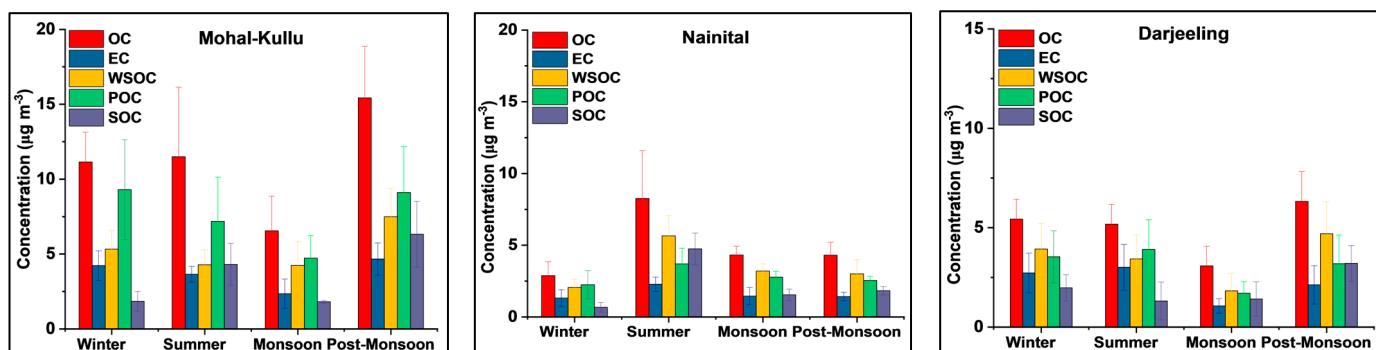


Figure S2. Seasonal variations in the concentration of carbonaceous components (OC, EC, WSOC, POC, SOC) of PM₁₀ at different study sites (in $\mu\text{g m}^{-3}$).

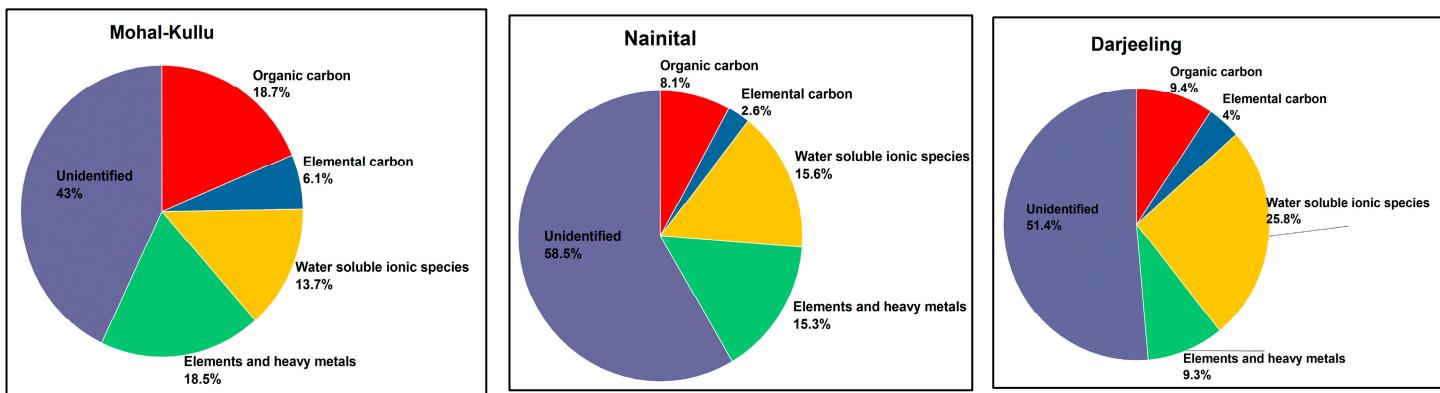


Figure S3. Pie chart showing percentage contribution of analyzed species to PM₁₀ at study sites in IHR.

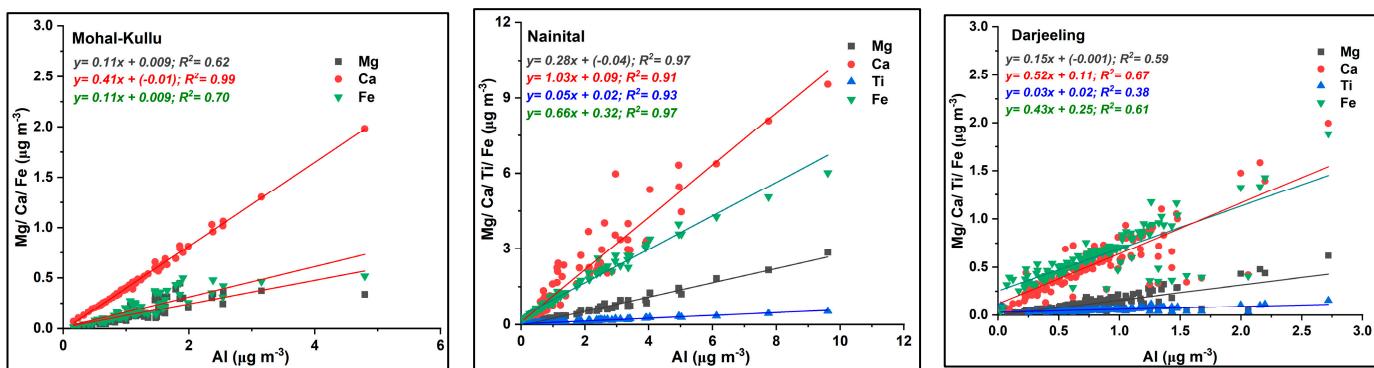


Figure S4. Scatter plots between crustal elements Al with Mg/Ca/Fe, and Ti of PM₁₀ at different study sites.

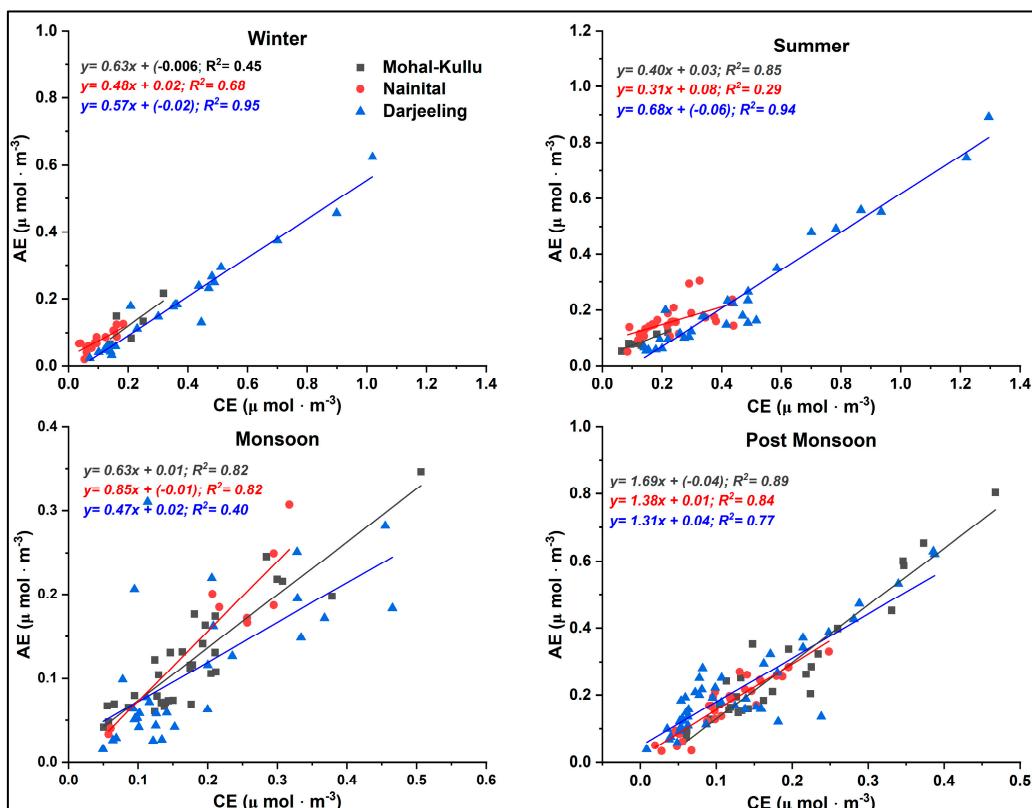


Figure S5. Seasonal CE/AE ratio for aerosol acidity at study sites.

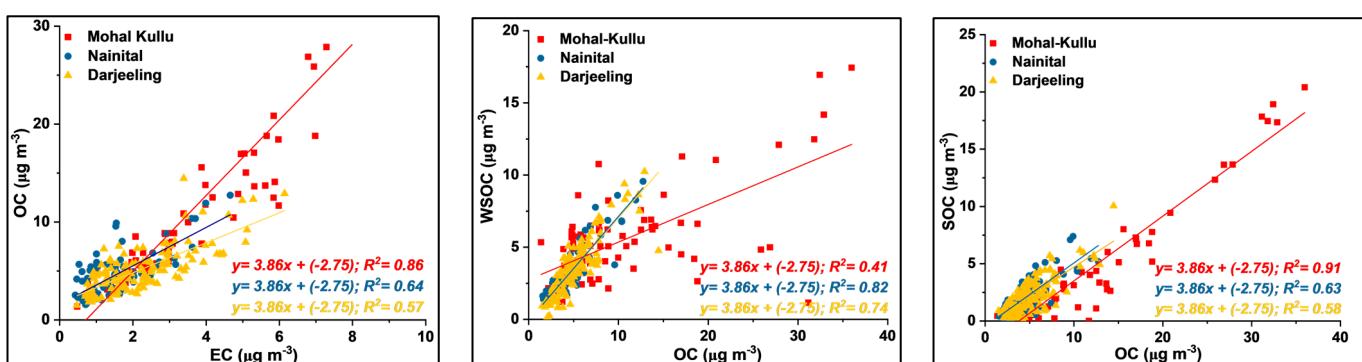


Figure S6. Scatter plots between OC/EC, WSOC/OC, SOC/OC of PM₁₀ at different study sites.