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Rapid transition in winter aerosol composition in Beijing from 2014 to 2017: response to clean air actions

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Table S1. Monthly descriptive statistic of the comparison between the observational $PM_{2.5}$ concentrations and CMAQ-simulated $PM_{2.5}$ concentrations in Beijing during the winters of 2014 and 2017. Concentration values are all reported in μg m^{-3}.

Year	Month	Mean_obs	Mean_sim	R	MB	ME	RMSE	NMB(%)	NME(%)
2014	12	58.75	77.87	0.91	19.12	24.53	39.57	13.73	31.24
2015	1	97.76	104.05	0.75	6.29	32.27	34.84	15.14	37.29
	2	93.36	92.66	0.86	-0.70	21.26	31.47	-1.29	29.34
2017	12	43.35	47.80	0.90	4.45	19.86	25.17	12.01	28.06
2018	1	33.87	55.76	0.79	21.89	35.97	36.41	29.45	44.45
	2	50.14	54.28	0.83	4.14	34.55	32.83	14.08	27.39

Table S2 (a). Correlations between simulated $PM_{2.5}$ compositions and observed PM_1 compositions in this study.

Period	OM	SO_4^{2-}	NO_3^-	NH_4^+	BC
2014 Winter	0.83	0.76	0.82	0.84	0.76
2017 Winter	0.72	0.81	0.89	0.90	0.73

Table S2 (b). Comparison of the simulation and observations of $PM_{2.5}$ compositions. The observation data was collected from the SPARTAN, Beijing Site (40.01°N, 116.207°E) (https://www.spartan-network.org/beijing-china); while the simulated data was extracted from the same grid where the Beijing Site located in the third domain. Concentration values are all reported in μg m^{-3}.

year	Date_range (dd/mm)	$PM_{2.5}$			SIA			SO_4^{2-}			NO_3^-		
		Obs	Sim	MB	Obs	Sim	MB	Obs	Sim	MB	Obs	Sim	MB
2014	01/12-28/12	64.12	70.31	6.19	15.14	18.90	3.75	5.56	6.38	0.82	6.41	8.05	1.64
2015	22/01-27/02	95.38	97.64	2.26	18.55	21.39	2.84	9.81	10.02	0.21	4.13	6.14	2.01
2017	27/11-12/12	39.14	42.70	3.56	14.21	15.56	1.35	2.29	3.49	1.20	8.72	9.72	1.00

year	Date_range (dd/mm)	NH_4^+			RM/OM			BC			OTHER		
		Obs	Sim	MB	Obs	Sim	MB	Obs	Sim	MB	Obs	Sim	MB
2014	01/12-28/12	3.17	4.46	1.29	30.64	24.30	-6.34	7.92	9.20	1.27	20.38	17.92	-2.46
2015	22/01-27/02	4.61	5.23	0.62	30.00	28.90	-1.10	8.82	12.57	3.74	42.59	34.79	-7.80
2017	27/11-12/12	3.20	2.35	-0.85	14.25	13.72	-0.53	3.31	3.37	0.06	6.56	10.04	3.48

[a] SIA represents the total amount of secondary inorganic aerosol, that the sum of SO_4^{2-}, NO_3^- and NH_4^+.

[b] RM represents the residual matter in SPARTAN observations. There is no direct organic composition in SPARTAN, however RM in SPARTAN measurements is dominated by organics, and has been used to evaluate the organic mass simulations (Weagle et al., 2018). OM represents the organic mass in CMAQ simulations.

[c] BC represents the black carbon.

[d] Other represents the rest of the $PM_{2.5}$ compositions, such as crustal matter, sea salt.

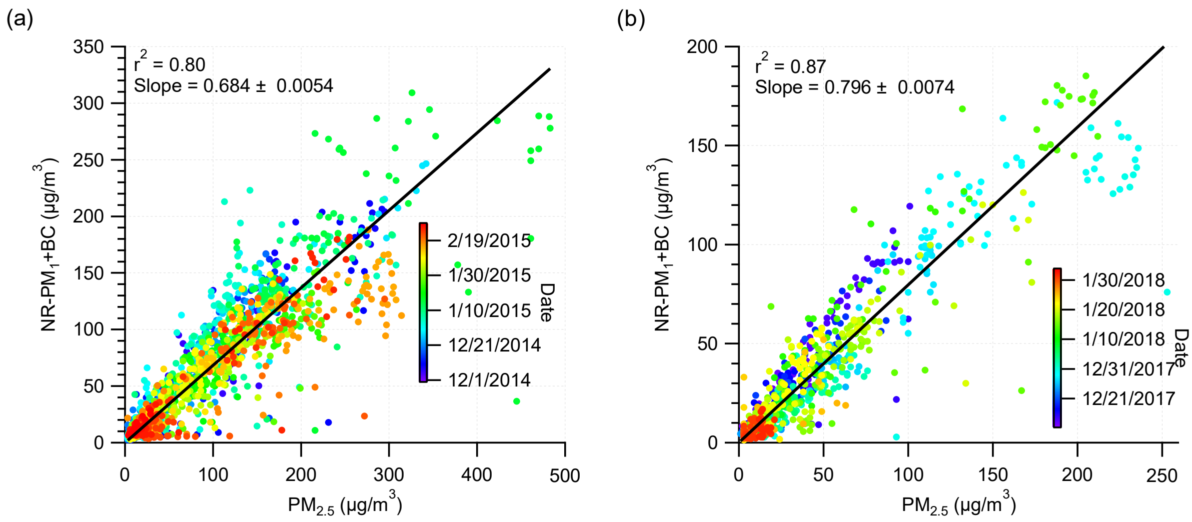

Figure S1. Correlation of total PM_1 concentration ($NR\text{-}PM_1$ plus BC) with $PM_{2.5}$ concentration during the winter of (a) 2014 and (b) 2017.

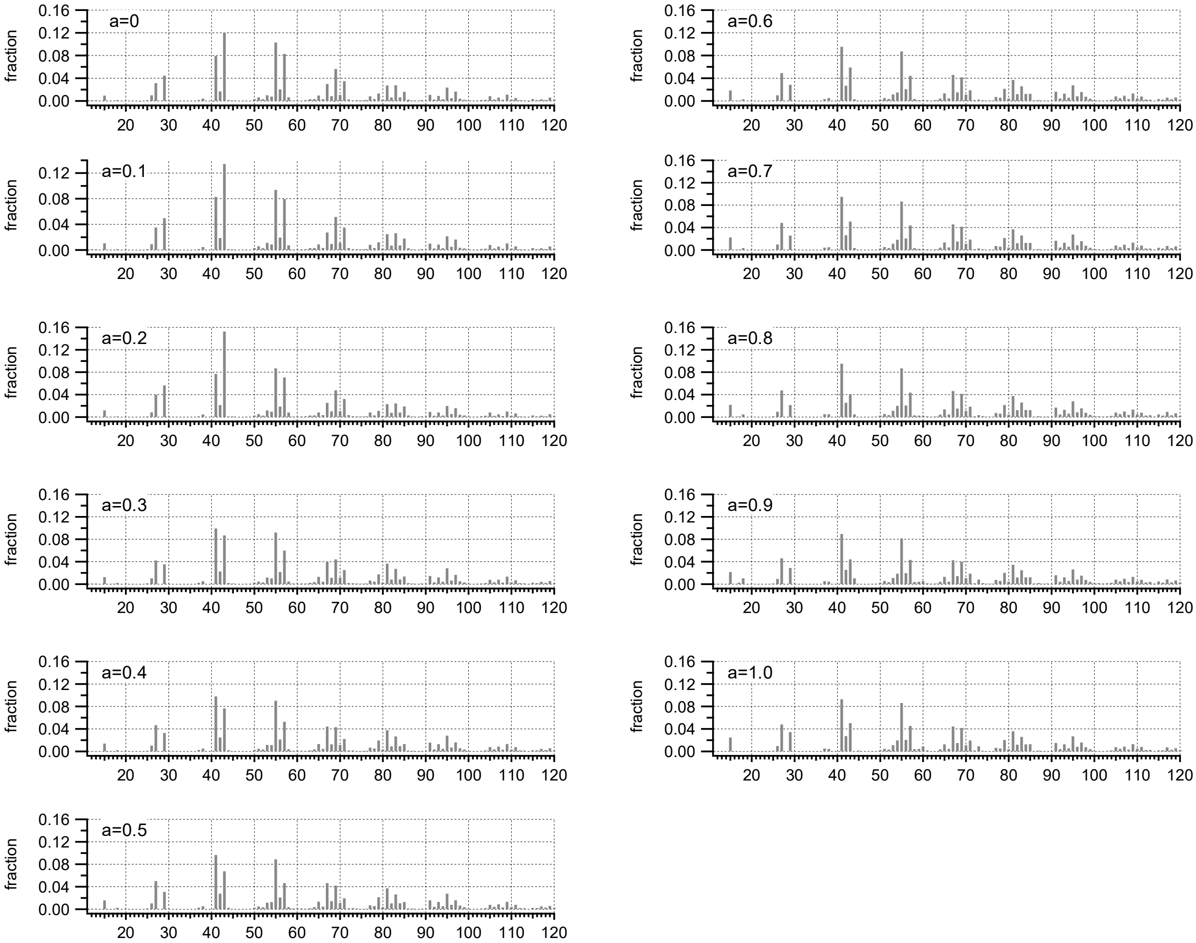

Figure S2. Factor profiles of HOA with different *a* values of ME-2 analysis.

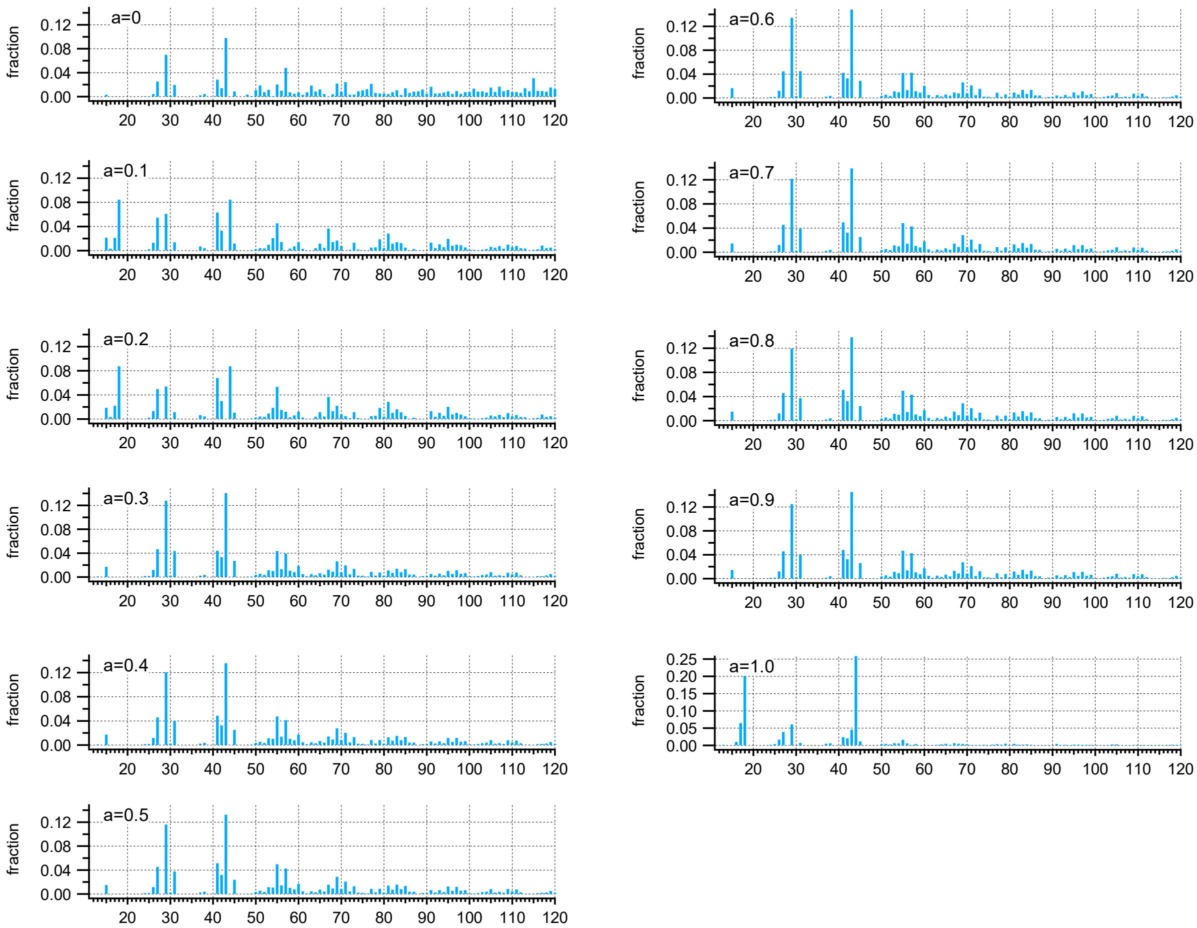

Figure S3. Factor profiles of CCOA with different *a* values of ME-2 analysis.

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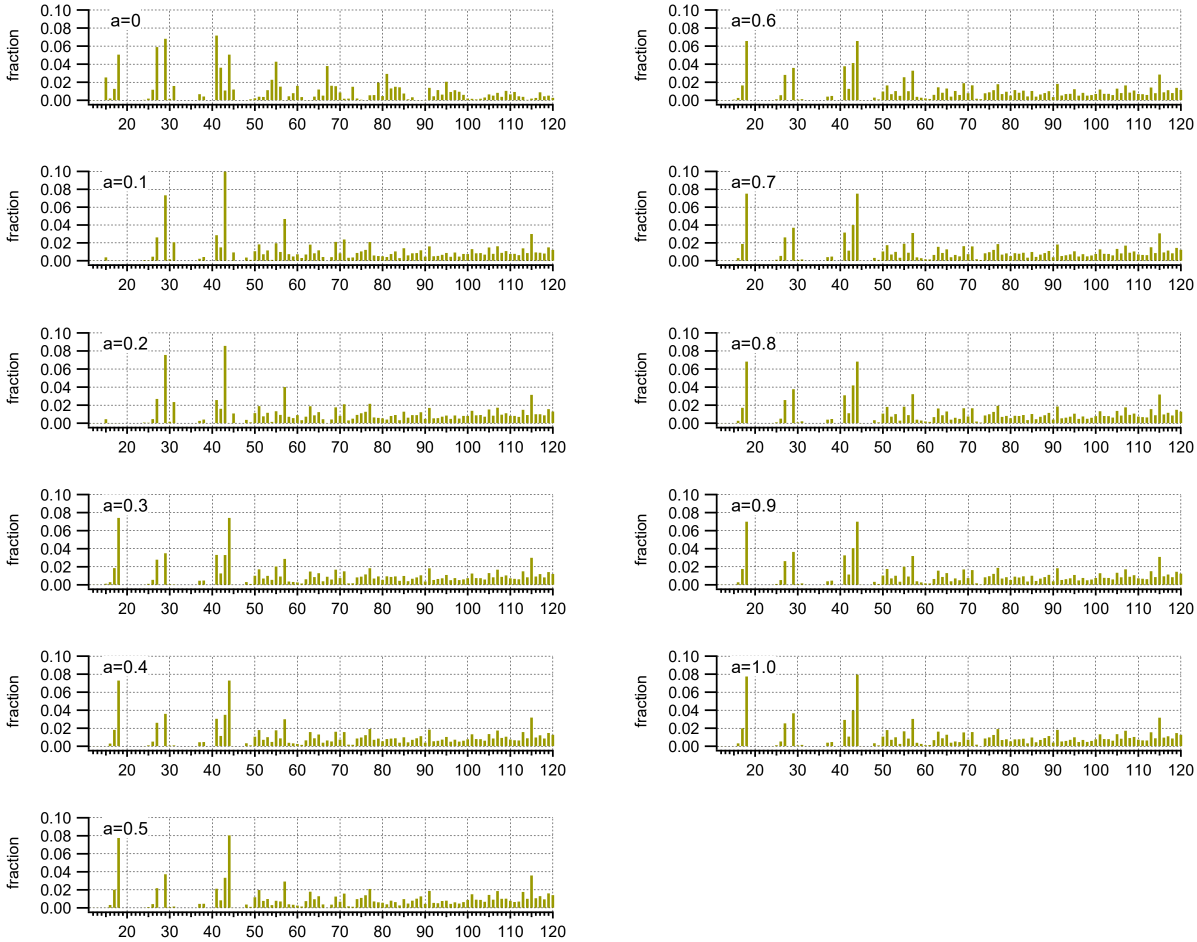


45 **Figure S4.** Factor profiles of BBOA with different *a* values of ME-2 analysis.

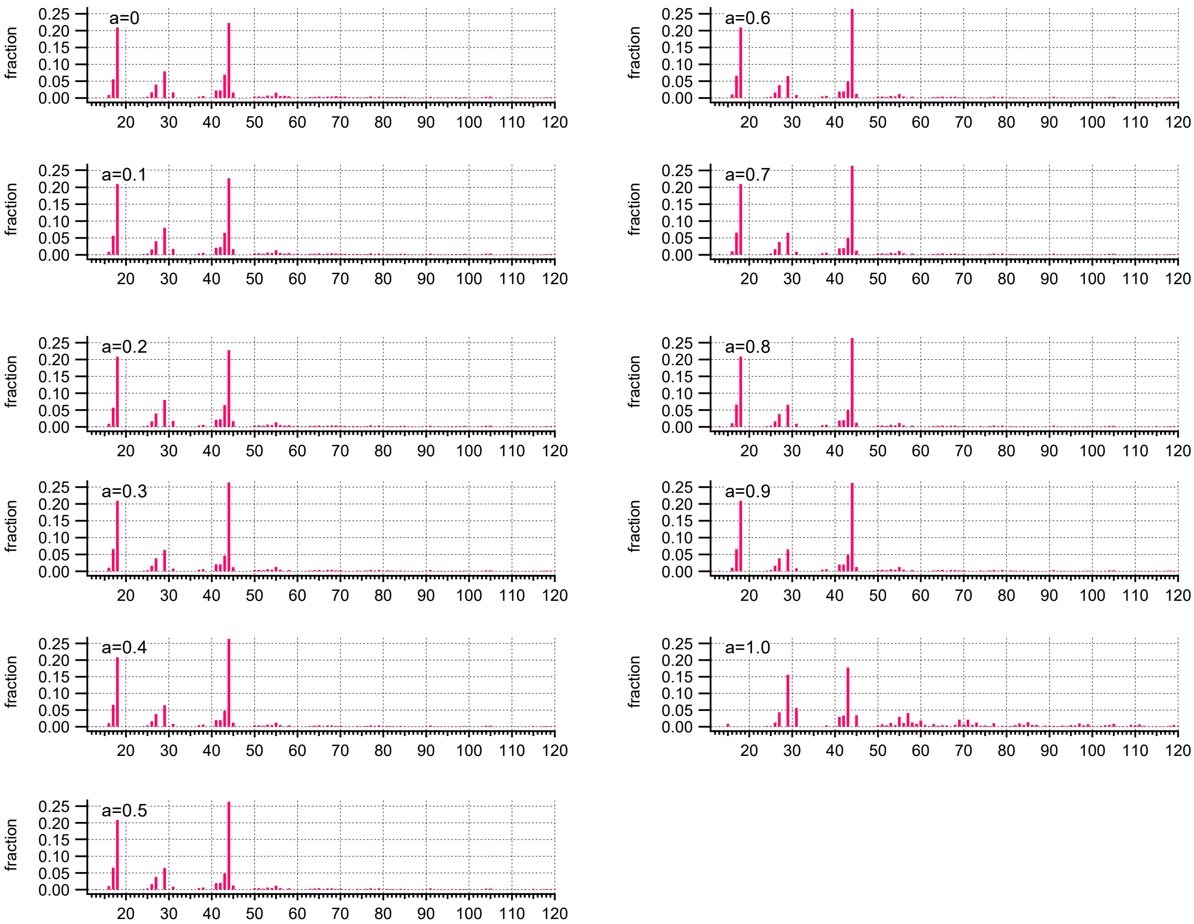


Figure S5. Factor profiles of OOA with different *a* values of ME-2 analysis.

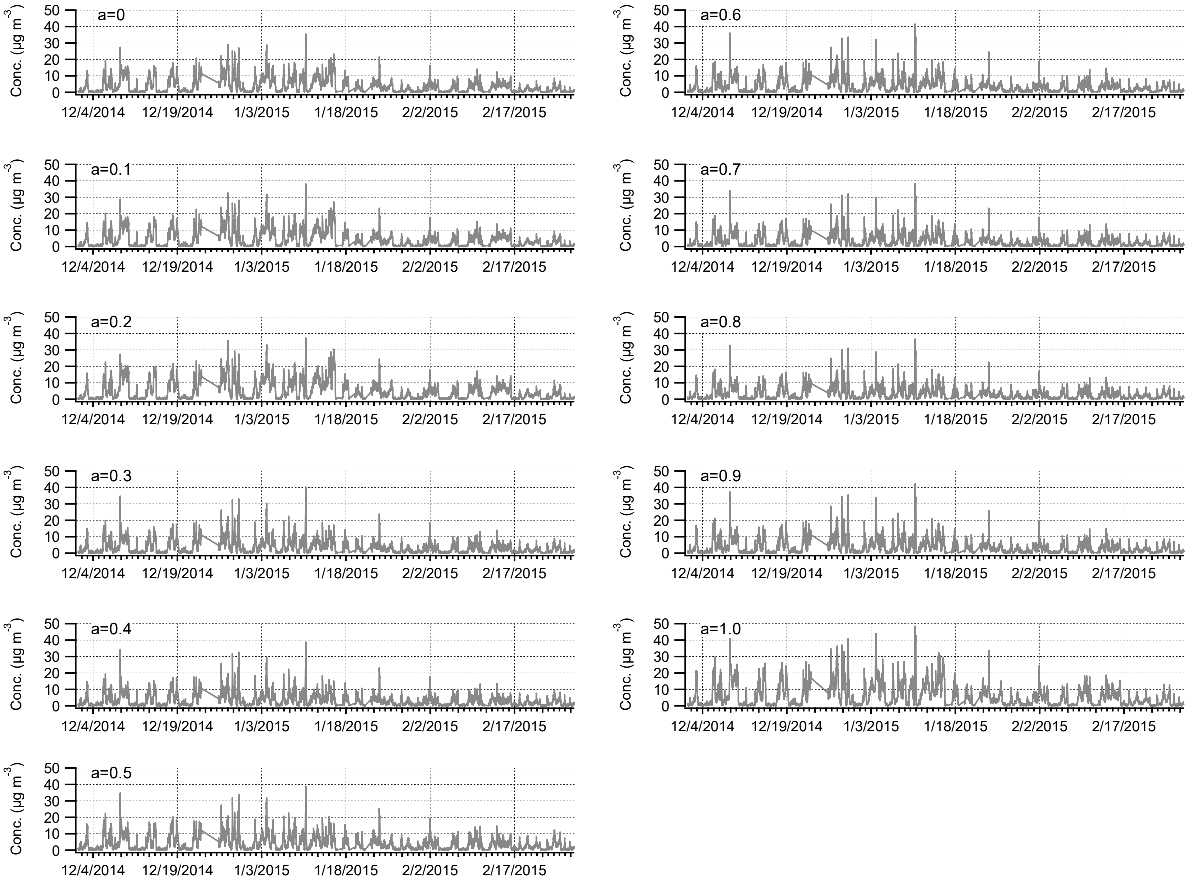


Figure S6. Time series of HOA with different *a* values of ME-2 analysis.

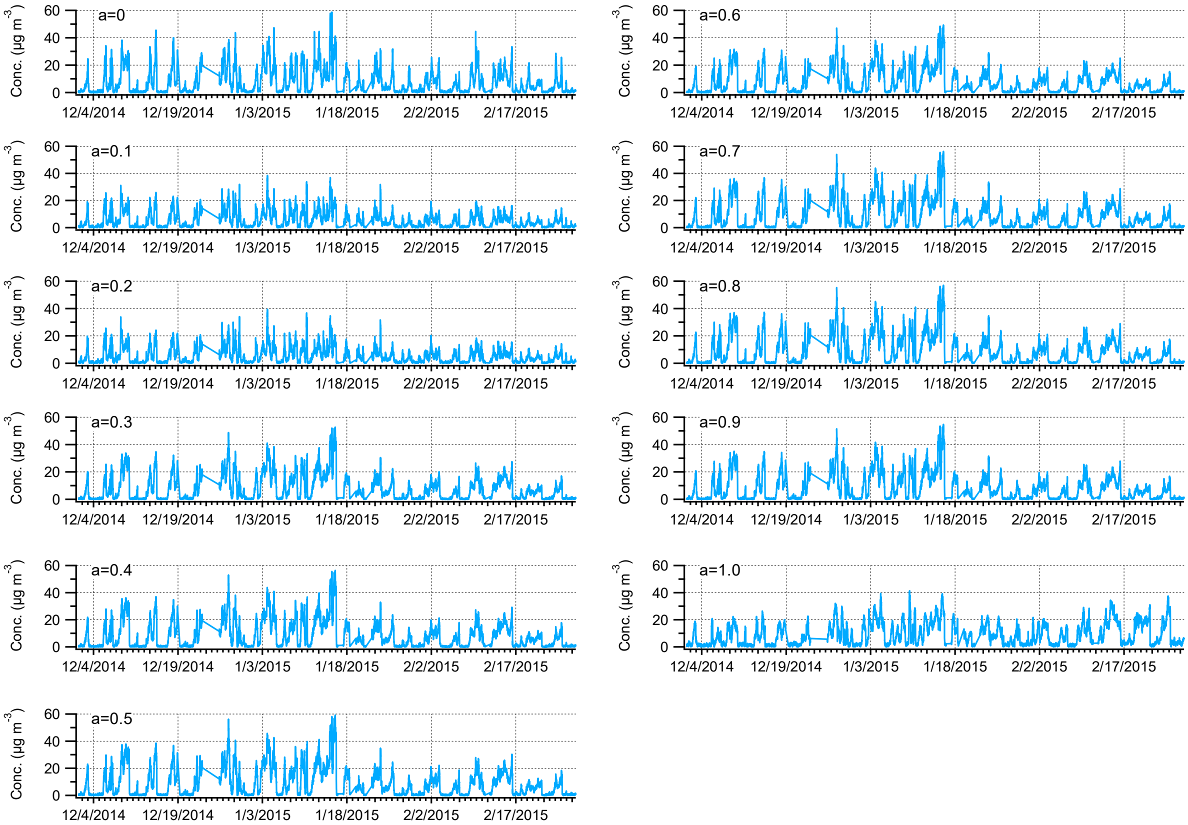


Figure S7. Time series of CCOA with different *a* values of ME-2 analysis.

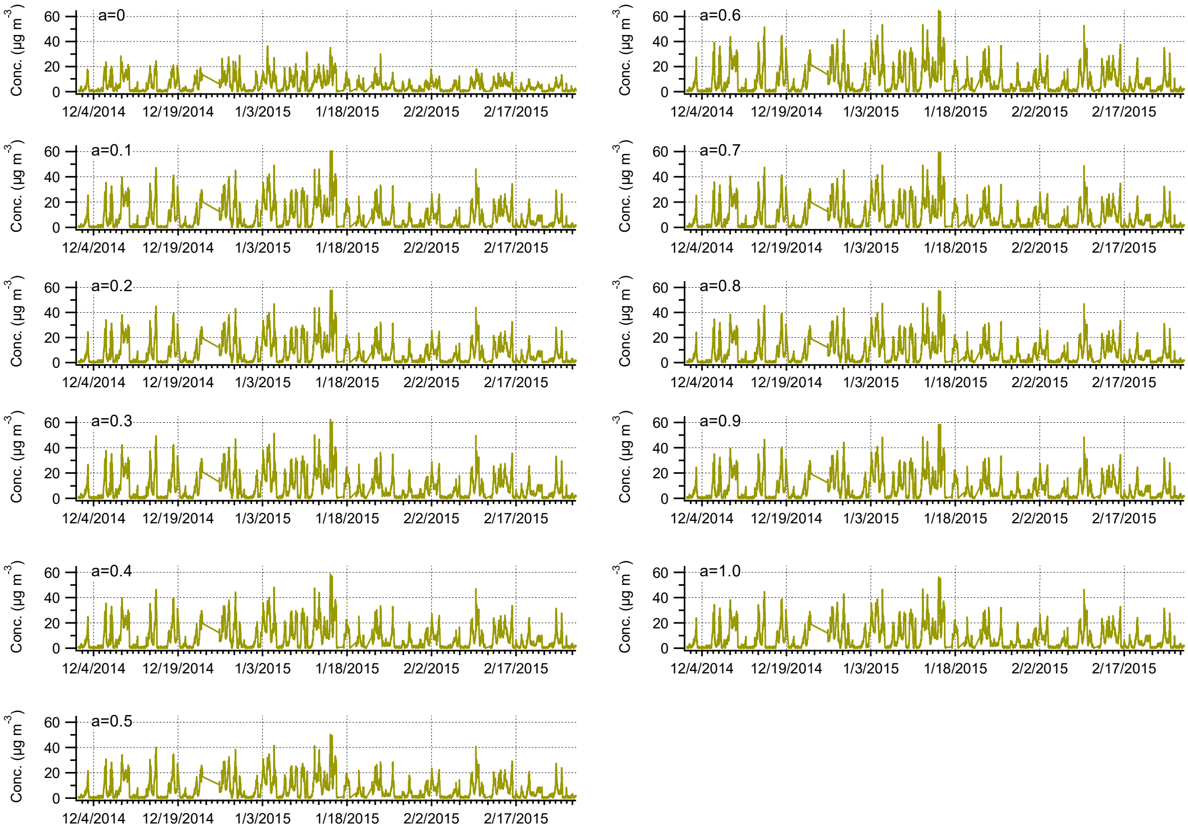


Figure S8. Time series of BBOA with different *a* values of ME-2 analysis.

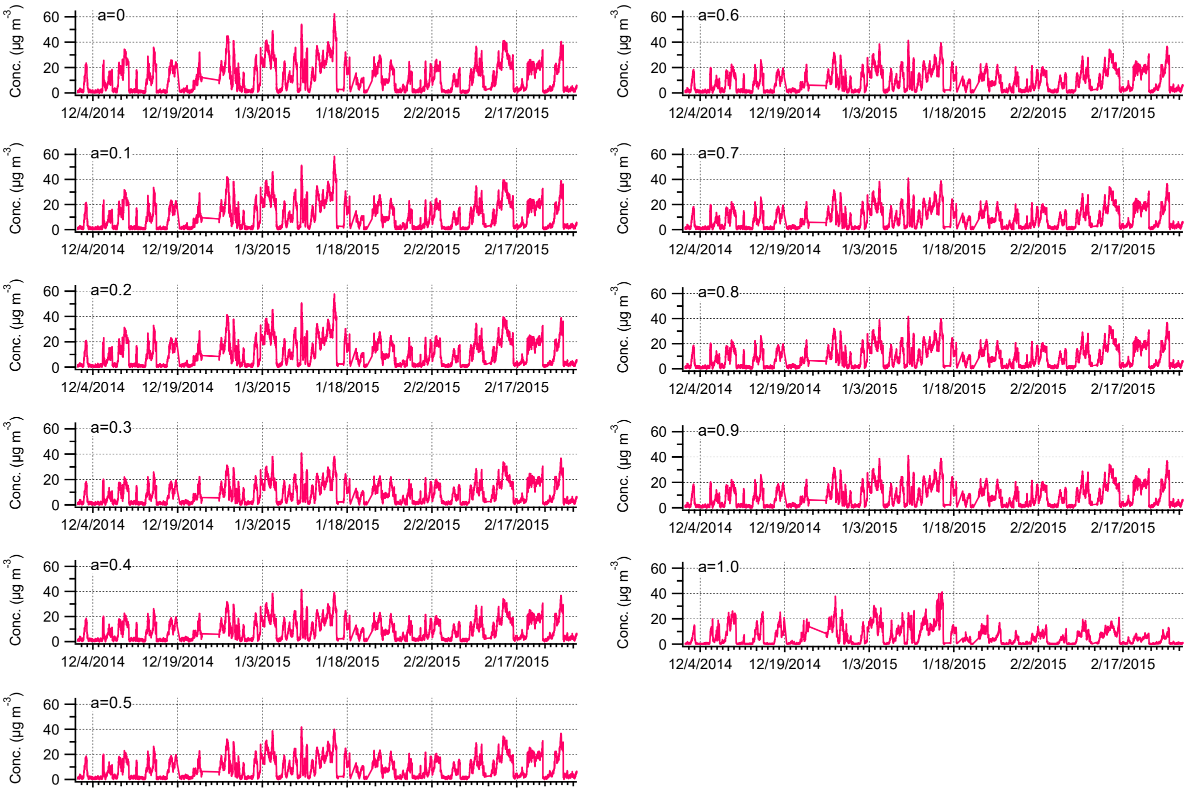
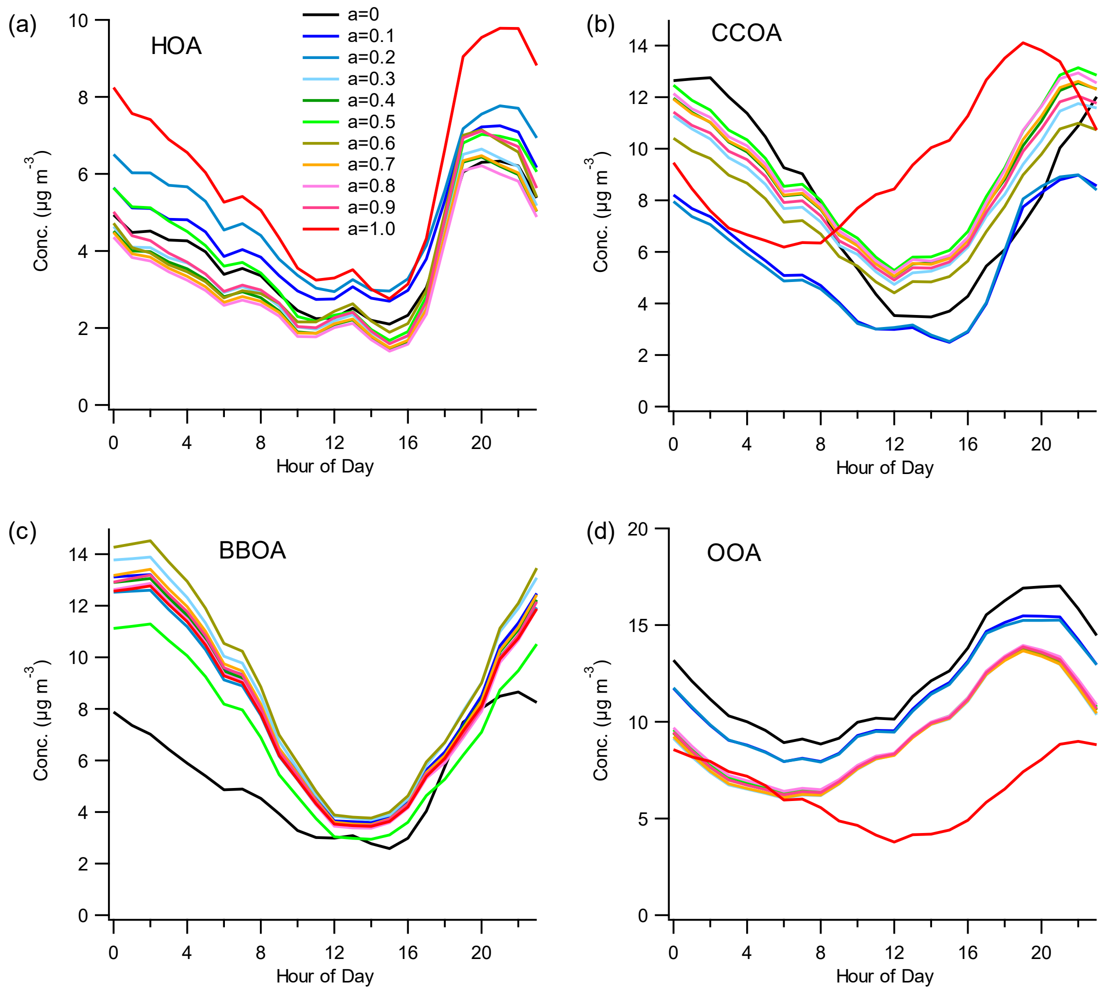


Figure S9. Time series of OOA with different *a* values of ME-2 analysis.



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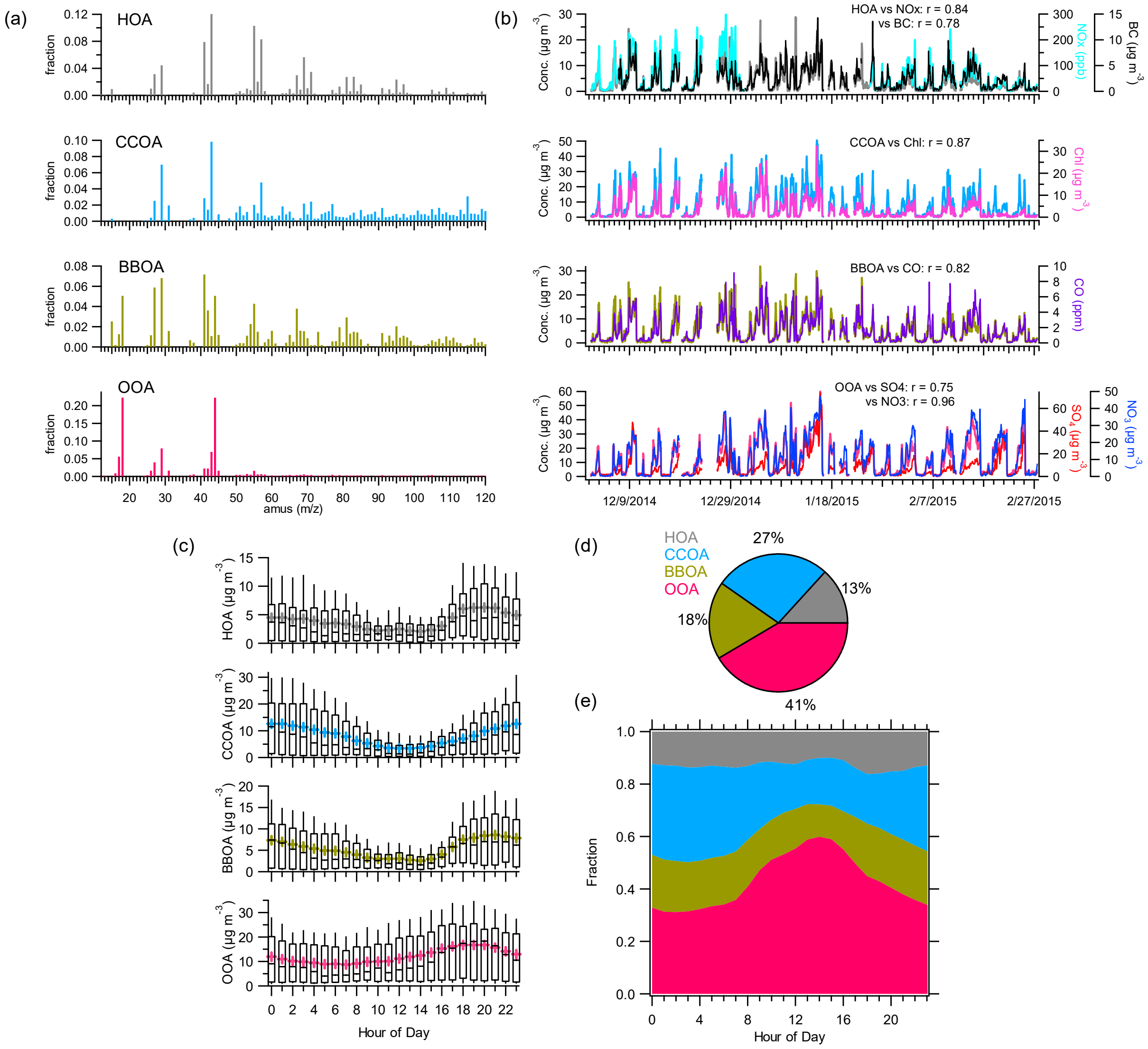
57 **Figure S10.** Diurnal cycles of (a) HOA, (b) CCOA, (d) BBOA, and (d) OOA with different *a*
58 values of ME-2 analysis.



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60 **Figure S11.** Source apportionment results of OA during the winter of 2014. (a) Mass spectra of
61 HOA, CCOA, BBOA, and OOA. (b) Time series of different OA factors and their corresponding
62 tracers. The correlation coefficients of OA factors with the tracers are also shown. (c) Diurnal
63 cycles of OA factors. (d) The average fractional pie chart of OA factors to total OA. (e) The
64 average diurnal mass contributions of OA factors to total OA.

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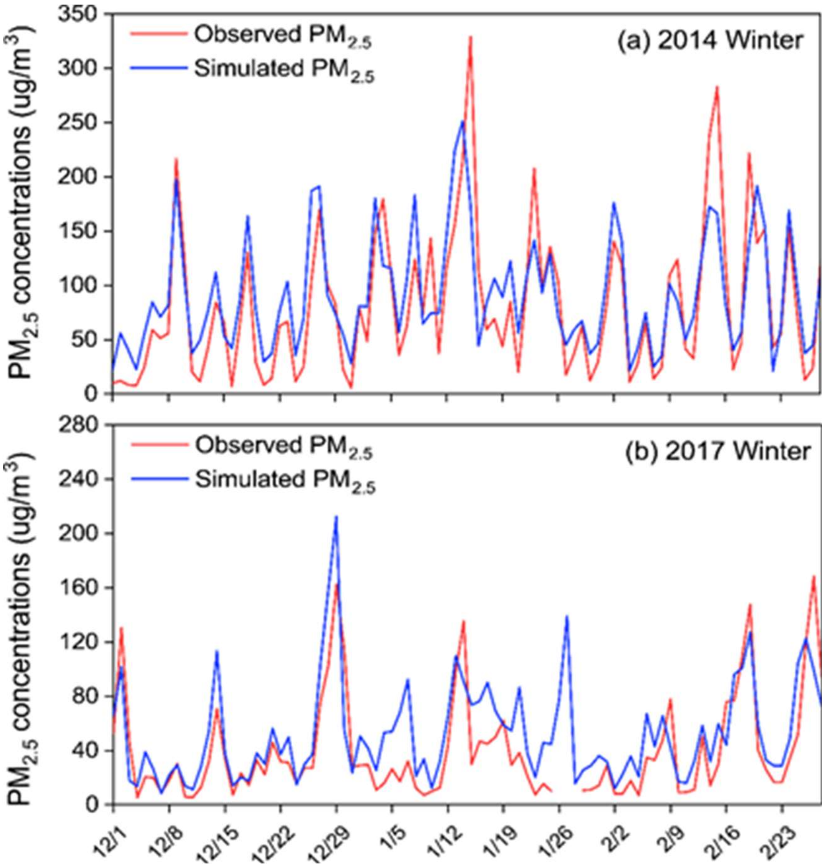
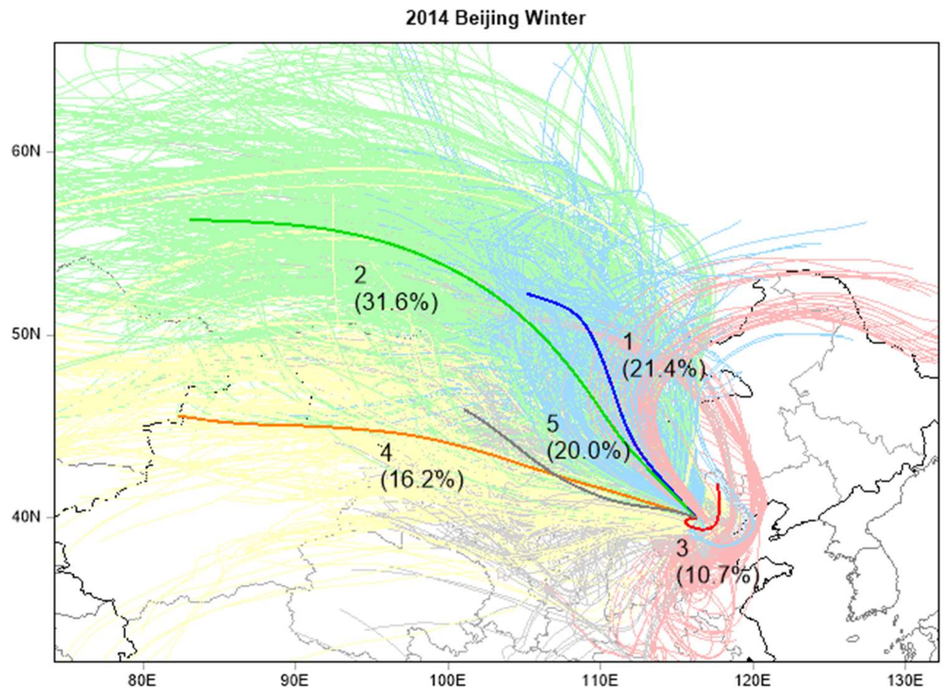


Figure S12. Comparison of observed (red) and CMAQ-simulated (blue) daily mean $PM_{2.5}$ concentrations over Beijing during the winters of 2014 (a) and 2017 (b). Observation data was obtained and averaged from 12 national observation stations in Beijing. Simulated concentrations were extracted from the grids corresponding to the station locations.



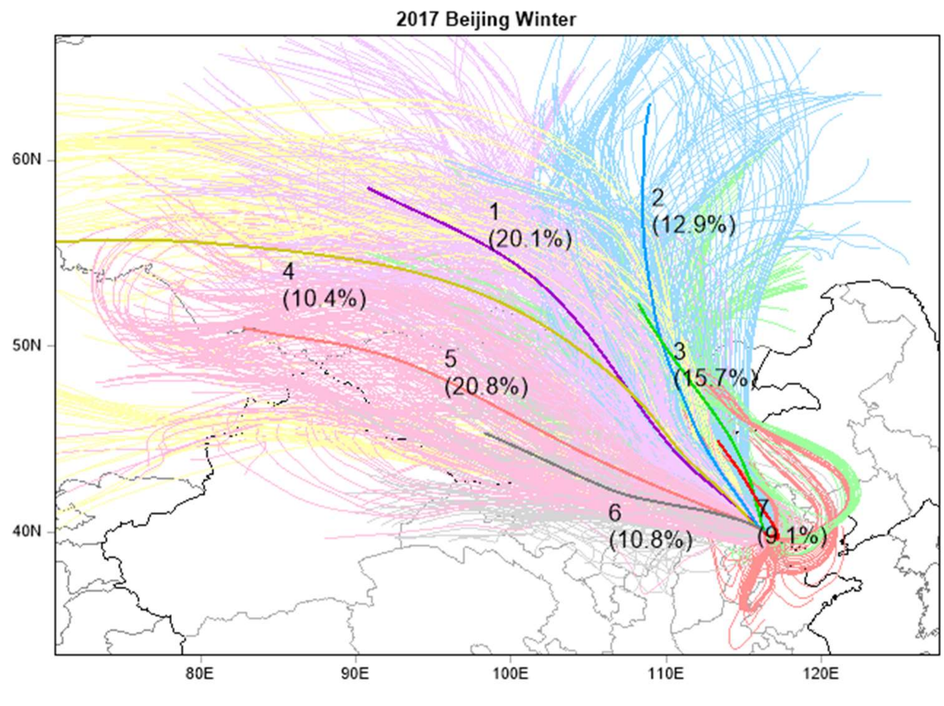


Figure S13. Back trajectories of air masses arriving in Beijing in winter 2014 and winter 2017 with unique colors for different clusters. The average percentage of each cluster is shown.

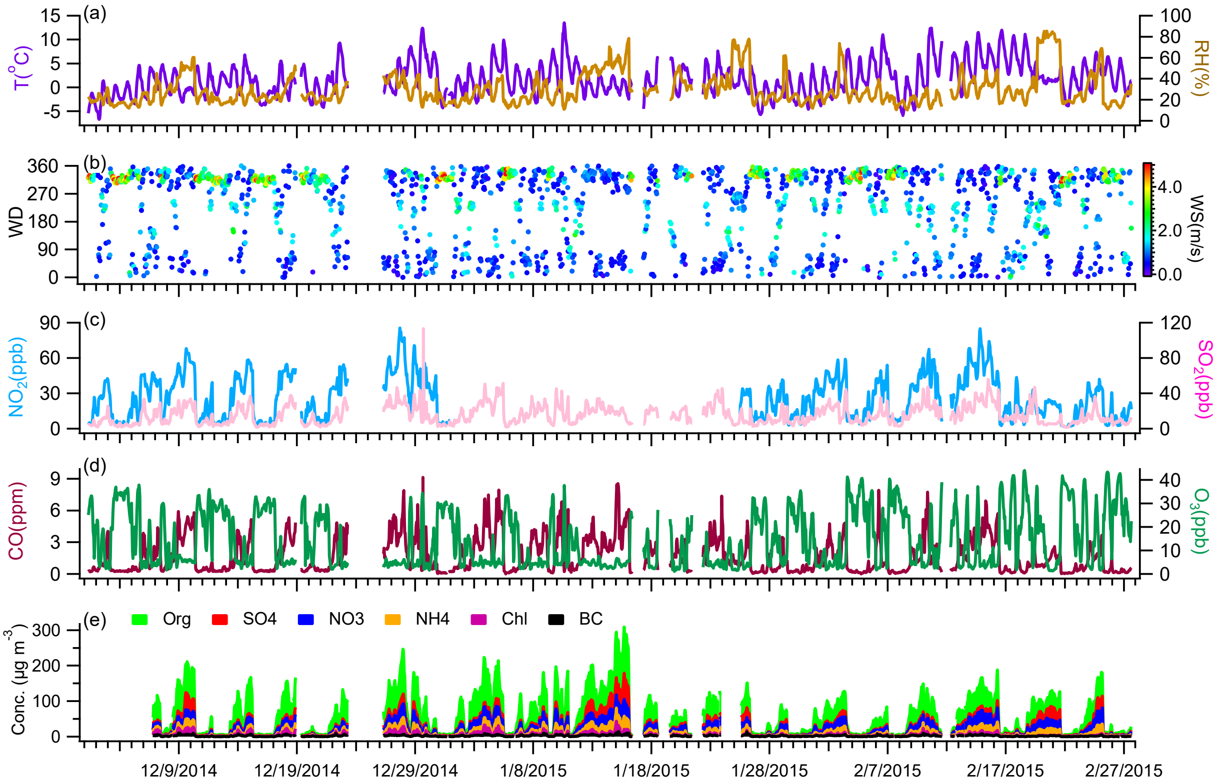


Figure S14. Temporal variations of (a, b) meteorological parameters, (c, d) gaseous species, and (e) aerosol species during the winter of 2014.

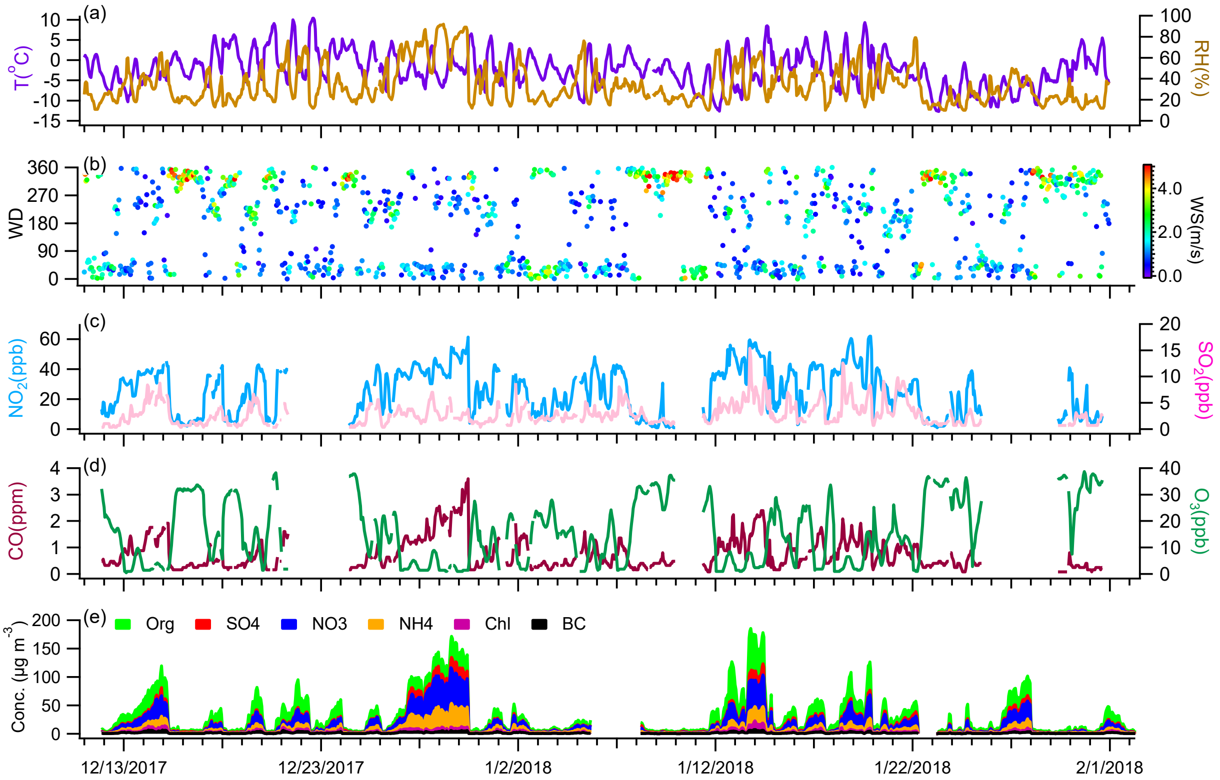


Figure S15. Temporal variations of (a, b) meteorological parameters, (c, d) gaseous species, and (e) aerosol species during the winter of 2017.

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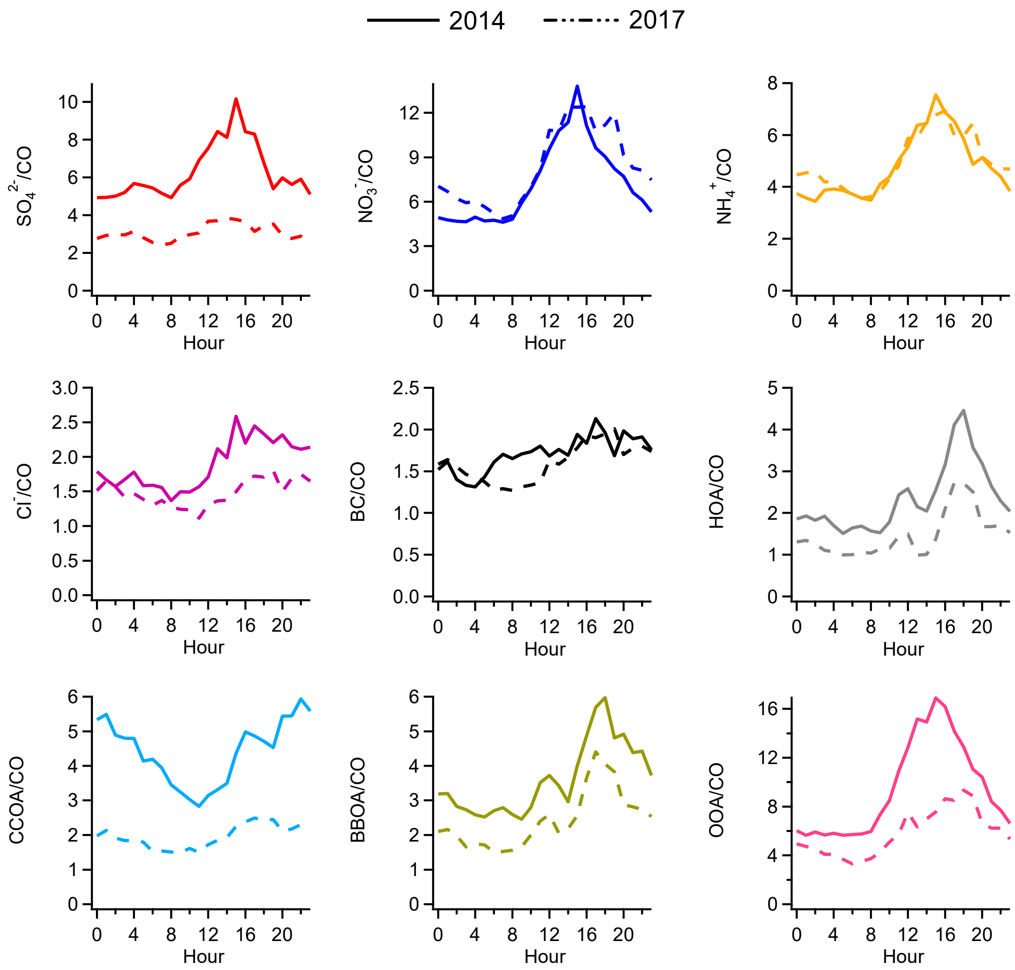

82 **Figure S16.** Average diurnal cycles of CO-scaled aerosol species in the winter of 2014 (solid line)
83 and winter of 2017 (dashed line).

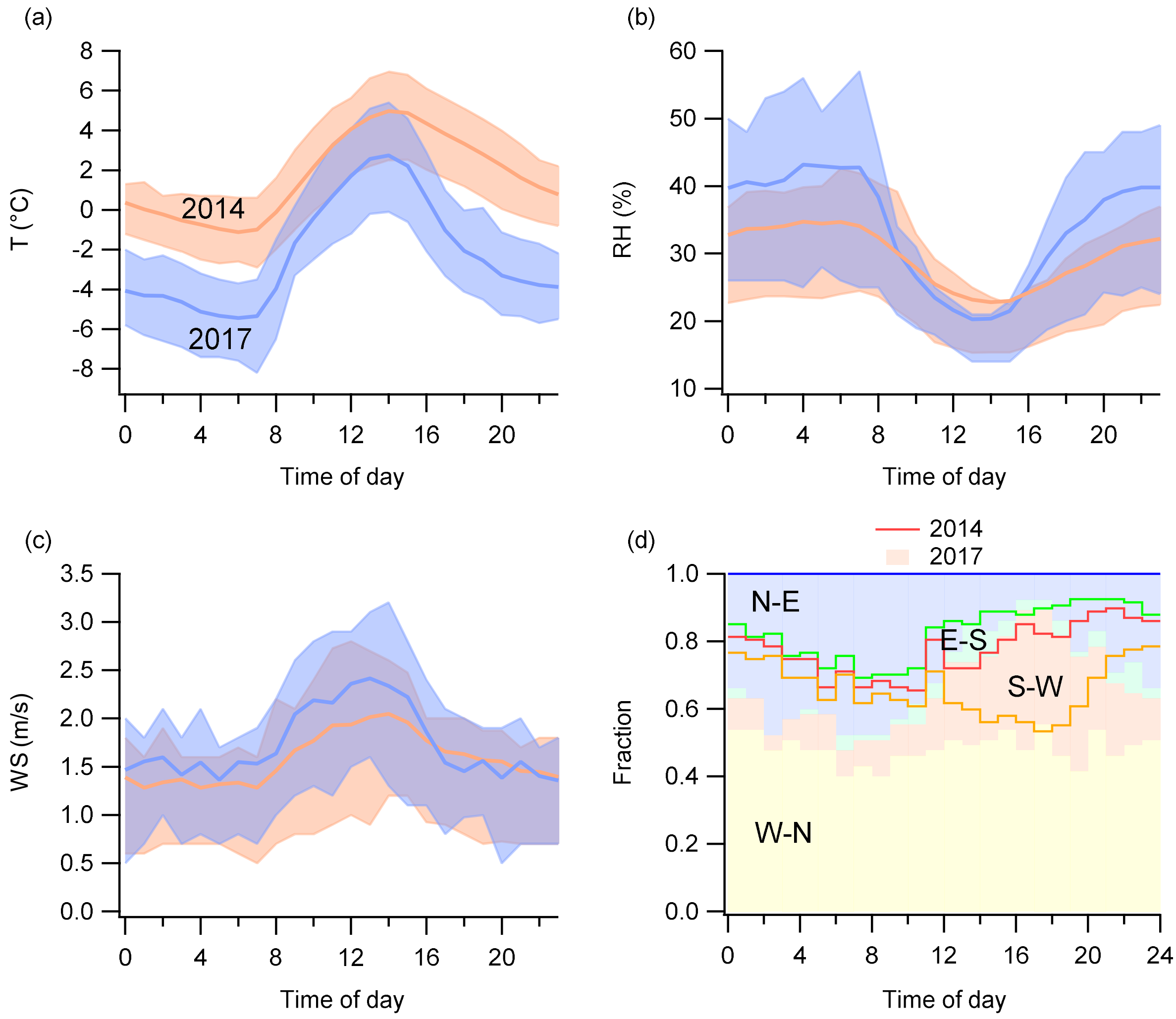

Figure S17. Diurnal cycles of meteorological parameters in 2014 and 2017. (a-c) The average variations of temperature, relative humidity (RH), and wind speed (WS). The shaded areas indicate 25th and 75th percentiles. (d) The distribution of different wind directions through the day. The lines and the stacked areas represent the year of 2014 and 2017, respectively.

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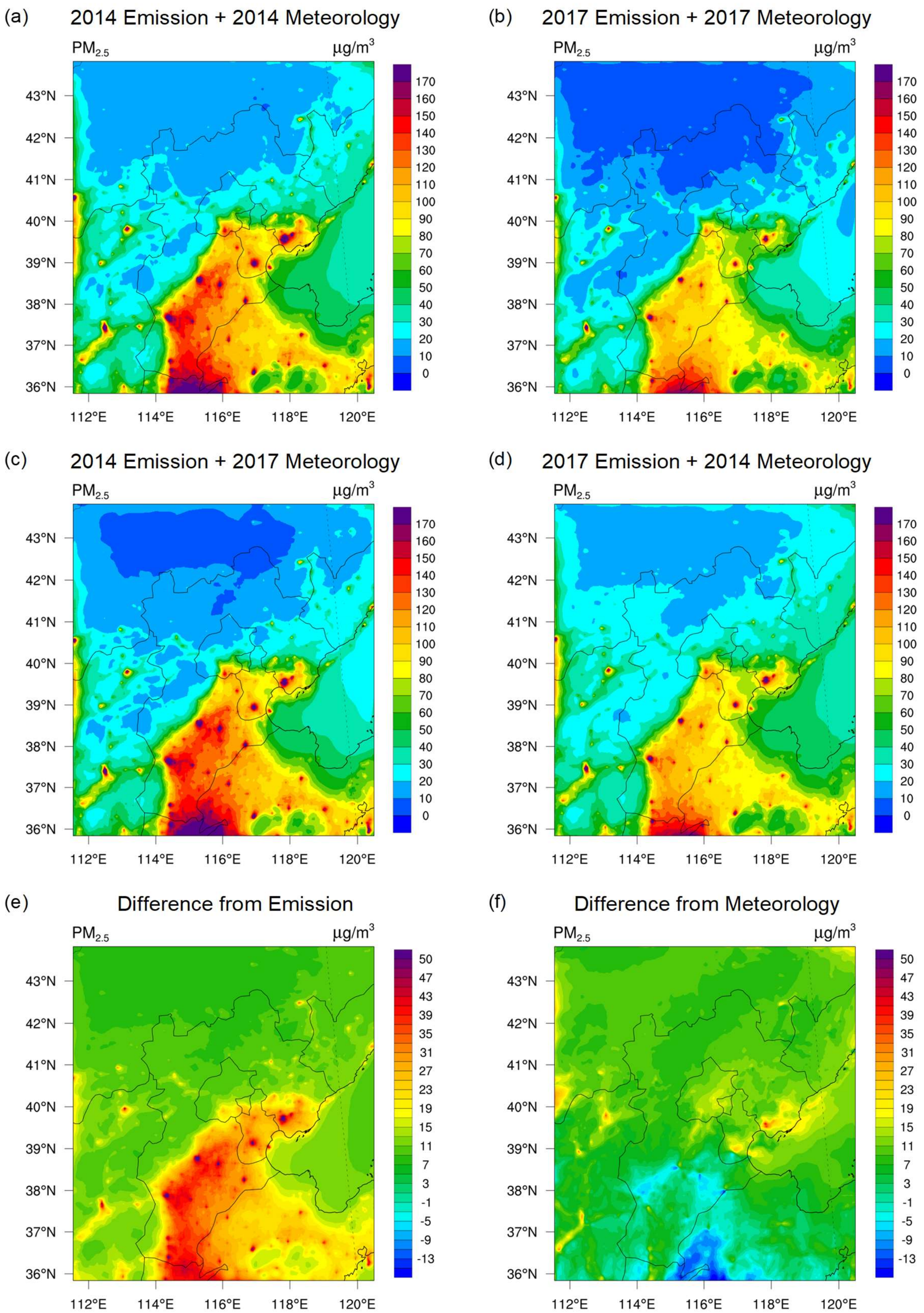
(a)
2014 Emission + 2014 Meteorology
(b)
2017 Emission + 2017 Meteorology
(c)
2014 Emission + 2017 Meteorology
(d)
2017 Emission + 2014 Meteorology
(e)
Difference from Emission
(f)
Difference from Meteorology
PM2.5
μg/m3
43°N
42°N
41°N
40°N
39°N
38°N
37°N
36°N
112°E
114°E
116°E
118°E
120°E
170
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Figure S18. WRF-CMAQ simulated $PM_{2.5}$ concentration under different scenarios for the observation periods of 2014 and 2017.

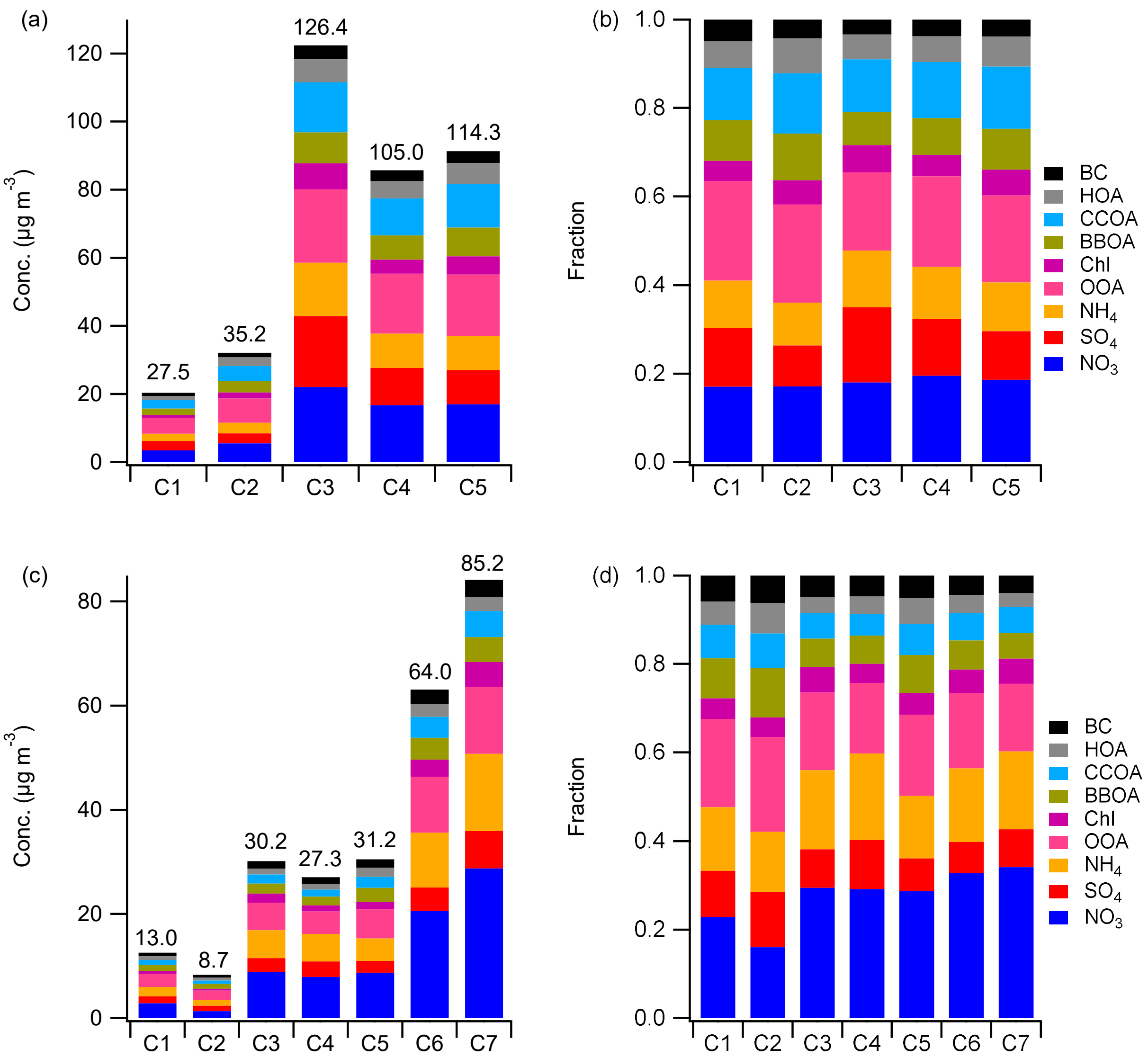

Figure S19. The mass concentrations of aerosol species and their fractional contributions in total PM_1 for different clusters in (a, b) winter 2014 and (c, d) winter 2017. The average PM_1 concentration of each cluster is shown on the top of the bar.

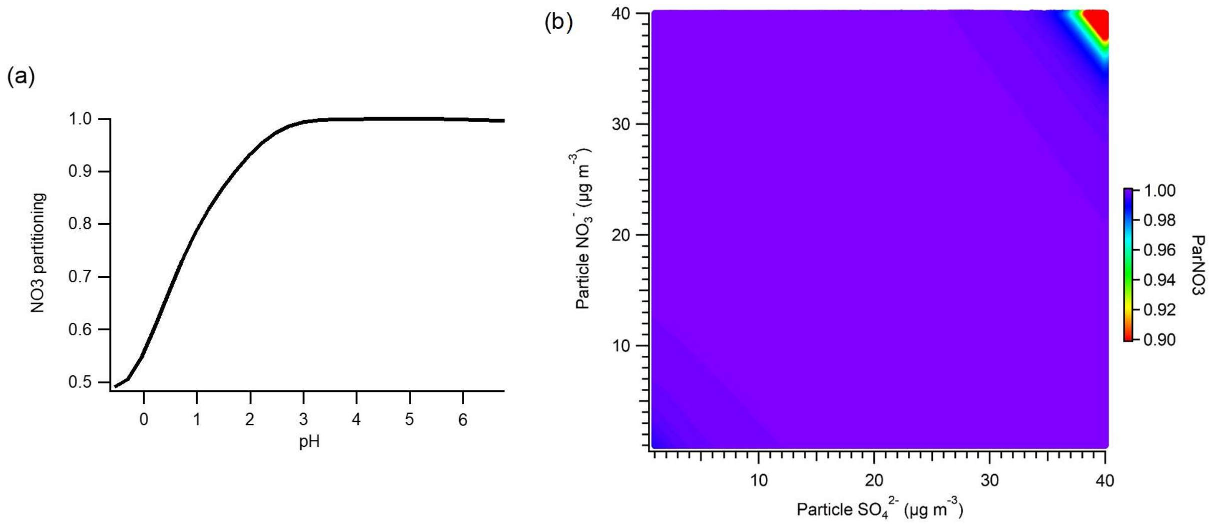

Figure S20. (a) Variation of $\in (NO_3{}^-)$ as a function of particle pH. (b) Sensitivity of the fraction of particulate nitrate to the mass concentrations of particulate sulfate.