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Measurement report: Formation of tropospheric brown carbon in a lifting air mass

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S1. UV−vis light absorption measurement

About 8 punches (12 cm^3) form each filter sample was extracted three time under sonication with 15 ml Milli-Q pure water (18.2 MΩ), and the extract was subsequently filtered through 0.45 μm PTFE pore syringe filter to remove the insoluble component in the suspension. A liquid waveguide capillary UV−vis spectrometer equipped with a 1 m long-effective path detection cell was applied to record the light absorption spectra of all extracts. The light absorption spectrum was finally converted into absorption coefficient (abs_λ, M/m) at a particular wavelength (λ) using the following equation.

$$abs_\lambda = (A_\lambda - A_{700})\frac{v_1}{v_a \times l} \times \ln(10) \quad \text{Eq.S1}$$

Where A_λ and A_{700} represent the light absorption of the extracts at wavelength λ and 700 nm, respectively. V_1 corresponds to the volume of the extract, e.g., 15 ml; V_a refers to the volume of the air through corresponding to filter punches; l (m) is the absorbing path length. While, ln(10) is used for converting common logarithm that provided by the spectrophotometer to natural logarithm.

Table S1 The result of bootstrap analysis.

Sources	Secondary formation	Fossil fuel combustion	Biomass burning	Fugitive dust	Unmapped
Secondary formation	50	0	0	0	0
Fossil fuel combustion	1	49	0	0	0
Biomass burning	0	0	50	0	0
Fugitive dust	0	5	0	45	0

Table S2 RF model performance for testing dataset.

	MF site	MS site
R^2	0.92	0.86
MSE	0.003	0.008
RMSE	0.054	0.091
MAE	0.04	0.07

Note: MSE: mean square error; RMSE: root-mean-square error; MAE: mean absolute error.

Table S3. Information and mass concentration (μg m^{-3}) of nitroaromatic compounds detected in this study.

Compounds	Molecular	Formula	CAS number	Abbreviation	MF site	MS site
4-Nitrophenol	139.11	$C_6H_5NO_3$	100-02-7	4NP	2.6±2.6	0.40±0.25
3-Methoxy-4-nitrophenol	153.14	$C_7H_7NO_3$	2581-34-2	3M4NP	0.07±0.05	0.01±0.01
4-Nitrocatechol	155.11	$C_6H_5NO_4$	59030-13-6	4NC	1.6±2.5	0.25±0.67
4-Methyl-5-nitrocatechol	169.13	$C_7H_7NO_4$	68906-21-8	4M5NC	11.4±11.4	1.8±1.3
3-Nitrosalicylic acid	183.12	$C_7H_5NO_5$	85038-1	3NSA	0.01±0.01	0.004±0.01
5-Nitrosalicylic acid	183.12	$C_7H_5NO_5$	96-97-9	5NSA	0.13±0.13	0.012±0.015

Table S4 NH_3 concentrations in different regions of China

Region	Location	Period	Ammonia (μg m^{-3})	Reference
NCP	Beijing	Summer of 2009	29.4±11.9	Meng et al. (2017)
	Gucheng	Mar.2016-May.2017	22.2±12.8	Kuang et al. (2020)
		May-Sep 2013	27.5±42.8	Meng et al. (2018)
	Luancheng	Dec.2015-Feb.2016	17.2	Pan et al. (2018)
	Cangzhou	Dec.2015-Feb.2016	22.2	
FWP	Xi'an	Winter of 2016	29±7.3	Wu et al. (2020a)
		Summer of 2016	38±9.4	
		2006-2007	12.9	Cao et al. (2009)
	Weinan	Dec.2015-Feb.2016	12.4	Pan et al. (2018)
	Mt. Hua-MS	Summer of 2020	3.1±1.9	Wu et al. (2022)
	Mt. Hua-MF	Summer of 2020	27.3±51	
YRD	Shanghai	Autumn of 2019	9.5	Wu et al. (2023)
		July-Dec 2013, Mar-June 2014	9.4±6.9	Wang et al. (2015)
		Dec.2019-Jan.2020	9.3±4.0	Lv et al. (2022)
	Nanjing	Dec.2015-Feb.2016	10.8	Pan et al. (2018)
	Taihu	Dec.2015-Feb.2016	6.3	
	Lin'an	Sep 2009-Dec 2010	12.5±8.5	Meng et al. (2014)
PRD	Guangzhou	Dec.2015-Feb.2016	5.8	Pan et al. (2018)
		Oct-Nov 2004	7.3±6.2	Hu et al. (2008)
	Dinghushan	Dec.2015-Feb.2016	2.8	Pan et al. (2018)
	Maoming	Dec.2015-Feb.2016	9.8	
	Hongkong	Autumn 2000	2.3±2.7	Yao et al. (2006)
TP	Lhasa	Dec.2015-Feb.2016	4.8	Pan et al. (2018)
	Ali	Dec.2015-Feb.2016	1.7	

Note: In some cities, the NH_3 unit is ppb, which was converted by the formula of standard atmospheric pressure and normal temperature in this study. The abbreviations of NCP, FWP, YRD, PRD and TP indicate North China Plain, Fen-wei Plain, Yangtze River Delta, Pearl River Delta and Tibet Plateau, respectively.

Table S5 MAE_{365} of water-soluble BrC in $PM_{2.5}$ among different cities in the world.

Region	Location	Year	Season	MAE_{365} (m^2 g^{-1})	Reference
NCP, China	Beijing	2011	Winter	1.2±0.1	Cheng et al. (2016)
		2010-2011	Winter	1.26	Du et al. (2014)
		2010-2011	Summer	0.51	
	Xingtai	2018-2019	Spring	1.4±0.18	Li et al. (2023a)
			Summer	0.95±0.18	
			Autumn	1.5±0.13	
			Winter	1.9±0.16	
	Jinan (TSP)	2016	Spring	1.00±0.23	Wen et al. (2021)
	Zhangbei	2016	Spring	1.32±0.34	
	Tianjin	2016–2017	Winter	1.54±0.33	Deng et al. (2022)
			Summer	0.84±0.22	
FWP, China	Xi'an	2016–2017	Winter	1.2±0.06	Wu et al. (2020b)
			Summer	1.1±0.2	
		2020	Winter	0.78 ± 0.96	Li et al. (2023b)
	Licun	2017	Winter	0.94±0.28	Li et al. (2020)
			Summer	1.01±0.18	
	Mt. Hua-MS	2016	Summer	0.69±0.2	This study
	Mt. Hua-MF			0.67±0.21	
YRD, China	Changzhou	2018	Winter	0.74	Tao et al. (2021)
	Yangzhou	2015-2016	Annual	0.75 ± 0.29	Chen et al. (2020)
	Nanjing	2015-2016	Spring	0.69	Chen et al. (2018)
			Summer	0.51	
			Autumn	0.55	
			Winter	1.04	
	Shanghai	2018-2019	Winter	1.18±0.42	Zhao et al. (2021)
PRD, China	Guangzhou	2012	Winter	0.81	Liu et al. (2018)
		2018	Winter	1.0 ± 0.21	Zou et al. (2023)
		2019	Winter	0.34	Wang et al. (2022)
		2016	Autumn	0.60±0.06	He et al. (2023)
	Taipei	2021	Annual	0.86±0.60	Ting et al. (2022)
TP, China	Lulang	2015-2016	Winter	0.75 ± 0.13	Zhu et al. (2018)
	Lhasa	2013-2014	Annual	0.74	Li et al. (2016)
	Southeast TP	2013-2014	Summer	0.27 ± 0.10	Wu et al. (2020c)
			Winter	0.86 ± 0.17	
	Nam Co	2015	Summer	0.38±0.16	Zhang et al. (2017)
India	Delhi	2016	Spring	2.5	Dasari et al. (2019)
Korea	Seoul	2012-2013	Winter	1.02	Kim et al. (2016)
			Summer	0.28	
USA	Los Angeles	2018-2019	Winter	0.7±0.2	Soleimanian et al. (2020)
			Summer	0.5±0.2	
	Yorkville	2010	Winter	0.41	Hecobian et al. (2010)
	Carolina	2013	Summer	0.29	Xie et al. (2019)
Switzerland		2013	Winter	0.9	Moschos et al. (2018)
			Summer	0.28	
Greece	Ioannina	2019	Summer	0.3 ± 0.1	Paraskevopoulou et al. (2023)

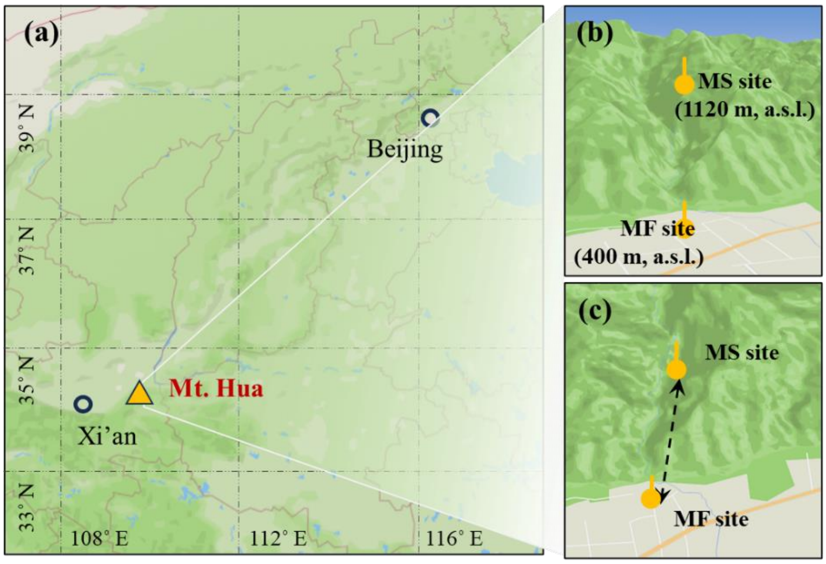

Figure S1. Locations of the sampling sites in China. **(a)** Topographic view, **(b)** vertical view and bird's-eye view **(c)** of Mt. Hua with the sampling sites marked. The maps are the reproductions from ©Mapbox (https://account.mapbox.com/, last access: 16 March 2024)

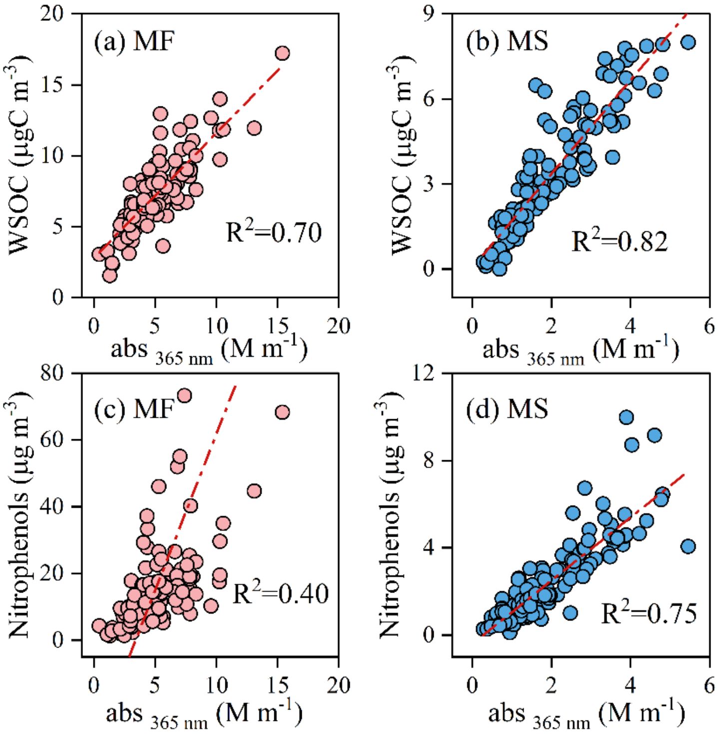

Figure S2. Linear fit regression analysis for Abs $_{\lambda=365\ nm}$ with WSOC and nitrophenols at MF and MS sites

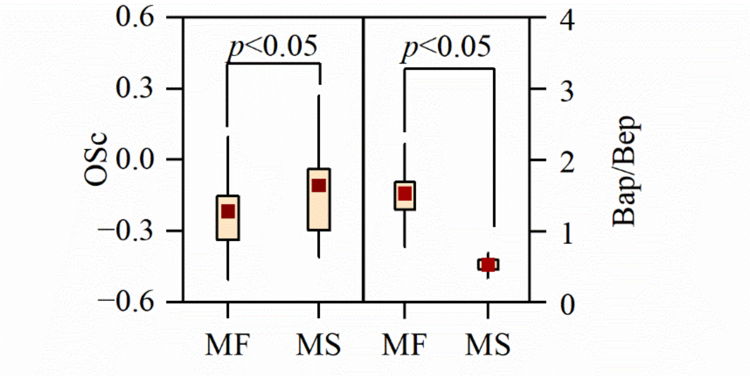

Figure S3 Comparison of OSc and BaP/BeP ratio among both sampling sites. The whisker boxes show mean (square), 25th–75th percentile ranges (box), and standard deviation values (whiskers)

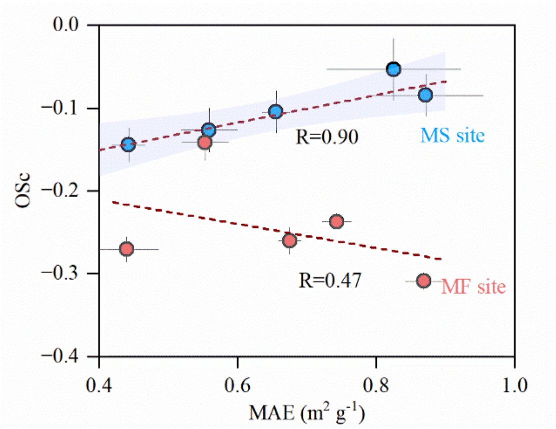

Figure S4. The correlativity between MAE_{365nm} and OSc value at both sampling sites.

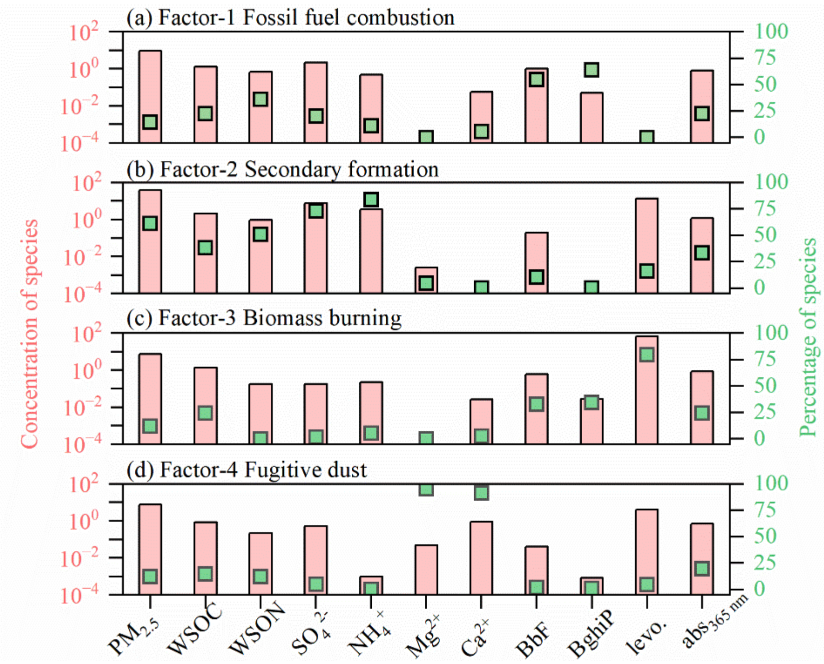

Figure S5. Source apportionment for light absorption of daytime water-soluble BrC ($abs_{365\ nm}$) of all the daytime samples collected during the whole campagin.

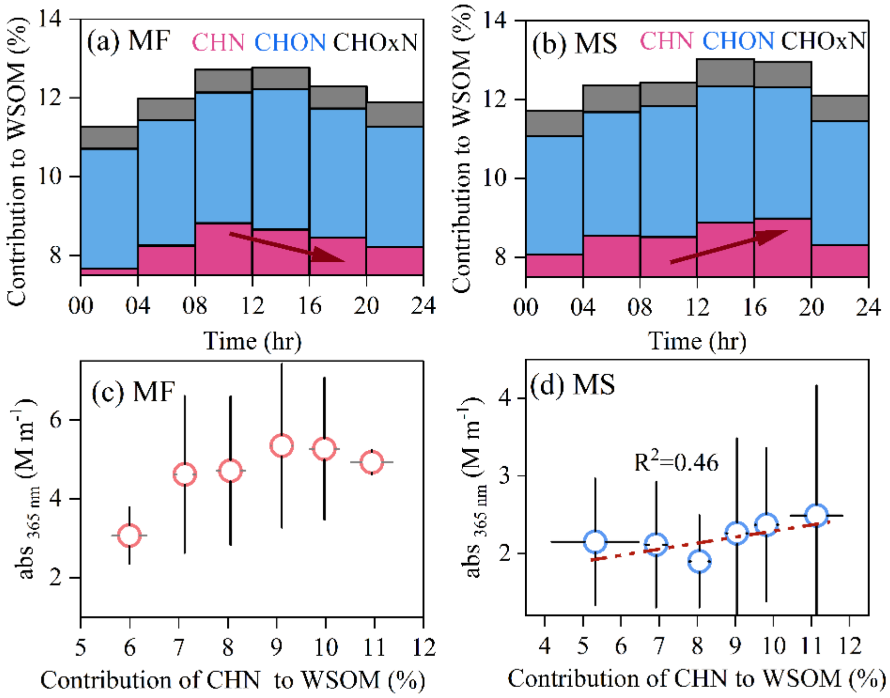

Figure S6. (a) and (b) Diurnal variations in relative contributions of nitrogen-containing organic fragments to WSOM at MF and MS sites, respetively. (c and d) The dependence of light absorption of WSOC ($Abs_{\lambda=365\ nm}$) on the relative contributions of CHN fragments to WSOM at the two sampling sites.

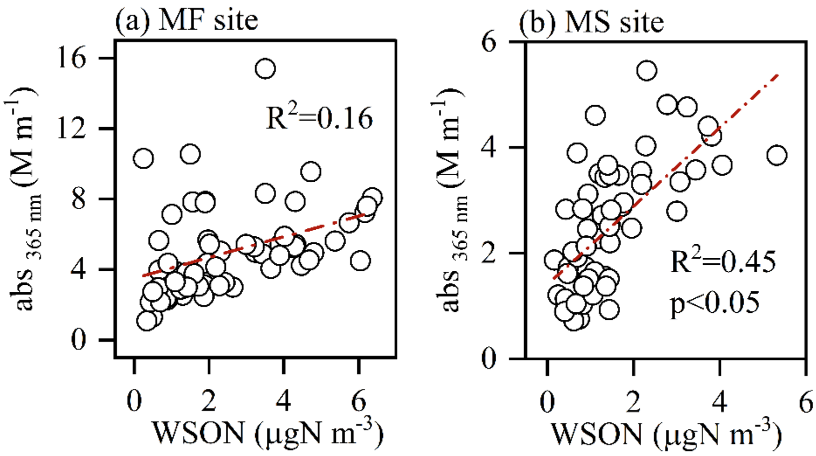

Figure S7 Linear fit regression analysis for Abs $_{\lambda=365\ nm}$ and WSON of the daytime samples at two sites

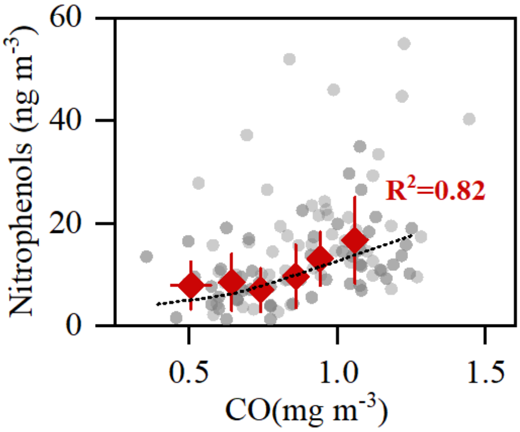

Figure S8 A correlation analysis of CO and NACs at MF site.

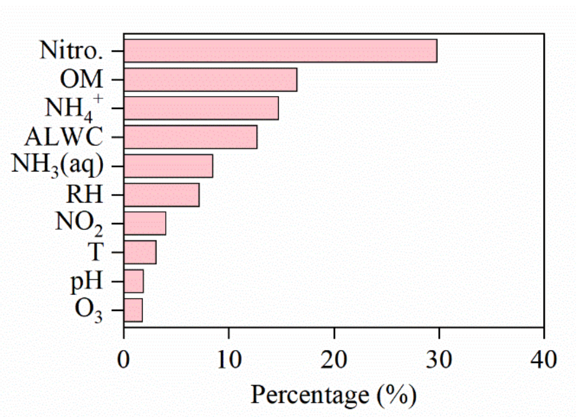

Figure S9 Importance assessment for the key factors affecting the daytime WSON at MF site.

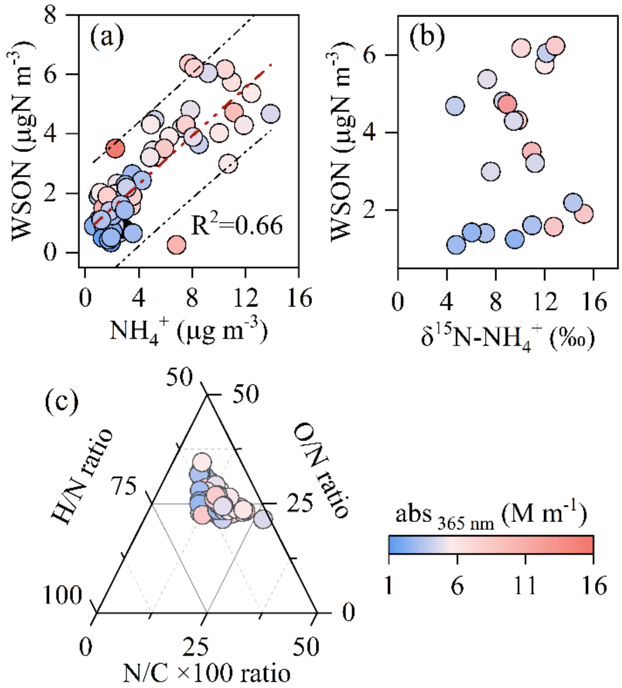

Figure S10 Impacts on WSON formation at MF site. Linear fit regressions for WSON with NH_4^++NH_3(aq) (a) and δ-^{15}N-NH_4^+ (b) at MF site and triangular chart for the elemental ratios of N/C, H/N and O/N of WSOC at MF site (c).

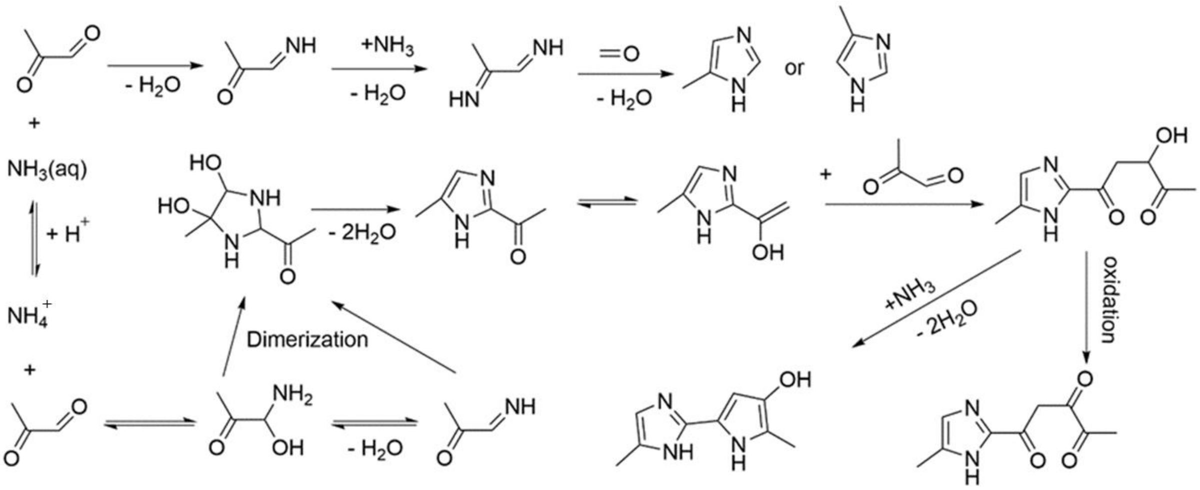

Figure S11 Simple reaction paths for imidazoles or N-heterocycles (Modified from Aiona et al. (2017) and Jang et al. (2013)).

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