

Supporting information to
Influence of Rain on the Abundance and Size Distribution of
Bioaerosols

Chathurika. M. Rathnayake,¹ Nervana. Metwali,² Thilina Jayarathne,¹ Josh Kettler,¹ Yuefan Huang,¹ Peter S. Thorne,^{2,3} Patrick T. O'Shaughnessy,^{2,3} and Elizabeth A. Stone¹

¹ Department of Chemistry, University of Iowa, Iowa 52242, United States.

² Occupational and Environmental Health, University of Iowa, Iowa 52242, United States.

³ Civil and Environmental Engineering, University of Iowa, Iowa 52242, United States.

Table S1: Detection limits and spike recoveries (\pm one standard deviation) of carbohydrates measured by High Performance Anion Exchange Chromatography with Pulsed Amperometric Detector.

Compound	Instrument detection limit (ppb) ¹	Method detection limit (ppb) ²	Spike recovery (%) (n=12)
Erythritol	1.0	5.9	99 \pm 6
Levoglucosan	2.0	5.8	102 \pm 5
Mannitol	0.6	1.7	100 \pm 6
Arabinose	1.9	3.3	103 \pm 5
Glucose	0.9	3.8	101 \pm 4
Fructose	0.6	2.8	100 \pm 5
Sucrose	0.7	3.7	102 \pm 6
Rhamnose	0.5	1.2	94 \pm 6

¹Instrumental detection limits were determined as three times the standard deviation of seven replicate injections of the lowest calibration point on the standard curve.

²Method detection limits were determined as three times the standard deviation of analyte concentrations recovered from seven spiked filter sample.

Table S2: Temperature, precipitation, PM concentrations and their size distribution in Iowa City from 17 April – 9 May, from 2010 to 2015.

Year	Temperature (°C)			Cumulative precipitation (mm)	PM _{2.5} (µg m ⁻³)			PM ₁₀ (µg m ⁻³)			Avg. fine fraction (%)	Std. Dev.	Reference
	Avg. temp. (°C)	±	Std. Dev.		Avg.	±	Std. Dev.	Range	Avg.	±			
2013			NA		7.1	± 3.0	1.8 - 13	15	± 8.9	2.0 - 32	57	± 17	This study
2010	13	± 3.4		93	8.2	± 6.2	3.2 - 25	20	± 12	7.0 - 38	61	± 22	EPA AQS*
2011	10	± 4.8		41	5.6	± 2.3	2.5 - 11	14	± 9.0	4.0 - 30	51	± 30	EPA AQS*
2012	14	± 5.2		111	8.3	± 5.1	3.4 - 26	21	± 9.3	10 - 37	55	± 18	EPA AQS*
2013	12	± 6.0		224	6.3	± 2.6	1.2 - 10	19	± 9.0	11 - 35	54	± 18	EPA AQS*
2014	13	± 4.2		93	7.5	± 4.3	1.1 - 15	18	± 7.8	7.0 - 30	51	± 18	EPA AQS*
2015	14	± 5.1		124	8.3	± 4.6	2.3 - 18	22	± 12	8.0 - 47	61	± 10	EPA AQS*

* PM_{2.5} averages were calculated using daily concentrations, and PM₁₀ averages were calculated using daily concentrations of 1-in-3 day sampling.

NA - not applicable

Table S3: Temperature, precipitation, PM concentrations and their size distribution in Iowa City from 15 August – 4 September, from 2010 to 2015.

Year	Avg. temp. (°C)			Cumulative precipitation (mm)	PM _{2.5} (µg m ⁻³)			PM ₁₀ (µg m ⁻³)			Avg. fine fraction (%)	Std. Dev	Reference
	Avg.	±	Std. Dev		Avg.	±	Std. Dev	Range	Avg.	±			
2013			NA		12	± 3.6	3.4 - 17	33	± 8.3	21 - 49	39	± 12	This study
2010	23	± 3.0		57	9.9	± 4.2	4.3 - 19	27	± 13	16 - 52	36	± 14	EPA AQS*
2011	23	± 2.9		51	9.9	± 4.7	4.9 - 27	26	± 4.6	21 - 33	36	± 7.9	EPA AQS*
2012	23	± 3.0		36	9.3	± 3.3	4.0 - 17	28	± 9.6	15 - 39	38	± 9.3	EPA AQS*
2013	24	± 3.8		1.0	12	± 2.9	7.5 - 17	31	± 5.7	25 - 39	41	± 7.9	EPA AQS*
2014	23	± 2.2		115	9.8	± 4.0	4.6 - 18	23	± 11	10 - 40	41	± 11	EPA AQS*
2015	21	± 3.5		54	9.4	± 4.9	2.2 - 19	24	± 9.2	8 - 36	40	± 16	EPA AQS*

* PM_{2.5} averages were calculated using daily concentrations, and PM₁₀ averages were calculated using daily concentrations of 1-in-3 day sampling.
NA - not applicable

Table S4 : Daily concentrations of PM mass and bioaerosol tracers measured from 17 April –9 May in Iowa City, IA, USA.

Date in 2013	PM _{2.5}							PM _{10-2.5}						
	PM mass ($\mu\text{g m}^{-3}$)	Glucose (ng m^{-3})	Fructose (ng m^{-3})	Sucrose (ng m^{-3})	Mannitol (ng m^{-3})	Glucan (ng m^{-3})	Endotoxins (EU m^{-3})	PM mass ($\mu\text{g m}^{-3}$)	Glucose (ng m^{-3})	Fructose (ng m^{-3})	Sucrose (ng m^{-3})	Mannitol (ng m^{-3})	Glucan (ng m^{-3})	Endotoxins (EU m^{-3})
Apr 17	2.93	1.06	0.70	0.65	1.03	<0.7	0.002	<0.03	5.53	<1.03	<1.37	5.92	4.65	0.01
Apr 18	1.82	0.60	<0.41	<0.25	0.65	1.31	<0.002	0.14	8.61	1.31	1.21	6.72	2.48	0.00
Apr 19	2.79	<0.16	<0.41	<0.25	0.42	1.18	<0.002	0.97	<1.37	<1.03	2.62	8.40	2.63	0.02
Apr 20	5.26	<0.16	<0.41	<0.25	0.51	<0.7	<0.002	5.06	3.09	<1.03	<1.37	2.12	13.5	0.05
Apr 21	5.92	0.51	0.31	<0.25	0.84	<0.7	<0.002	4.09	6.91	1.47	7.64	4.16	5.51	0.02
Apr 22	8.71	0.66	<0.41	<0.25	0.99	1.80	<0.002	5.47	8.91	1.42	15.4	5.78	59.0	0.04
Apr 23	5.62	0.55	0.39	<0.25	1.06	2.66	<0.002	2.70	1.54	<1.03	<1.37	1.45	4.32	0.03
Apr 24	7.27	0.55	0.32	<0.25	1.51	<0.7	<0.002	4.67	2.41	<1.03	<1.37	2.68	3.57	0.04
Apr 25	6.90	<0.16	0.23	<0.25	0.53	0.83	<0.002	8.58	2.85	<1.03	2.65	3.93	6.27	0.03
Apr 26	6.56	1.63	0.23	0.30	2.18	1.00	<0.002	9.10	14.4	1.20	5.38	8.48	6.02	0.07
Apr 27	9.77	1.92	0.41	<0.25	5.24	<0.7	<0.002	12.6	7.9	<1.03	2.79	13.1	1.12	0.05
Apr 28	13.0	1.45	<0.41	<0.25	2.36	1.12	<0.002	14.2	17.9	1.61	7.21	14.8	6.47	0.06
Apr 29	11.5	3.72	<0.41	<0.25	3.59	2.66	0.005	16.6	10.2	0.69	19.5	22.8	10.4	0.07
Apr 30	9.08	2.17	<0.41	0.39	2.48	<0.7	0.017	22.6	17.5	3.00	36.8	22.9	9.27	0.10
May 1	8.79	0.90	0.36	0.72	1.38	7.10	0.020	16.8	9.08	2.34	15.8	8.46	5.97	0.09
May 2	1.77	29.9	21.3	52.6	3.68	<0.7	<0.002	0.40	5.98	2.03	4.17	3.13	9.30	0.02
May 3	5.07	4.58	2.05	4.03	1.92	1.01	<0.002	1.41	11.1	<1.03	<1.37	10.4	13.3	0.01
May 4	6.65	8.81	3.05	3.88	3.47	<0.7	0.007	2.28	11.9	1.27	3.78	41.1	15.3	0.06
May 5	9.10	2.46	<0.41	<0.25	1.82	1.70	0.003	8.44	40.4	5.14	18.8	76.6	101	0.04
May 6	7.51	2.80	<0.41	0.31	2.31	2.01	0.007	8.79	19.1	4.53	22.9	55.6	21.9	0.04
May 7	11.1	2.71	0.34	0.81	3.01	0.90	0.002	13.7	19.0	5.60	15.7	63.8	45.9	0.04
May 8	9.55	1.35	<0.41	<0.25	1.52	<0.7	0.012	10.5	31.5	5.19	12.8	68.0	48.5	0.10
May 9	6.50	8.67	0.47	0.31	3.53	3.01	<0.002	1.73	53.9	3.64	6.76	68.8	8.40	0.06

Table S5 : Daily concentrations of PM mass and bioaerosol tracers measured from 15 August – 4 September in Iowa City, IA, USA.

Date in 2013	PM _{2.5}							PM _{10-2.5}						
	PM mass (µg m ⁻³)	Glucose (ng m ⁻³)	Fructose (ng m ⁻³)	Sucrose (ng m ⁻³)	Mannitol (ng m ⁻³)	Glucan (ng m ⁻³)	Endotoxins (EU m ⁻³)	PM mass (µg m ⁻³)	Glucose (ng m ⁻³)	Fructose (ng m ⁻³)	Sucrose (ng m ⁻³)	Mannitol (ng m ⁻³)	Glucan (ng m ⁻³)	Endotoxins (EU m ⁻³)
Aug 15	11.9	4.59	<0.46	<0.28	5.26	2.09	0.002	14.2	17.8	<1.03	4.66	32.7	10.3	0.08
Aug 16	10.1	3.41	<0.46	0.56	4.30	0.61	<0.002	22.9	15.9	<1.03	2.99	30.6	9.81	0.06
Aug 17	11.1	4.88	<0.46	<0.28	10.3	2.47	0.002	13.1	23.2	<1.03	9.98	44.3	15.7	0.1
Aug 18	12.5	4.41	<0.46	0.57	5.27	2.40	0.002	12.9	18.3	1.68	9.56	45.7	8.83	0.05
Aug 19	16.2	8.74	<0.46	<0.28	6.58	<0.70	0.003	15.1	29.5	4.59	7.96	62.1	<2.0	<0.004
Aug 20	13.8	2.60	<0.46	<0.28	1.96	1.73	0.005	19.5	26.3	3.6	29.4	42.3	10.2	0.11
Aug 21	15.9	5.17	<0.46	0.53	5.74	2.02	0.005	24.5	27.7	4.6	21.5	62.1	12.0	0.07
Aug 22	12.2	19.1	<0.46	<0.28	16.4	2.85	0.076	12.9	34.4	2.93	12.6	105	13.7	0.14
Aug 23	11.1	6.11	<0.46	<0.28	3.98	2.58	0.006	11.2	34.3	3.92	19.1	45.3	10.9	0.06
Aug 24	16.7	5.63	<0.46	<0.28	3.82	2.53	0.001	13.6	33.5	5.35	19.6	40.4	43.9	0.07
Aug 25	15.0	5.20	<0.46	<0.28	3.73	1.83	0.007	16.8	29.7	4.99	51.8	57.3	14.9	0.17
Aug 26	16.1	3.51	<0.46	<0.28	3.75	1.06	0.005	20.6	31.6	4.5	16.9	104	8.12	0.06
Aug 27	11.9	10.2	<0.46	1.28	5.86	2.21	0.026	28.9	77.5	6.78	33.1	64.9	6.95	0.16
Aug 28	15.7	3.24	<0.46	0.42	2.54	3.65	0.006	27.2	25.7	4.32	16.5	43.2	4.7	0.07
Aug 29	14.3	4.34	<0.46	<0.28	3.27	4.03	<0.002	17.2	19.2	2.5	5.39	35.9	3.27	0.07
Aug 30	10.8	5.99	<0.46	0.56	7.15	29.1	0.015	37.1	34.8	2.99	8.22	80.0	5.65	0.32
Aug 31 - Sep 1	15.2	5.47	<0.46	<0.28	7.73	22.1	0.008	34.1	65.4	6.00	6.02	177	42.2	0.39
Sep 2	3.4	3.58	<0.46	<0.28	3.35	2.09	0.011	17.9	17.3	<1.03	7.60	29.0	6.19	<0.004
Sep 3	5.59	2.44	<0.46	<0.28	3.30	0.61	0.009	19.4	10.8	<1.03	<1.37	23.1	<2.0	<0.004
Sep 4	7.58	3.31	<0.46	<0.28	13.2	2.47	0.005	29.5	13.7	<1.03	2.68	45.1	3.69	0.14

Table S6 : Absolute and percent contributions of pollens and fungal spores to PM_{2.5} and PM₁₀ mass during spring in Iowa City, IA, USA estimated by CMB modeling. Uncertainties in bioaerosol contributions represent standard errors of the estimate.

Date in 2013	Start time (local)	End time (local)	PM _{2.5} Pollens		PM _{2.5} Fungal spores		PM ₁₀ Pollens		PM ₁₀ Fungal spores	
			Mass ± Error (µg m ⁻³)	(%)	Mass ± Error (µg m ⁻³)	(%)	Mass ± Error (µg m ⁻³)	(%)	Mass ± Error (µg m ⁻³)	(%)
Apr 17	8:04	8:10	0.016 ± 0.004	(0.54)	0.019 ± 0.004	0.66	0.012 ± 0.005	(0.42)	0.134 ± 0.029	4.57
Apr 18	8:20	7:50	0.004 ± 0.004	(0.21)	0.014 ± 0.003	0.75	0.072 ± 0.024	(3.65)	0.143 ± 0.030	7.32
Apr 19	7:52	7:57	<0.002 ± 0.003	(<0.09)	0.008 ± 0.003	0.28	0.001 ± 0.004	(0.04)	0.171 ± 0.035	4.55
Apr 20	8:00	8:18	<0.002 ± 0.003	(<0.05)	0.010 ± 0.003	0.18	0.0002 ± 0.002	(0.002)	0.050 ± 0.021	0.49
Apr 21	8:34	7:41	0.004 ± 0.004	(0.07)	0.016 ± 0.004	0.26	0.122 ± 0.027	(1.22)	0.097 ± 0.026	0.97
Apr 22	7:44	7:32	0.004 ± 0.004	(0.05)	0.019 ± 0.004	0.22	0.175 ± 0.030	(1.23)	0.131 ± 0.029	0.93
Apr 23	7:40	7:47	0.003 ± 0.004	(0.05)	0.021 ± 0.006	0.38	0.0004 ± 0.002	(0.005)	0.050 ± 0.021	0.61
Apr 24	7:50	8:10	0.005 ± 0.004	(0.07)	0.029 ± 0.005	0.40	0.0005 ± 0.002	(0.005)	0.082 ± 0.022	0.68
Apr 25	8:15	7:34	0.0003 ± 0.004	(0.005)	0.010 ± 0.003	0.14	0.022 ± 0.014	(0.14)	0.085 ± 0.023	0.55
Apr 26	7:36	8:18	0.016 ± 0.004	(0.24)	0.043 ± 0.008	0.65	0.142 ± 0.028	(0.91)	0.207 ± 0.038	1.32
Apr 27	8:26	9:04	0.015 ± 0.004	(0.16)	0.101 ± 0.018	1.03	0.041 ± 0.015	(0.18)	0.355 ± 0.060	1.59
Apr 28	9:28	7:43	0.010 ± 0.004	(0.07)	0.047 ± 0.009	0.36	0.162 ± 0.031	(0.59)	0.333 ± 0.059	1.22
Apr 29	7:46	8:20	0.013 ± 0.004	(0.11)	0.070 ± 0.013	0.61	0.188 ± 0.022	(0.67)	0.512 ± 0.086	1.82
Apr 30	8:29	7:33	0.018 ± 0.004	(0.20)	0.048 ± 0.009	0.53	0.351 ± 0.047	(1.11)	0.494 ± 0.086	1.56
May 1	7:41	8:18	0.015 ± 0.004	(0.17)	0.027 ± 0.005	0.31	0.203 ± 0.032	(0.79)	0.191 ± 0.037	0.75
May 2	8:25	7:25	0.739 ± 0.081	(41.70)	0.069 ± 0.013	3.92	0.832 ± 0.092	(38.38)	0.129 ± 0.029	5.96
May 3	7:38	7:54	0.079 ± 0.011	(1.56)	0.037 ± 0.008	0.72	0.080 ± 0.012	(1.24)	0.238 ± 0.043	3.68
May 4	8:17	8:08	0.109 ± 0.014	(1.64)	0.068 ± 0.013	1.02	0.212 ± 0.032	(2.37)	0.865 ± 0.147	9.69
May 5	8:31	7:49	0.017 ± 0.004	(0.18)	0.035 ± 0.007	0.38	0.348 ± 0.047	(1.98)	1.521 ± 0.263	8.67
May 6	7:50	8:00	0.013 ± 0.004	(0.17)	0.045 ± 0.008	0.59	0.328 ± 0.043	(2.01)	1.123 ± 0.193	6.89
May 7	8:05	7:41	0.032 ± 0.005	(0.28)	0.058 ± 0.010	0.52	0.303 ± 0.041	(1.22)	1.296 ± 0.223	5.22
May 8	7:43	9:45	0.008 ± 0.004	(0.09)	0.029 ± 0.005	0.30	0.274 ± 0.038	(1.36)	1.348 ± 0.233	6.72
May 9	9:49	7:42	0.018 ± 0.005	(0.27)	0.068 ± 0.013	1.04	0.210 ± 0.037	(2.55)	1.403 ± 0.240	17.04

Table S7 : Absolute and percent contributions of fungal spores to PM_{2.5} and PM_{10-2.5} mass during late summer in Iowa City, IA, USA estimated using 1.7 pg mannitol spore⁻¹ (range from 1.2-2.4 pg mannitol spore⁻¹) and 33 pg spore⁻¹ in Bauer et al. (2008). Uncertainties in bioaerosol contributions represent standard errors of the estimate.

Date in 2013	Start time (local)	End time (local)	PM _{2.5} Fungal spores		PM _{10-2.5} Fungal spores	
			Mass ± Error (µg m ⁻³)	(%)	Mass ± Error (µg m ⁻³)	(%)
15-Aug	8:29	8:44	0.10 ± 0.02	(0.86)	0.63 ± 0.11	(4.47)
16-Aug	9:47	9:46	0.08 ± 0.01	(0.83)	0.59 ± 0.10	(2.59)
17-Aug	10:18	9:04	0.20 ± 0.03	(1.80)	0.86 ± 0.15	(6.56)
18-Aug	9:20	8:32	0.10 ± 0.02	(0.82)	0.89 ± 0.15	(6.88)
19-Aug	8:55	8:08	0.13 ± 0.02	(0.79)	1.21 ± 0.21	(7.98)
20-Aug	8:14	7:17	0.04 ± 0.01	(0.28)	0.82 ± 0.14	(4.21)
21-Aug	7:22	7:57	0.11 ± 0.02	(0.70)	1.21 ± 0.21	(4.92)
22-Aug	8:01	7:21	0.32 ± 0.05	(2.61)	2.04 ± 0.35	(15.80)
23-Aug	7:24	8:11	0.08 ± 0.01	(0.70)	0.88 ± 0.15	(7.85)
24-Aug	8:20	8:31	0.07 ± 0.01	(0.45)	0.78 ± 0.14	(5.77)
25-Aug	8:41	7:17	0.07 ± 0.01	(0.48)	1.11 ± 0.19	(6.62)
26-Aug	7:20	7:27	0.07 ± 0.01	(0.45)	2.02 ± 0.35	(9.80)
27-Aug	7:33	7:22	0.11 ± 0.02	(0.96)	1.26 ± 0.22	(4.36)
28-Aug	7:26	7:27	0.05 ± 0.01	(0.31)	0.84 ± 0.15	(3.08)
29-Aug	7:32	7:29	0.06 ± 0.01	(0.44)	0.70 ± 0.12	(4.05)
30-Aug	7:32	6:24	0.14 ± 0.02	(1.29)	1.55 ± 0.27	(4.19)
31-Aug -						
01-Sep	6:28	7:46	0.15 ± 0.03	(0.99)	3.44 ± 0.60	(10.07)
2-Sep	7:50	7:40	0.07 ± 0.01	(1.91)	0.56 ± 0.10	(3.14)
3-Sep	7:43	7:29	0.06 ± 0.01	(1.15)	0.45 ± 0.08	(2.31)
4-Sep	7:32	7:25	0.26 ± 0.04	(3.38)	0.88 ± 0.15	(2.97)

Figure S1: Comparison of CMB calculated R^2 values, which represent the fraction of the variance in the ambient measurements explained by the model, for PM_{10} (a) and $PM_{2.5}$ (b) for various pollen profiles. The comparison is limited to 26 April – 9 May, after the onset of the pollen season. Results indicated that the oak pollens fit the ambient data better than birch or willow, with the most variance explained by the combination of oak pollens and fungal spores.

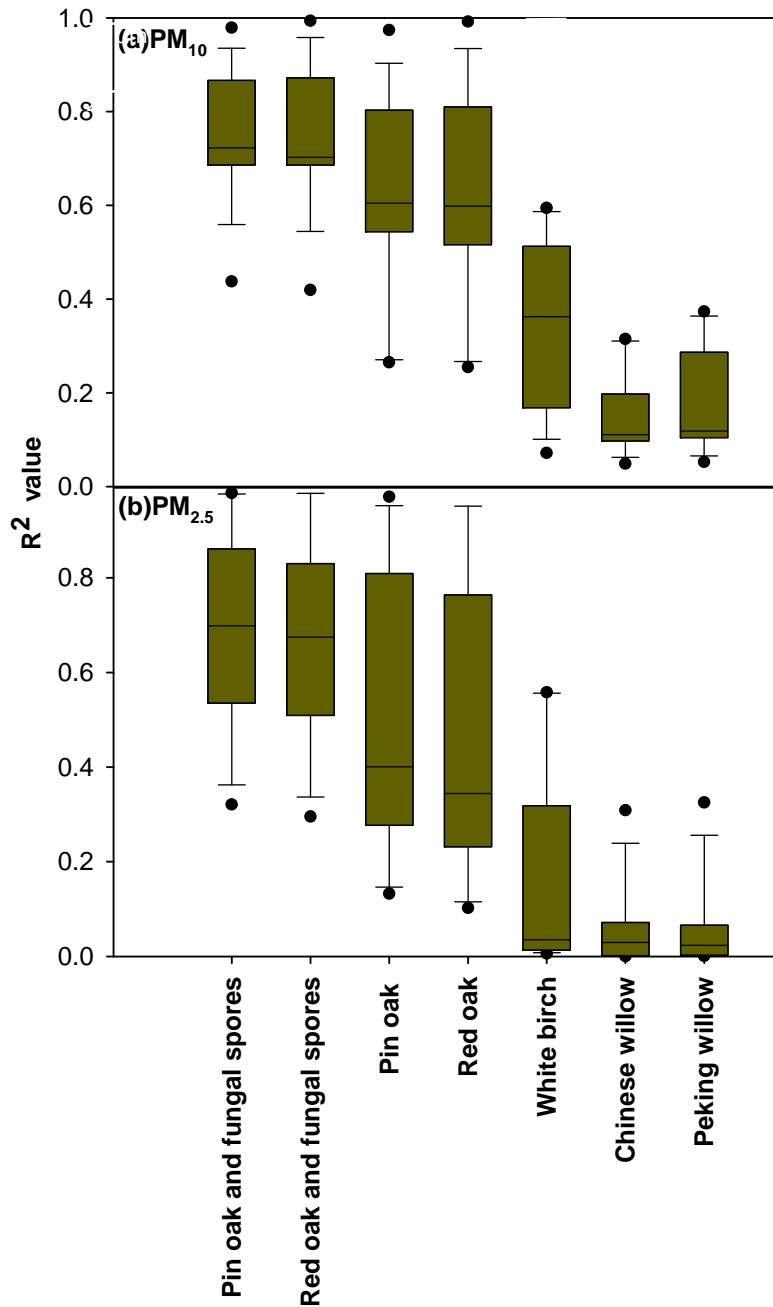
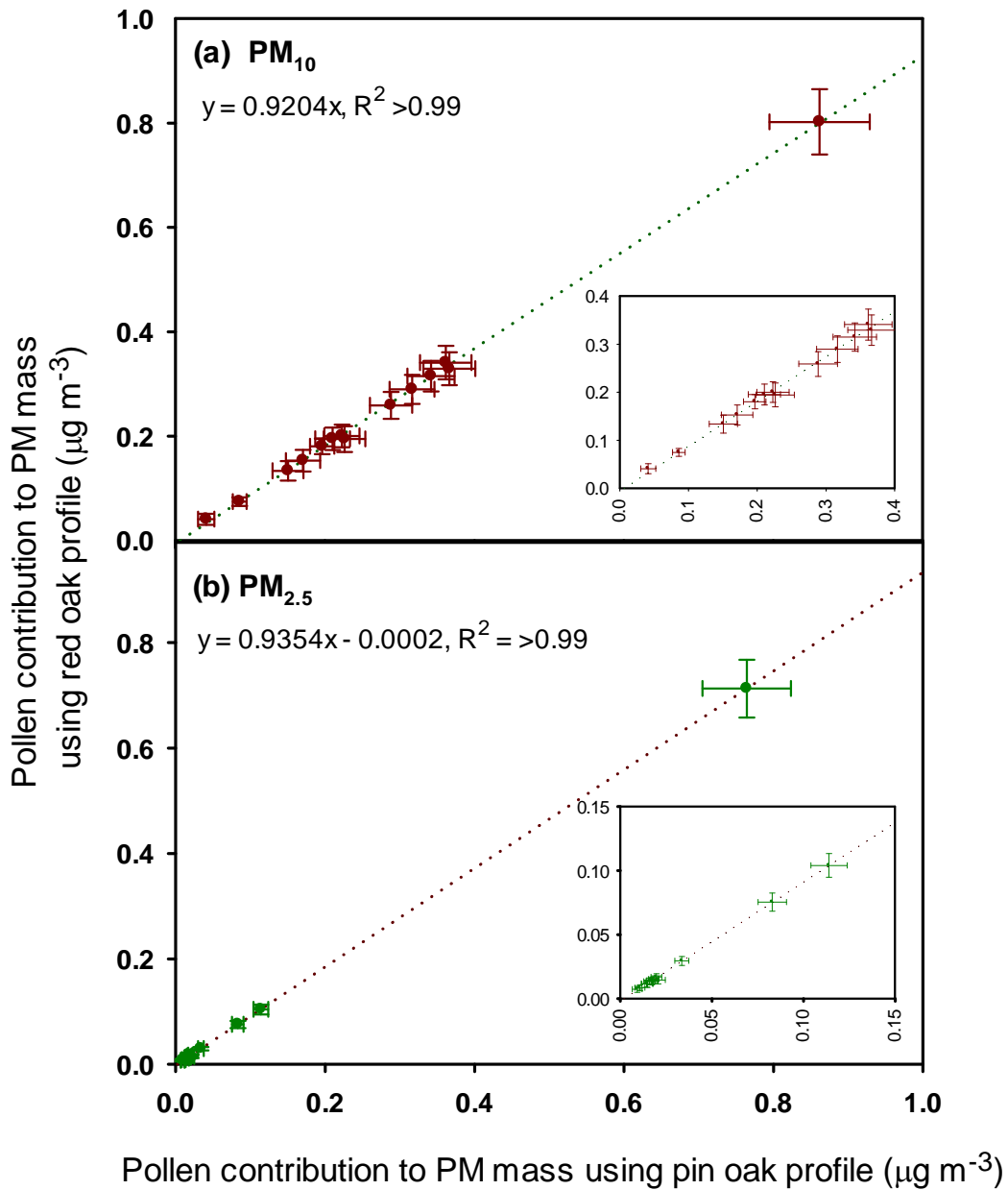


Figure S2: Comparison of pollen contributions to PM₁₀ (a) and PM_{2.5} (b) mass, estimated by the CMB model using pin oak and red oak pollen profiles. The comparison is limited to 26 April – 9 May, after the onset of the pollen season. The strong correlation ($R^2 > 0.99$) and numerical agreement (slopes close to 1) demonstrate the consistency in model results for when either profile is used. Consequently, source contributions from these two types were averaged to obtain the best estimate of pollen contributions to PM mass.



Works Cited

Bauer, H., Claeys, M., Vermeylen, R., Schueller, E., Weinke, G., Berger, A., and Puxbaum, H.: Arabitol and mannitol as tracers for the quantification of airborne fungal spores, *Atmos. Environ.*, 42, 588-593, 10.1016/j.atmosenv.2007.10.013, 2008.