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Changes in the hospitalization and ED visit rates for respiratory diseases associated with source-specific $PM_{2.5}$ in New York State from 2005 to 2016

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ABSTRACT

Prior work found increased rates for emergency department (ED) visits for asthma and hospitalizations for chronic obstructive pulmonary disease per unit mass of $PM_{2.5}$ across New York State (NYS) during 2014–2016 after significant reductions in ambient $PM_{2.5}$ concentrations had occurred following implementation of various policy actions and major economic disruptions. The associations of source-specific $PM_{2.5}$ concentrations with these respiratory diseases were assessed with a time-stratified case-cossover design and logistic regression models to identify the changes in the $PM_{2.5}$ that have led to the apparently increased toxicity per unit mass. The rates of ED visits and hospitalizations for asthma and COPD associated with increases in source-specific $PM_{2.5}$ concentrations in the prior 1, 4, and 7 days were estimated for 6 urban sites in New York State. Overall, there were similar numbers of significantly increased (n = 9) and decreased rates (n = 8) of respiratory events (asthma and COPD hospitalizations and ED visits) associated with increased source-specific $PM_{2.5}$ concentrations in the previous 1, 4, and 7 days. Associations of source-specific $PM_{2.5}$ concentrations with excess rates of hospitalizations for COPD for spark- and compression ignition vehicles increased in the 2014–2016 period, but the values were not statistically significant. Other source types showed inconsistent patterns of excess rates. For asthma ED visits, only biomass burning and road dust showed consistent positive associations with road dust having significant values for most lag times. Secondary nitrate also showed significant positive associations with asthma ED visits in the AFTER period compared to no associations in the prior periods. These results suggest that the relationships of asthma and COPD exacerbation with source-specific $PM_{2.5}$ are not well defined and further work will be needed to determine the causes of the apparent increases in the per unit mass toxicity of $PM_{2.5}$ in New York State in the 2014-16 period.

1. Introduction

The exacerbation of asthma and chronic obstructive pulmonary disease (COPD consisting of chronic bronchitis and emphysema) by exposure to ambient airborne particulate matter has been extensively reported and reviewed (Guarnieri and Balmes, 2014; Orellano et al., 2017; Anenberg et al., 2018; Li et al., 2016; Bloemsma et al., 2016; USEPA, 2018). Our recent study (Hopke et al., 2019) also found increased rates of hospitalizations and emergency department (ED) visits in New York State (NYS) associated with short term increases in ambient $PM_{2.5}$ concentrations. However, there were changes in the estimated excess rates ([relative rate – 1]*100%) of COPD hospitalization and adult asthma ED visits per unit mass of $PM_{2.5}$ concentration from 2005 to 2016, with the largest relative rates per $\mu g/m^3$ increase in $PM_{2.5}$ in the most recent (2014–2016) period compared to earlier periods (2005–2007; 2008–2013). Similar findings were observed in parallel studies of acute cardiovascular events (CVD) (Zhang et al., 2018) and respiratory infections (Croft et al., 2019a) across NYS. Overall rates of CVD hospitalizations (Zhang et al., 2018), adult asthma ED visits and COPD hospitalizations (Hopke, 2019), and influenza ED visits and

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culture-negative pneumonia hospitalizations decreased with decreasing $PM_{2.5}$ concentrations across NYS from 2005 to 2016 (Squizzato et al., 2018a). However, the rates did not decline proportionally to the declining $PM_{2.5}$ concentrations suggesting that the observed changes in PM composition (Squizzato et al., 2018b) resulted in increased PM toxicity.

Subsequently, Rich et al. (2019) reported the rates of these CVD hospitalizations were associated with source-specific $PM_{2.5}$ concentrations based on the source apportionment of speciated PM across NYS (Squizzato et al., 2018b). Analyses of U.S. Environmental Protection Agency Chemical Speciation Network data (Solomon et al., 2014) were performed for each site using positive matrix factorization (PMF) (Squizzato et al., 2018b) to provide estimates of the source-specific $PM_{2.5}$ concentrations. Interquartile range (IQR) increases in spark-ignition vehicular emissions (GAS) concentrations were associated with increased excess rates of cardiac arrhythmia hospitalizations (2.3%; 95% CI = 0.4%, 4.2%; IQR = 2.56 $\mu g/m^3$) and ischemic stroke hospitalizations (3.7%; 95% CI = 1.1%, 6.4%; 2.73 $\mu g/m^3$) on lag day 0. IQR increases in diesel (DIE) concentrations were associated with increased rates of congestive heart failure hospitalizations (0.7%; 95% CI = 0.2% 1.3%; 0.51 $\mu g/m^3$) and ischemic heart disease hospitalizations (0.8%; 95% CI = 0.3%, 1.3%; 0.60 $\mu g/m^3$) on lag day 0. Increased acute cardiovascular hospitalization rates were also associated with IQR increases in concentrations of road dust (RD), residual oil (RO), and secondary nitrate (SN) over lag days 0, 0–3, and 0–6, but not other sources including biomass burning (BB), secondary sulfate (SS), pyrolytic organic carbon (OP), and fresh (FSS) and aged sea salt (AGS) (Rich et al., 2019). These findings suggested that several sources of $PM_{2.5}$ in New York State (i.e. tailpipe emissions, non-tailpipe emissions such as brake and tire wear, residual oil, and nitrate that may also reflect traffic emissions) trigger acute cardiovascular events.

Croft et al. (2019b) has done similar analyses of the associations of source-specific $PM_{2.5}$ with hospitalizations and emergency department visits for respiratory infections (culture negative pneumonia or influenza). Increased rates of ED visits for influenza were associated with interquartile range increases in concentrations of GAS (Excess Rate [ER] = 9.2%; 95% CI: 4.3%, 14.3%) and DIE (ER = 3.9%; 95% CI: 1.1%, 6.8%) from exposure during lag days 0–3. There were similar associations between BB, SS, OP, and RO, influenza ED visits and hospitalizations, but not culture negative pneumonia hospitalizations or ED visits. They concluded that increased rates of influenza hospitalizations and ED visits that were associated with short term increases in $PM_{2.5}$ appear to be driven largely by PM from traffic and other combustion sources.

As part of the prior analyses (Zhang et al., 2018; Croft et al., 2019a), the measured organic carbon was separated into primary organic carbon (POC) and secondary organic carbon (SOC). It was hypothesized that SOC in the particles from these sources was likely to include reactive oxygen species (ROS) (Docherty et al., 2005; Hopke, 2015; Pagonis and Ziemann, 2018) or have substantial oxidative potential (Saffari et al., 2014) that could induce oxidative stress and systemic inflammation. Recent changes in light-duty vehicles from port fuel injection (PFI) to gasoline direct injection (GDI) technology coupled with changes in gasoline formulations appear to increase the formation of secondary organic aerosol (SOA) (Zhao et al., 2014, 2015; 2016, 2018) and thus, the potential for increased ROS associated with spark-ignition vehicular emissions (GAS). GAS PM had significant associations with several CVD outcomes as well as hospitalizations and ED visits for influenza. Diesel emissions (DIE) are reported to have substantial oxidative potential (Arimoto et al., 2005; Saffari et al., 2014) that would produce endogenous ROS (Hopke, 2015) and DIE PM was also associated with CVD and influenza rates of hospitalization and ED visits. It is not clear if the presence of oxidants or oxidative potential in $PM_{2.5}$ affects the rates of hospitalizations or ED visits for either asthma or COPD.

Therefore, a further study of whether these source-specific $PM_{2.5}$ components also exacerbate adult asthma and/or COPD leading to hospitalizations or ED visits was conducted. We hypothesized that the ROS-associated source components such as spark-ignition and diesel vehicle emissions, road dust, and residual oil combustion particles would be associated with increased rates of hospitalizations and ED visits for these respiratory diseases.

2. Methods and materials

2.1. Exposure and Meteorological Data

We have used the same approach in this study as that described by Rich et al. (2019) and Croft et al. (2019b) to obtain the exposure and associated weather data. The source-specific $PM_{2.5}$ concentrations were estimated from the chemical speciation data obtained from the analysis of samples collected at six urban sites in NYS (Albany, Bronx, Buffalo, Manhattan, Queens, and Rochester). The locations and details of sample collection for these sites are presented in Table S1 in the supplemental material file. Details of the sample collection and analyses were presented by Solomon et al. (2014). The PMF analyses and resulting source identifications were presented by Squizzato et al. (2018b) while the trends in the source contributions were reported by Masiol et al. (2019). Because of the limited sample collection schedules, source-specific $PM_{2.5}$ concentrations were available only every 3rd or 6th day depending on the site (Table S1). Minimum values for the source specific mass concentrations often have small negative values as a result of the uncertainties in the PMF analysis (Paatero et al., 2014). However, we did not left censor the values to avoid the potential bias that such truncation would induces (e.g., Cain et al., 2011).

Hourly temperature and relative humidity data were obtained from the National Weather Service (National Climate Data Center, https://www.ncdc.noaa.gov/cdo-web/datatools/lcd) for the nearest major airport (BUF - Buffalo, ROC - Rochester, ALB - Albany, LGA - Bronx, and JFK - Queens) or the closest weather station (Central Park for Manhattan). For each study subject living within 15 miles of our six monitoring stations, we assigned $PM_{2.5}$ source contributions, temperature, and relative humidity measurements from the nearest applicable monitoring site. If a person lived < 15 miles from more than one monitor (e.g. Bronx vs. Manhattan), we assigned concentrations/values from the closer monitor to that person.

2.2. Study Population and Hospital Admissions Data

The hospitalization and ED visit data used have been previously described by Zhang et al. (2018), Croft et al. (2019a), and Hopke et al. (2019). Hospital admissions and ED visit records were obtained from the New York State Department of Health Statewide Planning and Research Cooperative System (SPARCS) database, which covers ~95% of hospitals in NYS, but not federal (e.g. Veterans Affairs Hospitals) and psychiatric facilities. The SPARCS data include a primary diagnoses and up to 24 other comorbidity diagnoses assigned at the time of hospital admission or ED visit as well as patient demographic information. Using each residential address for every subject hospitalization or ED visit, we retained those adult subjects (≥18 years old) with 1) a successfully geocoded address (using ArcGIS 10.3.1; ©ESRI, Inc. Redlands, CA) that was within 15 miles of each monitoring station (Buffalo, Rochester, Albany, Bronx, Manhattan, Queens); 2) a "principal" diagnoses of asthma (ICD9 = 493; ICD10 = J45) or COPD (ICD9 = 491, 492; ICD10 = J41, J43); and 3) an admission date between January 1, 2005 and December 31, 2016 (N = 43,315 asthma hospitalizations; N = 128,055 asthma ED visits; N = 32,240 COPD hospitalizations; N = 10,854). This study was reviewed and approved by the Institutional Review Board at the State University of New York at Albany.

Table 1a
Characterization of the asthma patients requiring hospitalizations.

	Albany (N)	Albany (%)	Bronx (N)	Bronx (%)	Buffalo (N)	Buffalo (%)	Manhattan (N)	Manhattan (%)	Queens (N)	Queens (%)	Rochester (N)	Rochester (%)
N	833	100	16,515	100	880	100	13,482	100	10,078	100	1527	100
Male	237	28.45	4932	29.86	223	25.34	4112	30.5	3087	30.63	401	26.26
AGE (years; mean (STD))	53.86 (18.16)		53.39 (17.59)		55.88 (17.92)		55.93 (17.71)		57.45 (18.32)		53.53 (17.26)	
18-39	185	22.21	3552	21.51	154	17.5	2404	17.83	1676	16.63	302	19.78
40-49	196	23.53	3339	20.22	177	20.11	2375	17.62	1685	16.72	324	21.22
50-69	156	18.73	3658	22.15	193	21.93	3068	22.76	2131	21.15	371	24.3
60-69	125	15.01	2716	16.45	138	15.68	2379	17.65	1760	17.46	254	16.63
70-79	74	8.88	1968	11.92	109	12.39	1844	13.68	1488	14.76	139	9.1
≥80	97	11.64	1282	7.76	109	12.39	1412	10.47	1338	13.28	137	8.97
RACE/ETHNICITY												
White	516	61.94	2722	16.48	514	58.41	3464	25.69	3553	35.26	727	47.61
Black	248	29.77	5517	33.41	273	31.02	6097	45.22	3733	37.04	601	39.36
American Indian	1	0.12	69	0.42	6	0.68	64	0.47	81	0.80	1	0.07
Asian	1	0.12	104	0.63	4	0.45	298	2.21	404	4.01	5	0.33
Native Hawaii	1	0.12	1	0.01	0	0.00	3	0.02	6	0.06	0	0
Hispanic	29	3.48	4997	30.26	61	6.93	2682	19.89	1493	14.81	130	8.51
YEAR												
2005	N/A	N/A	2129	12.89	76	8.64	1687	12.51	713	7.07	141	9.23
2006	N/A	N/A	2238	13.55	69	7.84	1436	10.65	904	8.97	130	8.51
2007	44	5.28	2283	13.82	65	7.39	1534	11.38	946	9.39	134	8.78
2008	112	13.45	2240	13.56	98	11.14	1308	9.70	866	8.59	163	10.67
2009	130	15.61	2148	13.01	112	12.73	1389	10.3	852	8.45	164	10.74
2010	100	12.00	701	4.24	75	8.52	920	6.82	1033	10.25	147	9.63
2011	105	12.61	N/A	N/A	74	8.41	1188	8.81	917	9.10	102	6.68
2012	98	11.76	N/A	N/A	98	11.14	279	2.07	922	9.15	97	6.35
2013	40	4.80	N/A	N/A	61	6.93	1107	8.21	839	8.33	83	5.44
2014	107	12.85	1756	10.63	59	6.70	1201	8.91	846	8.39	140	9.17
2015	62	7.44	1833	11.10	62	7.05	839	6.22	712	7.06	121	7.92
2016	35	4.20	1187	7.19	31	3.52	594	4.41	528	5.24	105	6.88
SEASON												
Fall	197	23.65	3815	23.1	214	24.32	3461	25.67	2442	24.23	378	24.75
Spring	216	25.93	4782	28.96	253	28.75	3670	27.22	2804	27.82	407	26.65
Summer	171	20.53	3210	19.44	155	17.61	2420	17.95	2040	20.24	309	20.24
Winter	249	29.89	4708	28.51	258	29.32	3931	29.16	2792	27.70	433	28.36
length of stay (days, M (STD))	4.04 (3.44)		3.70 (4.2)		4.21 (3.71)		4.03 (4.13)		4.32 (6.88)		3.63 (6.41)	

Table 1b
Characterization of the asthma patients requiring ED visits.

	Albany (N)	Albany (%)	Bronx (N)	Bronx (%)	Buffalo (N)	Buffalo (%)	Manhattan (N)	Manhattan (%)	Queens (N)	Queens (%)	Rochester (N)	Rochester (%)
N	4140	100	64,398	100	4030	100	51,901	100	29,675	100	5911	100
Male	1557	37.61	25,576	39.72	1477	36.65	21,042	40.54	11,754	39.61	2167	36.66
AGE (years; mean(STD))	38.38 (14.5)		40.92 (14.95)		39.36 (15.04)		41.47 (15.29)		40.67 (15.52)		38.74 (15.24)	
18-39	2361	57.03	30,519	47.39	2175	53.97	24,306	46.83	14,736	49.66	3259	55.13
40-49	862	20.82	15,062	23.39	876	21.74	11,711	22.56	6252	21.07	1317	22.28
50-69	594	14.35	11,525	17.9	576	14.29	9299	17.92	5181	17.46	768	12.99
60-69	198	4.78	5075	7.88	246	6.10	4344	8.37	2203	7.42	333	5.63
70-79	71	1.71	1690	2.62	104	2.58	1684	3.24	900	3.03	132	2.23
≥80	54	1.30	527	0.82	53	1.32	557	1.07	403	1.36	102	1.73
RACE/ETHNICITY												
White	1841	44.47	7469	11.60	1829	45.38	7954	15.33	7118	23.99	2221	37.57
Black	1739	42.00	28,356	44.03	1679	41.66	29,803	57.42	14,298	48.18	2525	42.72
American Indian	5	0.12	183	0.28	14	0.35	124	0.24	127	0.43	4	0.07
Asian	16	0.39	273	0.42	15	0.37	470	0.91	912	3.07	15	0.25
Native Hawaii	0	0.00	3	0.01	1	0.02	4	0.01	16	0.05	0	0.00
Hispanic	232	5.60	14,046	21.81	285	7.07	6887	13.27	4901	16.52	775	13.11
YEAR												
2005	N/A	N/A	7970	12.38	288	7.15	5458	10.52	1837	6.19	410	6.94
2006	N/A	N/A	8185	12.71	316	7.84	4284	8.25	2418	8.15	413	6.99
2007	204	4.93	7902	12.27	284	7.05	4953	9.54	2309	7.78	456	7.71
2008	543	13.12	7258	11.27	358	8.88	4199	8.09	1948	6.56	495	8.37
2009	425	10.27	6556	10.18	409	10.15	3863	7.44	2544	8.57	554	9.37
2010	461	11.14	2265	3.52	361	8.96	3047	5.87	2911	9.81	556	9.41
2011	531	12.83	N/A	N/A	314	7.79	4113	7.92	2724	9.18	392	6.63
2012	527	12.73	N/A	N/A	425	10.55	1094	2.11	3010	10.14	438	7.41
2013	312	7.54	N/A	N/A	290	7.20	4237	8.16	3053	10.29	497	8.41
2014	605	14.61	7703	11.96	306	7.59	5493	10.58	2405	8.10	627	10.61
2015	267	6.45	8411	13.06	352	8.73	5217	10.05	2252	7.59	498	8.42
2016	265	6.40	8148	12.65	327	8.11	5943	11.45	2264	7.63	575	9.73
SEASON												
Fall	1221	29.49	15,792	24.52	1122	27.84	14,540	28.01	8110	27.33	1670	28.25
Spring	1031	24.90	19,078	29.63	1043	25.88	14,426	27.80	8359	28.17	1430	24.19
Summer	831	20.07	13,131	20.39	890	22.08	9665	18.62	5733	19.32	1334	22.57
Winter	1057	25.53	16,397	25.46	975	24.19	13,270	25.57	7473	25.18	1477	24.99
length of stay (days, M (STD))	0.09 (0.31)		0.05 (0.28)		0.13 (0.37)		0.06 (0.35)		0.09 (0.30)		0.26 (0.54)	

Table 1c
Characterization of the COPD patients requiring hospitalizations.

	Albany (N)	Albany (%)	Bronx (N)	Bronx (%)	Buffalo (N)	Buffalo (%)	Manhattan (N)	Manhattan (%)	Queens(N)	Queens (%)	Rochester (N)	Rochester (%)
N	2085	100	7174	100	1863	100	9074	100	9685	100	2359	100
Male	851	40.82	3297	45.96	773	41.49	4294	47.32	4434	45.78	1009	42.77
AGE (years; mean (STD))	69.26 (11.84)		70.55 (12.4)		70.54 (11.88)		71.19 (12.5)		72.91 (12.3)		69.83 (12.27)	
18-39	11	0.53	41	0.57	6	0.32	61	0.67	56	0.58	12	0.51
40-49	73	3.50	332	4.63	74	3.97	386	4.25	294	3.04	115	4.87
50-69	376	18.03	1055	14.71	279	14.98	1246	13.73	1131	11.68	374	15.85
60-69	594	28.49	1759	24.52	480	25.76	2121	23.37	2106	21.74	604	25.6
70-79	571	27.39	2099	29.26	544	29.2	2703	29.79	2830	29.22	690	29.25
≥80	460	22.06	1888	26.32	480	25.76	2557	28.18	3268	33.74	564	23.91
RACE/ETHNICITY												
White	1858	89.11	3017	42.05	1517	81.43	5212	57.44	6395	66.03	1924	81.56
Black	163	7.82	1984	27.66	287	15.41	2248	24.77	1972	20.36	348	14.75
American Indian	1	0.05	28	0.39	2	0.11	57	0.63	42	0.43	1	0.04
Asian	7	0.34	65	0.91	4	0.21	297	3.27	313	3.23	7	0.3
Native Hawaii	1	0.05	0	0.00	0	0.00	1	0.01	5	0.05	1	0.04
Hispanic	31	1.49	1006	14.02	21	1.13	863	9.51	634	6.55	58	2.46
YEAR												
2005	N/A	N/A	956	13.33	148	7.94	1216	13.4	668	6.9	247	10.47
2006	N/A	N/A	936	13.05	146	7.84	876	9.65	866	8.94	205	8.69
2007	112	5.37	934	13.02	145	7.78	1025	11.3	840	8.67	231	9.79
2008	319	15.3	1039	14.48	223	11.97	935	10.3	901	9.3	262	11.11
2009	253	12.13	1049	14.62	209	11.22	875	9.64	968	9.99	265	11.23
2010	282	13.53	381	5.31	181	9.72	637	7.02	1079	11.14	238	10.09
2011	295	14.15	N/A	N/A	180	9.66	825	9.09	978	10.1	168	7.12
2012	245	11.75	N/A	N/A	198	10.63	203	2.24	999	10.31	206	8.73
2013	148	7.1	N/A	N/A	151	8.11	839	9.25	931	9.61	192	8.14
2014	296	14.2	1027	14.32	129	6.92	935	10.3	870	8.98	178	7.55
2015	134	6.43	834	11.63	153	8.21	688	7.58	562	5.8	162	6.87
2016	1	0.05	18	0.25	0	0	20	0.22	23	0.24	5	0.21
SEASON												
Fall	496	23.79	1430	19.93	368	19.75	2211	24.37	2297	23.72	533	22.59
Spring	581	27.87	2201	30.68	538	28.88	2576	28.39	2631	27.17	632	26.79
Summer	402	19.28	1560	21.75	395	21.20	1676	18.47	2020	20.86	472	20.01
Winter	606	29.06	1983	27.64	562	30.17	2611	28.77	2737	28.26	722	30.61
length of stay (days, M (STD))	5.21 (8.84)		5.58 (7.88)		5.18 (6.43)		5.83 (6.62)		6.31 (6.49)		4.72 (5.5)	

Table 1d
Characterization of the COPD patients requiring ED visits.

	Albany (N)	Albany (%)	Bronx (N)	Bronx (%)	Buffalo (N)	Buffalo (%)	Manhattan (N)	Manhattan (%)	Queens (N)	Queens (%)	Rochester (N)	Rochester (%)
N	1124	100	2648	100	1162	100	2716	100	1845	100	1359	100
Male	451	40.12	1358	51.28	496	42.69	1469	54.09	919	49.81	635	46.73
AGE (years; mean (STD))	62.28 (13)		62.45 (12.76)		63.89 (12.81)		62.08 (12.81)		64.04 (13.99)		63.92 (12.74)	
18-39	29	2.58	81	3.06	26	2.24	99	3.65	65	3.52	26	1.91
40-49	144	12.81	275	10.39	142	12.22	318	11.71	186	10.08	144	10.6
50-69	339	30.16	811	30.63	269	23.15	769	28.31	486	26.34	353	25.97
60-69	267	23.75	697	26.32	310	26.68	761	28.02	442	23.96	396	29.14
70-79	213	18.95	517	19.52	278	23.92	519	19.11	379	20.54	248	18.25
≥80	132	11.74	267	10.08	137	11.79	250	9.2	287	15.56	192	14.13
RACE/ETHNICITY												
White	920	81.85	701	26.47	863	74.27	1153	42.45	1024	55.5	1042	76.67
Black	175	15.57	1175	44.37	245	21.08	1018	37.48	548	29.7	276	20.31
American Indian	0	0.00	8	0.30	4	0.34	3	0.11	11	0.60	0	0.00
Asian	1	0.09	16	0.6	1	0.09	45	1.66	38	2.06	4	0.29
Native Hawaii	0	0	0	0	0	0	0	0	2	0.11	0	0
Hispanic	15	1.33	281	10.61	17	1.46	226	8.32	159	8.62	36	2.65
YEAR												
2005	0	0	248	9.37	86	7.4	214	7.88	119	6.45	104	7.65
2006	0	0	304	11.48	72	6.2	162	5.96	116	6.29	97	7.14
2007	35	3.11	258	9.74	67	5.77	212	7.81	113	6.12	116	8.54
2008	133	11.83	293	11.06	101	8.69	211	7.77	119	6.45	114	8.39
2009	122	10.85	316	11.93	79	6.8	214	7.88	144	7.8	140	10.3
2010	97	8.63	98	3.7	112	9.64	144	5.3	224	12.14	115	8.46
2011	181	16.1	0	0	88	7.57	262	9.65	199	10.79	109	8.02
2012	150	13.35	0	0	134	11.53	63	2.32	220	11.92	126	9.27
2013	102	9.07	0	0	127	10.93	332	12.22	232	12.57	115	8.46
2014	211	18.77	534	20.17	129	11.1	502	18.48	191	10.35	193	14.2
2015	92	8.19	569	21.49	162	13.94	368	13.55	147	7.97	123	9.05
2016	1	0.09	28	1.06	5	0.43	32	1.18	21	1.14	7	0.52
SEASON												
Fall	280	24.91	569	21.49	282	24.27	693	25.52	474	25.69	327	24.06
Spring	287	25.53	774	29.23	305	26.25	719	26.47	465	25.2	369	27.15
Summer	250	22.24	694	26.21	266	22.89	626	23.05	413	22.38	333	24.5
Winter	307	27.31	611	23.07	309	26.59	678	24.96	493	26.72	330	24.28
length of stay (days, M (STD))	0.19 (0.53)		0.08 (0.32)		0.2 (0.5)		0.1 (0.36)		0.13 (0.39)		0.39 (0.73)	

2.3. Study Design and Statistical Analyses

To estimate the relative rates of asthma or COPD associated with short term increases in $PM_{2.5}$ source contributions (e.g. GAS or DIE), we used the same time-stratified, case-crossover design and conditional logistic regression analyses (Levy et al., 2001; Maclure, 1991) used in our analysis of source-specific $PM_{2.5}$ and acute cardiovascular hospitalizations (Rich et al., 2019). The case-crossover design compares pollutant concentrations immediately before the hospitalization or ED visit event (case periods) to other times matched to the case period by calendar month and weekday when the subject did not have an event (3–4 control periods per case). For example, if an event occurred on March 15, 2012, then the case-day would be March 15th, and the control days would be March 1st, 8th, 22nd, and 29th. Since the case and control are matched by subject, time-invariant factors such as age, gender, and long term health history are controlled by design. We then contrast the air pollutant concentrations on the case and control days. To do so, we separately fit conditional logistic regression models for asthma hospitalizations, asthma ED visits, COPD hospitalizations, and COPD ED visit for each $PM_{2.5}$ source described above. The case–control status (i.e., case = 1, control = 0) was regressed against the mean specific $PM_{2.5}$ source concentration and mean residual $PM_{2.5}$ ($rPM_{2.5}$) concentration (i.e. $rPM_{2.5}$ = $PM_{2.5}$ concentration – specific $PM_{2.5}$ source concentration). The use of the $rPM_{2.5}$ term was to account for possible confounding by the correlation between the source-specific $PM_{2.5}$ and total $PM_{2.5}$ concentrations as discussed by Mostofsky et al. (2012). The use of the $rPM_{2.5}$ substantially reduced the correlations between the source-specific $PM_{2.5}$ and the marker of total $PM_{2.5}$ as shown in Tables S2 and S3. This same approach was used in the prior studies of source-specific $PM_{2.5}$ and cardiovascular disease (Rich et al., 2019). In each analysis, an indicator variable was included for holidays. Temperature and relative humidity (same lag days as $PM_{2.5}$), time varying potential confounders, were modeled with a natural spline with 4 degrees of freedom. Akaike's Information Criterion (Aho et al., 2014) was used to select the df for each natural spline. This case-crossover model provided estimates of the rate ratio and its 95% confidence interval. The excess rate is the percent (%) increase in the rate per interquartile range increase in source-specific $PM_{2.5}$ concentration (i.e. [rate ratio – 1.0] * 100%). We then re-ran this set of models for each $PM_{2.5}$ source for lag days 0, 0–3, and 0–6 for each outcome (asthma hospitalizations, asthma ED visits, COPD hospitalizations, and COPD ED visits). Since there were 3 lagged effects estimated for each outcome/$PM_{2.5}$ source in separate models, a $p < 0.017$ (0.05/3) was used to define statistical significance. In further exploratory analyses, we then ran the model with an interaction term among periods [BEFORE (2005–2007), DURING (2008–2013), and AFTER (2014–2016)] and each individual source-specific $PM_{2.5}$ concentration (e.g. GAS * DURING, GAS * AFTER). The site-specific OR estimates were pooled into an overall value across the study area with a fixed-effect meta-analysis based on the generic inverse variance method (Viechtbauer, 2010). As a sensitivity analysis, we reran the meta-analysis with the mixed-effect model and found that the overall OR estimates were identical. All meta-analyses were done with the rma function in the metafor package ((https://cran.r-project.org/web/packages/metafor/metafor.pdf)). All data management and statistical analyses were done using R version 3.0.1 (https://www.r-project.org/). With the reduced number of subjects included in these analyses compared to the $PM_{2.5}$ analyses (Hopke et al., 2019), particularly given the loss of Albany and Bronx PM speciation data for several years each (Table S1), we could not examine any trends in the number of adverse health events for those 2 sites.

3. Results

The demographical characterization of the subjects in each of the 4 outcome groups (asthma hospitalizations, asthma ED visits, COPD hospitalizations, and COPD ED visit) are presented in Table 1(a–d). The numbers of subjects in this study were smaller than in Hopke et al. (2019) because of the decreased frequency of the ambient $PM_{2.5}$ species sampling (every 3rd or 6th day) compared to the $PM_{2.5}$ concentrations (every day) used in Hopke et al. (2019).

As discussed by Hopke et al. (2019), the majority of asthma patients only visiting the ED (discharged without being hospitalized) were predominantly female, while patients with asthma hospitalizations, COPD hospitalizations, and COPD ED visits were more often male. ED visit subjects were younger than those hospitalized patients, while asthma patients were younger than COPD patients. The majority of upstate (Albany, Buffalo, Rochester) requiring hospitalization for asthma or COPD were white (Table 1a and c). In NYC, the largest fractions of asthma and COPD hospitalized subjects were white (Table 1a and c). For asthma ED visits (Table 1b), less than half of the subjects were white, but ~50% were black at each site. For COPD ED visits (Table 1d), the majority of patients were white at each site, except for the Bronx and Manhattan. For the Bronx, 44% of the subjects were black and almost 11% were Hispanic while in Manhattan, whites were the plurality with about 37.5% black and 8.3% Hispanic.

The distributional characteristics of the source-specific $PM_{2.5}$ concentrations ($\mu g/m^3$) by site, period, and source, separated for the case and control days for the 4 outcomes are presented in Tables S4–S7. Since not all $PM_{2.5}$ sources were resolved at all sites, the 'N/A' entries pertain to periods for which no speciation samples were collected. The distributional characteristics and the results of Kruskal-Wallis ANOVA on ranks among the three periods are shown in Figs. S1–S9 for all of the sources except road salt, fresh sea salt and aged sea salt that represent same and constant contributors to the ambient $PM_{2.5}$. For most sources, concentrations of the resolved source contributions were least in the AFTER period. However, GAS increased in the AFTER period and DIE varied from site to site. Detailed trend analyses for each of the sources were presented in Masiol et al. (2019).

Table 2 presents the mean annual incidence rates per 1000 persons

Table 2
Mean annual incidence rate (#/1000 persons per year) of hospital admissions and ED visits by period for asthma and COPD.

	Period	Overall	Albany	Bronx	Buffalo	Manhattan	Queens	Rochester
Asthma hospitalizations	BEFORE	0.42	0.03	0.76	0.08	0.44	0.29	0.19
	DURING	0.26	0.18	0.29	0.10	0.29	0.30	0.17
	AFTER	0.28	0.12	0.52	0.06	0.24	0.23	0.17
COPD hospitalizations	BEFORE	0.28	0.07	0.32	0.16	0.29	0.27	0.31
	DURING	0.24	0.46	0.14	0.22	0.20	0.33	0.30
	AFTER	0.17	0.26	0.21	0.11	0.15	0.16	0.16
Asthma ED visits	BEFORE	1.37	0.12	2.75	0.33	1.39	0.74	0.59
	DURING	0.86	0.84	0.90	0.41	0.95	0.90	0.67
	AFTER	1.44	0.68	2.66	0.37	1.51	0.75	0.77
COPD ED visits	BEFORE	0.07	0.02	0.09	0.08	0.06	0.04	0.15
	DURING	0.07	0.24	0.04	0.12	0.06	0.06	0.16
	AFTER	0.09	0.18	0.12	0.11	0.08	0.04	0.15

Table 3
Excess rate (%) of hospitalization or ED visits for asthma and COPD across NYS for the 2005–2014 period.

Outcome	Lag (days)	IQR	N Cases	Excess Risk (%)	P value
Secondary Nitrate					
Asthma hospitalizations	0	1.36	175,852	0.1 (−0.6, 0.8)	0.795
	0–3	1.59	114,047	0.3 (−1.0, 1.6)	0.705
	0–6	1.60	123,240	0.3 (−1.3, 2.0)	0.699
COPD hospitalizations	0	1.38	127,282	−0.3 (−1.1, 0.6)	0.569
	0–3	1.59	76,137	−0.1 (−1.7, 1.5)	0.858
	0–6	1.55	85,869	0.1 (−1.8, 2.1)	0.894
Asthma ED visits	0	1.12	657,749	−0.2 (−0.5, 0.2)	0.319
	0–3	1.53	437,102	−1.1 (−1.8, −0.5)	0.001
	0–6	1.51	470,947	−1.2 (−2.1, −0.4)	0.005
COPD ED visits	0	1.20	43,219	0.8 (−0.6, 2.3)	0.249
	0–3	1.57	24,592	3.2 (0.0, 6.4)	0.049
	0–6	1.60	29,831	0.2 (−3.5, 4.1)	0.898
Secondary Sulfate					
Asthma hospitalizations	0	2.07	175,852	1.2 (0.4, 2.0)	0.003
	0–3	2.06	114,047	0.9 (−0.4, 2.2)	0.176
	0–6	1.74	123,240	2.1 (0.7, 3.5)	0.004
COPD hospitalizations	0	2.05	127,282	1.0 (0.0, 1.9)	0.041
	0–3	2.03	76,137	0.6 (−0.9, 2.2)	0.428
	0–6	1.66	85,869	1.7 (0.1, 3.3)	0.039
Asthma ED visits	0	2.08	657,749	0.1 (−0.3, 0.5)	0.659
	0–3	2.03	437,102	−1.0 (−1.7, −0.3)	0.005
	0–6	1.62	470,947	−1.5 (−2.2, −0.9)	< 0.001
COPD ED visits	0	2.06	43,219	−0.9 (−2.6, 0.7)	0.271
	0–3	1.89	24,592	−1.0 (−3.8, 1.7)	0.458
	0–6	1.46	29,831	−0.4 (−2.9, 2.2)	0.749
Spark-ignition					
Asthma hospitalizations	0	2.44	175,852	−1.0 (−3.2, 1.2)	0.360
	0–3	1.63	114,047	−3.7 (−6.1, −1.3)	0.003
	0–6	1.42	123,240	−2.4 (−4.9, 0.3)	0.079
COPD hospitalizations	0	2.35	127,282	1.5 (−1.1, 4.1)	0.250
	0–3	1.69	76,137	2.5 (−0.8, 5.9)	0.146
	0–6	1.34	85,869	4.2 (1.1, 7.5)	0.008
Asthma ED visits	0	2.55	657,749	−0.4 (−1.5, 0.8)	0.513
	0–3	1.67	437,102	−0.5 (−1.7, 0.8)	0.472
	0–6	1.34	470,947	−0.4 (−1.6, 0.8)	0.516
COPD ED visits	0	2.55	43,219	3.1 (−1.4, 7.7)	0.175
	0–3	1.67	24,592	−0.2 (−5.4, 5.2)	0.929
	0–6	1.33	29,831	0.8 (−3.9, 5.7)	0.745
Diesel					
Asthma hospitalizations	0	0.54	175,852	0.5 (−0.3, 1.4)	0.214
	0–3	0.81	114,047	−0.2 (−2.4, 2.1)	0.865
	0–6	0.72	123,240	−0.2 (−2.6, 2.3)	0.895
COPD hospitalizations	0	0.59	127,282	0.7 (−0.4, 1.7)	0.220
	0–3	0.65	76,137	−0.5 (−2.7, 1.6)	0.625
	0–6	0.73	85,869	0.4 (−2.4, 3.3)	0.784
Asthma ED visits	0	0.49	657,749	−0.4 (−0.8, 0.0)	0.058
	0–3	0.56	437,102	−0.9 (−1.7, −0.1)	0.027
	0–6	0.63	470,947	−1.4 (−2.5, −0.2)	0.022
COPD ED visits	0	0.50	43,219	−0.8 (−2.5, 1.0)	0.397
	0–3	0.51	24,592	1.1 (−2.3, 4.6)	0.526
	0–6	0.58	29,831	−3.0 (−7.0, 1.2)	0.156
Biomass Burning					
Asthma hospitalizations	0	0.58	175,852	0.2 (−1.3, 1.8)	0.786
	0–3	0.43	114,047	−0.2 (−2.2, 1.8)	0.809
	0–6	0.36	123,240	0.7 (−1.4, 2.8)	0.531
COPD hospitalizations	0	0.58	127,282	−0.5 (−2.1, 1.1)	0.508
	0–3	0.44	76,137	0.1 (−2.2, 2.3)	0.964
	0–6	0.38	85,869	2.1 (−0.2, 4.4)	0.077
Asthma ED visits	0	0.56	657,749	0.7 (−0.1, 1.6)	0.082
	0–3	0.42	437,102	0.5 (−0.6, 1.6)	0.419
	0–6	0.36	470,947	1.3 (0.1, 2.5)	0.031
COPD ED visits	0	0.56	43,219	2.5 (−0.2, 5.2)	0.067
	0–3	0.43	24,592	2.3 (−2.0, 6.8)	0.305
	0–6	0.41	29,831	0.8 (−3.3, 5.2)	0.704
Road Dust					
Asthma hospitalizations	0	0.31	175,852	0.7 (−0.2, 1.7)	0.118
	0–3	0.26	114,047	0.4 (−0.9, 1.7)	0.539
	0–6	0.25	123,240	1.4 (−0.2, 3.0)	0.087
COPD hospitalizations	0	0.31	127,282	−0.4 (−1.5, 0.8)	0.525
	0–3	0.25	76,137	−2.2 (−3.8, −0.6)	0.006
	0–6	0.26	85,869	−2.5 (−4.5, −0.5)	0.015

(*continued on next page*)

Table 3 (*continued*)

Outcome	Lag (days)	IQR	N Cases	Excess Risk (%)	P value
Asthma ED visits	0	0.30	657,749	1.1 (0.6, 1.5)	< 0.001
	0–3	0.22	437,102	1.1 (0.6, 1.7)	< 0.001
	0–6	0.24	470,947	2.2 (1.4, 2.9)	< 0.001
COPD ED visits	0	0.30	43,219	−0.6 (−2.6, 1.5)	0.600
	0–3	0.21	24,592	3.1 (0.5, 5.7)	0.017
	0–6	0.21	29,831	1.2 (−1.9, 4.4)	0.453
Pyrolytic Organic Carbon					
Asthma hospitalizations	0	1.38	114,149	−0.3 (−1.6, 1.0)	0.639
	0–3	1.01	71,074	−0.7 (−2.2, 0.9)	0.376
	0–6	0.86	77,885	−0.4 (−2.2, 1.3)	0.625
COPD hospitalizations	0	1.48	82,890	1.1 (−0.6, 2.8)	0.210
	0–3	0.98	48,191	0.7 (−1.1, 2.6)	0.446
	0–6	0.93	55,291	1.5 (−0.8, 3.9)	0.199
Asthma ED visits	0	1.41	458,685	−0.0 (−0.7, 0.6)	0.900
	0–3	0.87	296,284	0.4 (−0.3, 1.1)	0.269
	0–6	0.76	322,121	1.1 (0.2, 1.9)	0.011
COPD ED visits	0	1.46	32,319	1.3 (−1.6, 4.3)	0.377
	0–3	0.88	18,104	3.2 (0.2, 6.2)	0.034
	0–6	0.83	22,204	0.1 (−3.4, 3.7)	0.963
Residual Oil					
Asthma hospitalizations	0	0.92	163,484	−0.9 (−2.6, 0.8)	0.282
	0–3	0.82	109,438	−3.1 (−5.5, −0.5)	0.020
	0–6	0.81	115,477	0.3 (−2.8, 3.4)	0.875
COPD hospitalizations	0	0.86	103,762	−3.5 (−5.6, −1.4)	0.001
	0–3	0.78	67,954	−2.9 (−6.1, 0.3)	0.074
	0–6	0.76	71,260	−4.6 (−8.3, −0.9)	0.016
Asthma ED visits	0	0.84	604,317	0.1 (−0.8, 0.9)	0.905
	0–3	0.73	418,065	−0.7 (−1.9, 0.6)	0.282
	0–6	0.68	437,278	0.5 (−1.0, 1.9)	0.536
COPD ED visits	0	0.83	29,465	−2.9 (−6.7, 1.0)	0.140
	0–3	0.73	20,039	0.2 (−5.5, 6.1)	0.958
	0–6	0.72	21,070	0.2 (−6.4, 7.4)	0.943
Aged Sea Salt					
Asthma hospitalizations	0	0.69	163484.00	−0.4 (−1.7, 0.9)	0.540
	0–3	0.64	109438.00	0.2 (−1.7, 2.1)	0.846
	0–6	0.61	115477.00	−2.3 (−4.6, 0.1)	0.059
COPD hospitalizations	0	0.70	103762.00	−0.2 (−1.9, 1.5)	0.822
	0–3	0.64	67954.00	−0.5 (−2.9, 2.0)	0.708
	0–6	0.63	71260.00	−2.8 (−5.9, 0.3)	0.079
Asthma ED visits	0	0.68	604317.00	0.3 (−0.3, 1.0)	0.351
	0–3	0.61	418065.00	1.5 (0.6, 2.5)	0.001
	0–6	0.59	437278.00	1.4 (0.2, 2.6)	0.019
COPD ED visits	0	0.68	29465.00	−0.3 (−3.4, 2.9)	0.854
	0–3	0.61	20039.00	0.8 (−3.5, 5.3)	0.712
	0–6	0.55	21070.00	0.1 (−5.0, 5.5)	0.964
Fresh Sea Salt					
Asthma hospitalizations	0	0.11	163,484	−0.1 (−0.4, 0.1)	0.174
	0–3	0.17	109,438	−0.0 (−0.6, 0.5)	0.862
	0–6	0.22	115,477	−0.6 (−1.4, 0.3)	0.196
COPD hospitalizations	0	0.12	103,762	−0.2 (−0.5, 0.1)	0.251
	0–3	0.17	67,954	−0.7 (−1.5, −0.0)	0.047
	0–6	0.21	71,260	−0.6 (−1.6, 0.6)	0.325
Asthma ED visits	0	0.09	604,317	0.1 (−0.0, 0.2)	0.199
	0–3	0.15	418,065	0.2 (−0.1, 0.5)	0.116
	0–6	0.18	437,278	0.5 (0.2, 0.9)	0.005
COPD ED visits	0	0.1	29,465	0.0 (−0.4, 0.5)	0.857
	0–3	0.16	20,039	−0.2 (−1.5, 1.1)	0.762
	0–6	0.18	21,070	−1.0 (−2.8, 0.9)	0.318

per year for asthma and COPD hospitalizations and ED visits for all subjects and each site individually. Incidence rates for asthma hospitalizations and ED visits in NYC were generally higher than those for the upstate cities. The Bronx had higher annual incidence rates for asthma ED visits and hospitalizations compared to Queens or Manhattan. COPD incidence rates among the NYC sites were comparable. However, rates of COPD hospitalizations and COPD ED visits did not have a clear upstate versus NYC pattern.

Table 3 presents the estimated excess rates (%) of hospitalization or ED visits for asthma or COPD associated with each interquartile range increase in source-specific $PM_{2.5}$ concentration on the day of the event (lag day 0), the average of the values on the day of the event and the sample from the prior 3rd day (lag days 0–3), and the 2 or 3 samples from every 6th or 3rd day prior, respectively, and the event day (lag days 0–6). Overall, there were similar numbers of significantly increased (n = 9) and decreased rates (n = 8) of respiratory events (asthma and COPD hospitalizations and ED visits) associated with increased source-specific $PM_{2.5}$ concentrations on lag days 0, 0–3, and 0–6 (Table 3). For example, an interquartile range (IQR) increase in GAS $PM_{2.5}$ concentration was associated with both decreased rates of asthma hospitalizations (−3.7%, 95% CI = −6.1%, −1.3%) and increased rates of COPD hospitalizations (4.2%, 95% CI = 1.1%, 7.5%). Similarly, IQR increases in concentrations of road dust $PM_{2.5}$ were associated with increased rates of asthma ED visits (e.g. previous 0–6 days: 2.2%, 95% CI = 1.4%, 2.9%), and increased rates of COPD ED visits (e.g. previous 0–3 days: 3.1%; 95% CI = 0.5%, 5.7%), but also

decreased rates of COPD hospitalizations (e.g. previous 0–3 days: 2.2%, 95% CI = −3.8%, −0.6%). Similarly for secondary sulfate, there were both positive and negative excess rates. Decreased rates of asthma ED visits were associated with IQR increases in secondary nitrate $PM_{2.5}$ concentration in the previous 0–3 and 0–6 days (e.g. −1.2%, 95% CI = −2.1%, −0.4%), while increased rates of asthma ED visits were associated with IQR increases in pyrolytic organic carbon $PM_{2.5}$ concentrations in the previous 0–6 days (1.1%, 95% CI = 0.2%, 1.9%) and aged sea salt concentration in the previous 0–3 days (1.5%, 95% CI = 0.6%, 2.5%). Last, IQR increases in residual oil $PM_{2.5}$ concentration were associated with significantly decreased rates of COPD hospitalizations (e.g. previous 0–6 days: 4.6%, 95% CI = −8.3%, −0.9%) (Table 3).

Hopke et al. (2019) saw only consistent patterns of increased rates of asthma ED visits and COPD hospitalizations associated with increased $PM_{2.5}$ concentrations in the AFTER period. To assess the results with the smaller data set resulting from only having $PM_{2.5}$ data on every 3rd or 6th day, we repeated the total $PM_{2.5}$ analyses in the same manner as Hopke et al. (2019) but with only the $PM_{2.5}$ concentration measured on the same days as the speciation samples were collected. These results are presented in Table S8. The results are very consistent with those reported by Hopke et al. (2019) suggesting that the smaller data set did not affect the previously observed patterns.

Thus, each period's (BEFORE, DURING, and AFTER) source-specific PM analyses were used to help understand which PM source(s) may have triggered these respiratory events. The excess rates of asthma and COPD hospitalizations and ED visits associated with source-specific $PM_{2.5}$ concentrations in the BEFORE, DURING, and AFTER periods are shown in Figs. 1 and 2 and Table S9. In the BEFORE period, there were no clear patterns of increased or decreased rates of COPD and asthma hospitalizations and ED visits across $PM_{2.5}$ sources with similar numbers of statistically significant increases (n = 9) and decreases in excess rates (n = 11). In the DURING period, there were again no clear increased or decreased excess rate patterns with only n = 8 statistically significant results (n = 7 increased excess rates and n = 1 decreased excess rate).

However, in the AFTER period, increased rates of asthma hospitalizations and ED visits were associated with multiple $PM_{2.5}$ sources at multiple lag times. For example, increased rates of asthma hospitalizations were significantly associated with IQR increases in concentrations of secondary nitrate $PM_{2.5}$ (e.g. during lag days 0–3: 8.2%, 95% CI = 3.3%, 13.4%), secondary sulfate $PM_{2.5}$ (e.g. during lag days 0–6: 10.8%, 95% CI = 4.8%, 17.1%), road dust $PM_{2.5}$ (e.g. during lag days 0–3: 2.5%, 95% CI = 0.0% 5.1%), pyrolytic organic carbon $PM_{2.5}$ (e.g. lag day 0: 3.0%, 95% CI = 0.3%, 5.7%), and aged sea salt $PM_{2.5}$ (e.g. during lag days 0–6: 4.4%, 95% CI = 0.2%, 8.9%). Similarly, increased rates of asthma ED visits were associated with IQR increases in concentrations of secondary nitrate $PM_{2.5}$ (e.g. during lag days 0–6 3.0%, 95% CI = 0.2%, 5.8%), secondary sulfate $PM_{2.5}$ (e.g. lag day 0: 2.6%, 95% CI = 1.1%, 4.2%), road dust $PM_{2.5}$ (e.g. lag day 0: 1.1%, 95% CI = 0.4%, 1.9%), and aged sea salt $PM_{2.5}$ (e.g. during lag days 0–3: 2.8%, 95% CI = 1.4%, 4.3%).

The excess rate patterns for COPD hospitalization and COPD ED visits across $PM_{2.5}$ sources were less clear. In the AFTER period, increased rates of COPD hospitalizations were associated with increased concentrations of pyrolytic organic carbon $PM_{2.5}$ (e.g. during lag days 0–6 days: 8.1%, 95% CI = 3.6%, 12.7%), while increased rates of COPD ED visits were associated with increased concentrations of diesel $PM_{2.5}$ (e.g. during lag days 0–3 days: 9.7%, 95% CI = 0.1%, 20.2%). There were no clear patterns of associations between COPD hospitalizations and ED visits and other $PM_{2.5}$ sources (Table S9).

4. Discussion

There was a lack of clear patterns of asthma or COPD hospitalization and ED visits associated with increased source-specific $PM_{2.5}$ concentrations across the whole study period (2005–2016) (Table 3). The only strong association was for asthma ED visits with road dust (RD) for all 3 lag days. Other sources that had some strong positive associations for varying lag days included secondary sulfate (SS) with both COPD and asthma hospitalizations, gasoline vehicles (GAS) with COPD hospitalizations, biomass burning (BB), pyrolized organic carbon (OP), aged sea salt (AGS) and fresh sea salt (FSS) with asthma ED visits and secondary nitrate (SN), RD, and OP with COPD ED visits. Thus, our results do not support our hypothesis that increased PM concentrations from oxidant-related sources (e.g., GAS, DIE, and RO) would be the major contributors to increased rates of hospitalizations or ED visits for COPD or asthma. The sources that showed patterns of positive associations with asthma or COPD events (ED visits or hospitalizations) on lag days 0, 0–3, and 0–6 although with many imprecise values are seen in Figs. 1 and 2, respectively. For asthma (Fig. 1 and Table S9), consistent patterns of association were observed for SN and SS and hospitalizations, and for RD with ED visits over the 2005–2016 period. There were increased asthma hospitalization rates in the AFTER period for SN, SS, RD, RO, and AGS. There were increased rates of asthma ED visits for BB and AGS. For COPD (Fig. 2 and Table S9), there are associations of hospitalization for OP at all lag days and SN, GAS and BB for the lag days 0–6 and for DIE for lag days 0 and 0–3 during the AFTER period.

Previously, Bell et al. (2014) examined hospital admission rates for COPD (ICD codes 490–492) in Connecticut and Massachusetts for adults older than 65 years associated with source-specific $PM_{2.5}$ and some of its major constituent species over lags of 0, 1, or 2 days. Increased rates of respiratory diseases/events were associated with increased concentrations of $PM_{2.5}$ road dust, and sea salt, as well as crustal species aluminum, calcium, silicon, and titanium, sea salt elements (chlorine), residual oil combustion markers (nickel and vanadium), and black carbon (typically used as a marker of traffic). They found that individual chemical species were better indicators than the resolved source-specific PM. Krall et al. (2017) assessed the association between short term increases in source-specific $PM_{2.5}$ concentrations (lags of 0–2 days) and respiratory disease emergency department (ED) visits in 4 U.S. cities: Atlanta, Georgia; Birmingham, Alabama; St. Louis, Missouri; and Dallas, Texas. Although increased rates of respiratory disease ED visits were associated with increased concentrations of biomass burning $PM_{2.5}$, rates of respiratory ED visits associated with diesel and gasoline $PM_{2.5}$ were frequently imprecise and null. They found little evidence of associations with crustal $PM_{2.5}$ materials.

Previously, we reported increased rates of cardiovascular, respiratory infectious disease, and respiratory hospitalizations and ED visits associated with increased $PM_{2.5}$ concentrations in the previous 0–6 days (Zhang et al., 2018; Croft et al., 2019a; Hopke et al., 2019). Increases in source-specific $PM_{2.5}$ concentrations in the previous 0, 0–3, and 0–6 days, including traffic (GAS, DIE, RD) and non-traffic emissions (secondary nitrate) were associated with increased excess rates of cardiovascular hospitalizations or ED visits (Rich et al., 2019). These results focus largely on motor vehicle emissions as the major influencing factor in the increased particulate toxicity. Spark-ignition vehicle (GAS) contributions to $PM_{2.5}$ had increased during the 2005 to 2016 period while diesel (DIE) and road dust (RD) contributions had remained largely constant (Masiol et al., 2019). Thus, as secondary nitrate and sulfate concentrations declined, these traffic-related sources represented a larger fraction of the residual $PM_{2.5}$ mass concentrations. Secondary organic carbon (SOC) also increased (Zhang et al., 2018; Croft et al., 2019a) and was moderately well correlated with the GAS contributions (Rich et al., 2019). The r^2 values between SOC and GAS were 0.423, 0.548, 0.504, 0.303 0.533, 0.533 for Albany, the Bronx, Buffalo, Manhattan, Queens, and Rochester, respectively. Thus, for all sites except Albany and Manhattan, the majority of the SOC variance was related to GAS mass contributions. More detailed discussion of the correlations of the source specific component and SOC are discussed below. For the cardiovascular outcomes, residual oil (RO) and secondary nitrate (SN) also were associated with increased rates of

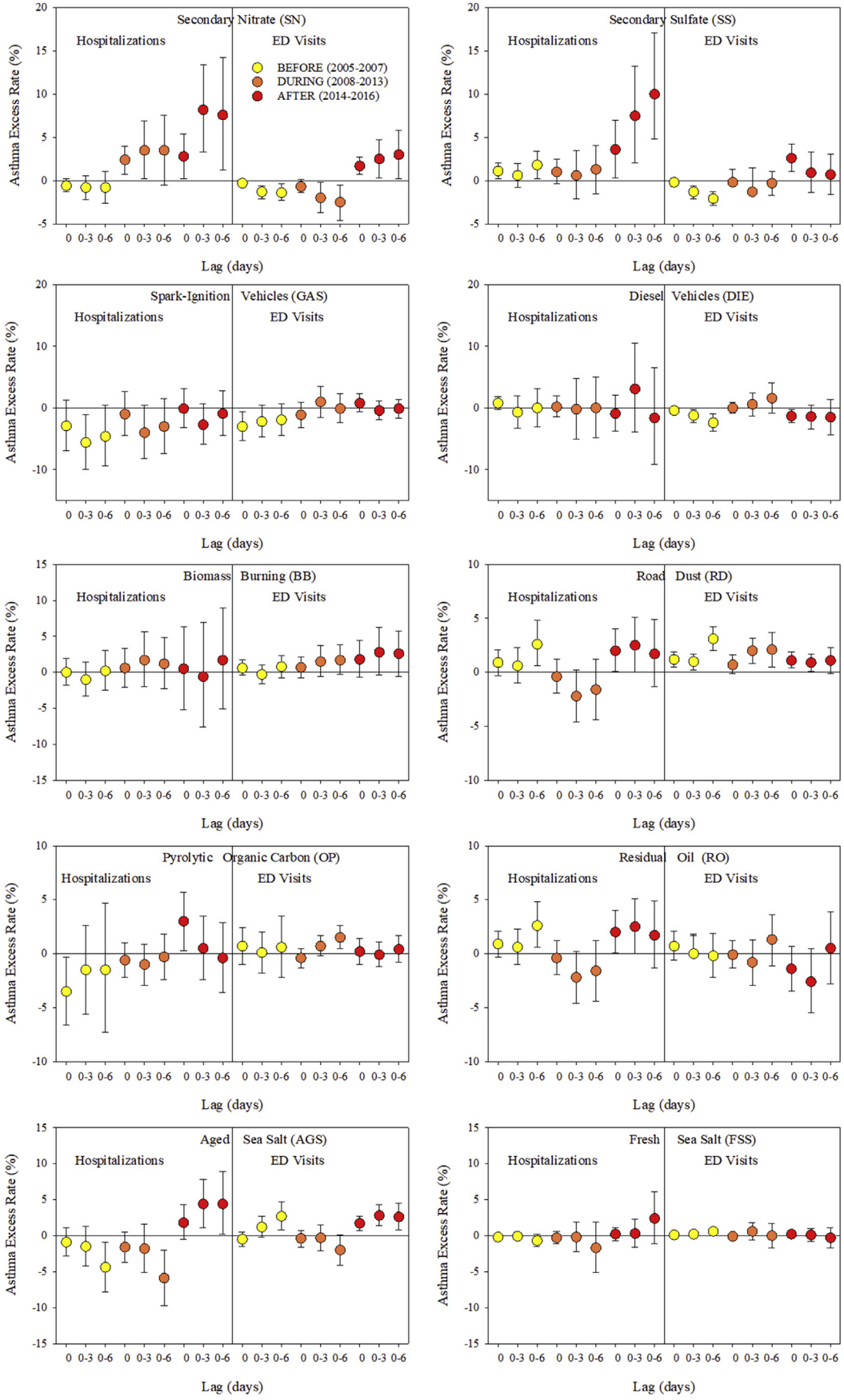

Fig. 1. Excess rates (%) of hospitalizations and ED visits for asthma by source and period.

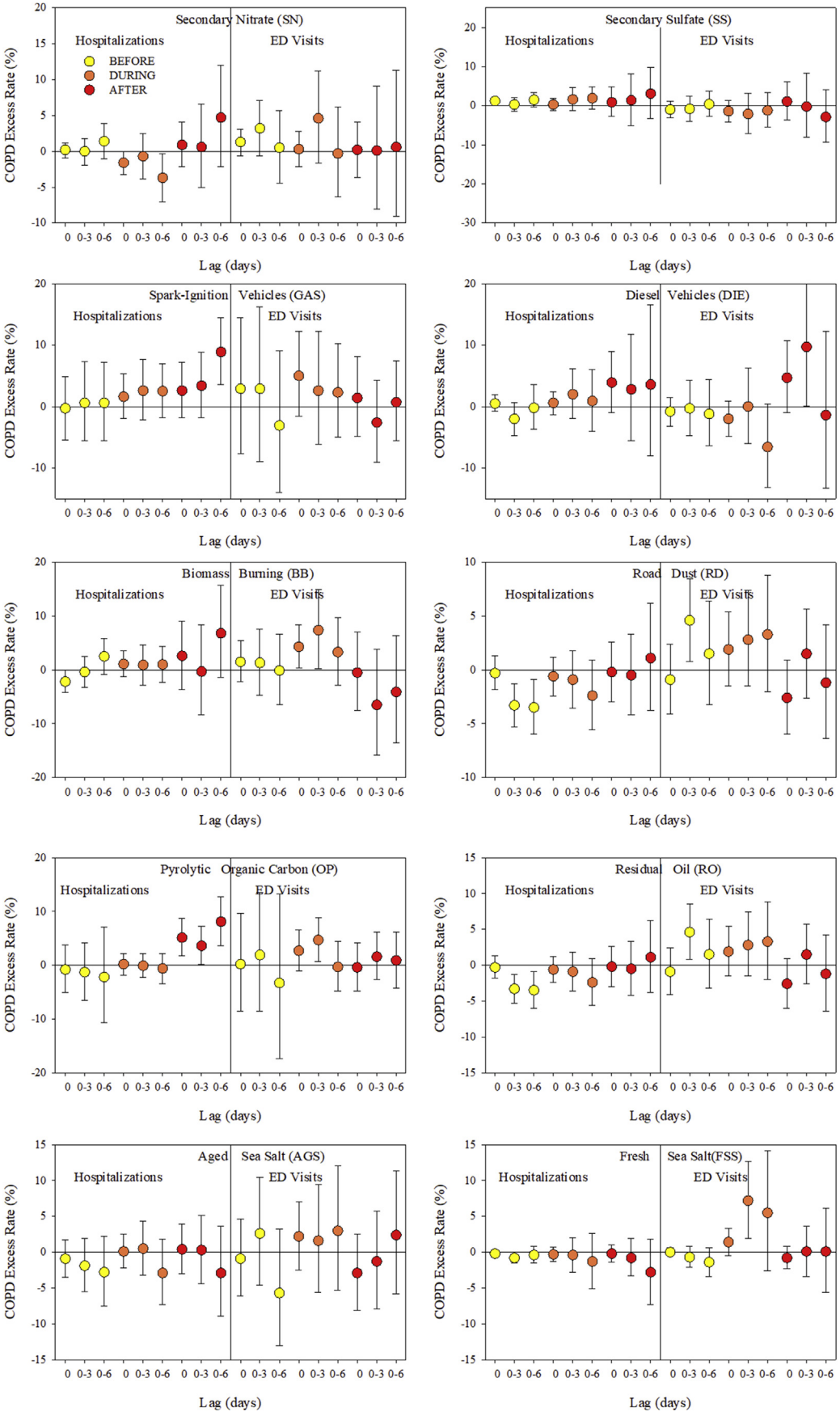

Fig. 2. Excess rates (%) of hospitalizations and ED visits for COPD by source and period.

hospitalizations. There were no strong patterns of temporal changes in the toxicity of the source-specific PM for the cardiovascular outcomes across the study period (Rich et al., 2019), suggesting that it was primarily the changes in relative concentrations that drove the increase per unit mass toxicity.

Croft et al. (2019b) reported the associations between respiratory infections (influenza and culture negative pneumonia) and source-specific $PM_{2.5}$ concentrations. For influenza, increased hospitalization rates were associated with interquartile range (IQR) increases in the mean $PM_{2.5}$ concentrations on lag days 0 and 0–3. Increases in SN concentrations on lag day 0 were associated with an increase in the influenza hospitalization rates. Increased but uncertain rates of influenza hospitalization were associated with increased concentrations of SS, GAS, DIE, RO, and BB on lag day 0. No consistent associations were observed between influenza hospitalizations and the OP, RD, FSS or AGS concentrations. The patterns of influenza ED visit rates were similar. Increased rates of influenza ED visits were also associated with increases in GAS concentration during lag days 0–3 and DIE concentration during several lag periods. Imprecise, increased rates of influenza ED visits were also associated with increases in SS, OP, RO, and BB during lag days 0–3. However, increased RD, RSS and AGS concentrations were not associated with increased influenza ED visit rates. No consistent patterns of effects were observed for increases in source specific $PM_{2.5}$ contributions and the rates of culture negative pneumonia hospitalizations or ED visits.

In the prior study, sharp increases in the rate of asthma ED visits and COPD hospitalizations associated with increased $PM_{2.5}$ concentrations in the previous 0–6 days (i.e. greater PM toxicity) were found in the AFTER period (Hopke et al., 2019). As noted above, increased asthma hospitalization rates were found in the AFTER period for SN, SS, RD, RO, and AGS and increased rates of asthma ED visits for BB and AGS. For COPD, there were increased hospitalizations associated with increased OP for all lag periods and with SN, GAS and BB for only lag days 0–6 and for DIE for lag days 0 and 0–3. Thus, these source types appear to drive the previously observed increased rates during the AFTER period.

To investigate the interrelationships among the sources and other PM species including primary organic carbon (POC), secondary organic carbon (SOC) as well as with temperature, relative humidity, and measured gaseous pollutants where available, pairwise correlation coefficients were calculated. Tables S10–S19 present the correlation coefficient matrices for the Queens and Rochester sites, respectively. These sites represent the relationships seen in NYC and in the upstate cities and are the two sites with the most complete data records. The correlation coefficients have been calculated for the entire study period (Tables S10 and S14) and for the BEFORE (Tables S11 and S15), DURING (Tables S12 and S16), and AFTER periods (Tables S13 and S17). These period-specific results could provide indications of changes in the interrelationships among these variables over the study subperiods. In these tables, correlations coefficients greater than 0.50 are shown in bold. Large negative correlations < -0.50 are shown in red and bold.

The formation of secondary organic carbon (SOC) results in the concurrent formation of oxidant species (Chen et al., 2011). Thus, SOC may provide a surrogate for exogenous oxidants (Hopke, 2015). Thus, the correlations between source specific $PM_{2.5}$ concentrations and SOC may provide an indication of sources that are associated with oxidants. The SOC concentrations estimated at the Queens site in the BEFORE period were strongly correlated with secondary sulfate ($r = 0.608$) and GAS ($r = 0.749$), while POC was strongly correlated with secondary nitrate ($r = 0.608$) and DIE ($r = 0.785$). POC was also strongly correlated with the gaseous species that would be associated with diesel exhaust including NO, NO_2, NOx, SO_2, and CO (Table S9). In the DURING period (Table S10), the SOC correlations with the secondary aerosol species (AN and AS) were lower, but SOC remained well correlated with GAS ($r = 0.766$). SOC was now also moderately correlated with biomass burning ($r = 0.535$) and pyrolyzed organic carbon ($r = 0.624$). The POC correlations with nitrate ($r = 0.529$) and DIE ($r = 0.483$) weakened. However, GAS was now about equally correlated with POC ($r = 0.512$), and remained correlated with the pollutant gases. In the AFTER period (Table S11), POC and SOC correlations with GAS and OP were even larger, but the POC and SOC correlations with biomass burning decreased. There was also a strong correlation between POC and SOC ($r = 0.597$) in the AFTER period. Similar correlation patterns across and within periods were also seen in Rochester (Tables S13–S15). However, the vehicular factors (i.e. GAS and DIE) were only associated with increased COPD hospitalizations and ED visits (Figs. 1 and 2). However, increased asthma rates were not observed suggesting that exposure to the hypothesized exogenous ROS was not a major driver of asthma exacerbation. However, RD and RO were associated with asthma hospitalizations in the AFTER period and RD was associated with ED visits across all 3 periods suggesting that redox active metals that can form endogenous oxidants may be important for inducing asthmatic effects. Road dust in $PM_{2.5}$ provided only a relatively small contribution to the PM mass because it represents only the lower end of the coarse particle size distribution that extends into the $PM_{2.5}$. Thus, the actual road dust exposures could be much larger since the bulk of the RD mass would be in particle sizes from 2.5 to 10 μm.

The types of source-specific $PM_{2.5}$ that were associated with increased rates of asthma hospitalizations and ED visits in the AFTER period were SN, SS, OP, RD, and AGS. The only source-specific $PM_{2.5}$ associated with increased COPD hospitalizations and ED visits in the AFTER period in addition to DIE, GAS, was OP that was significant for all 3 lag days. There are no obvious relationships among the source types identified in our study that affect asthma except that they are likely to represent larger sized particles within the $PM_{2.5}$ compared to the smaller traffic related species (Kodros et al., 2018). Asthma affects the bronchial/bronchiolar region of the respiratory tract and thus, could be affected by those particle sizes that can more effectively deposit in this region. It has been reported that coarse mode particles may play a larger role in the exacerbation of respiratory diseases (USEPA, 2018) and thus, sources producing larger particles might be more effective in inducing the observed asthma associations. Additionally, Feng et al. (2017) reported a strong correlation between oxidative potential (the ability to induce the formation of endogenous oxidants) and the surface area of the PM. Several of these identified source types (SN and SS) represent the bulk of the ambient $PM_{2.5}$ surface area (Zhang et al., 2005) and may represent sources of endogenous oxidants and the associations of these source-specific PM with asthma events. However, these results do not provide as clear a picture of the source-specific PM concentrations and the changes over time in the associations for asthma and COPD hospitalization and ED visit reported by Hopke et al. (2019).

A major limitation of using source-specific $PM_{2.5}$ concentrations is the substantial reduction in the number of subjects for which exposure data are available, and thus reduced statistical power, since samples were only collected every 3rd or 6th day for chemical speciation. However, inference was made primarily by examining the pattern of excess rates across outcomes, $PM_{2.5}$ sources, and lag times, and secondly by whether any individual results were statistically significant. Second, the PMF results for each site (Squizzato et al., 2018b) were obtained from a single analysis using all 12 years of data. The individual sources were identified and named based on common chemical compositions across these 12 years (Squizzato et al., 2018b). However, it is possible that if the source apportionment was done separately for each individual time period (e.g. 2005–2007, 2008–2013, and 2014–2016), there might be differences in the daily concentrations of individual source-specific mass concentrations (e.g. secondary sulfate) from those used in this analysis. Assuming that this exposure misclassification is non-differential with regard to time (i.e. not different for case and control periods), then it would likely result in a bias toward the null and an underestimate of effects. Third, additional exposure

misclassification may have arisen because all study subjects (those with a COPD or asthma hospitalization or ED visit) lived within 15 miles of a given $PM_{2.5}$ monitoring site, and were assigned the same source-specific $PM_{2.5}$ contribution for that specific day irrespective of the distance between their residence and the site. For the same reasons, such misclassification would result in biases toward the null and additional underestimation of effects. Fourth, we also had the problem of a change in the hospital admission diagnosis codes used in SPARCS starting October 1, 2015 when they shifted from ICD-9 to ICD-10 codes. Certain ICD-9 codes could be divided into multiple specific ICD-10 codes, resulting in possible misclassification or undercounting of cases. However, since the case-crossover design contrasts pollutant concentrations between case and control time periods within a short period (one month), the change of case classification should have less impact on excess rates since the both periods used similar case definitions. Fifth, as most COPD cases are limited to older adults, our study population included adults only, and children who have the highest rate of asthma were not included in this study. Finally, case-crossover designs analyzed with conditional logistic regression cannot fully adjust for possible overdispersion (Armstrong et al., 2014) and that could result in larger confidence intervals than we reported.

5. Conclusions

Although Hopke et al. (2019) observed large increases in the rates of ED visits for asthma and hospitalizations for COPD per unit mass of $PM_{2.5}$ across NYS in 2014–2016 compared to 2005–2007 and 2008–2013, there were not similar increases in associations of these outcomes with source-specific $PM_{2.5}$. In Rich et al. (2019), the decreases in secondary sulfate and secondary nitrate meant that traffic-related sources (spark-ignition vehicles, diesel, and road dust) and sources with redox metals like residual oil combustion became larger fractions of the decreased $PM_{2.5}$ concentrations. Spark-ignition vehicles had the strongest correlation with secondary organic carbon that was hypothesized to represent a source of short-lived reactive oxygen species that drove the association of these source-specific $PM_{2.5}$ concentrations and the observed rates of ED visits and hospitalizations for cardiovascular disease. The analyses of the associations of source-specific $PM_{2.5}$ concentrations with hospitalizations for COPD found that excess rates for GAS and DIE did increase in the AFTER period as hypothesized, but none of the values were statistically significant. The other source types showed inconsistent patterns of excess rates. For asthma ED visits, only biomass burning and road dust showed consistent positive associations with road dust having significant values for most lag times. Secondary nitrate also showed significant positive associations with asthma ED visits in the AFTER period compared to no associations in the prior periods. The asthma ED results did not support the hypothesized effects of the major traffic sources (GAS and DIE) as the major drivers of the observed increase in per unit mass toxicity during the AFTER period. These results suggest that the relationships of asthma and COPD exacerbations by source-specific $PM_{2.5}$ are not as well defined as those for some of the cardiovascular outcomes previously reported and further work will be needed to determine what has caused the apparent increased the per unit mass toxicity of $PM_{2.5}$ in New York State in the AFTER (2014–16) period.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2019.108912.

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