A CONCEPTUAL MODEL FOR AMBIENT FINE PARTICULATE MATTER OVER SOUTHEAST MICHIGAN: HIGH CONCENTRATION DAYS

Version 1.0 October 1, 2008

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EXECUTIVE SUMMARY

In 2006, the USEPA promulgated revisions to the National Ambient Air Quality Standard (NAAQS) for particulate matter (PM) which tightened the 24-hour (daily) $PM_{2.5}$ standard from 65 $\mu g/m^3$ to 35 $\mu g/m^3$. This revision profoundly increased the frequency of exceedance days observed in Southeast Michigan with fourteen of the fifteen compliance monitors violating the standard for the period 2004-2006. An understanding of the drivers for high $PM_{2.5}$ days is needed to support the development of effective controls strategies; this study seeks to provide insights into such drivers.

At most sites in Southeast Michigan, exceedances occur predominantly – and in some cases exclusively – on days when the entire network is exhibiting high PM. For thirteen of the fifteen monitors, at least 85% of exceedances from 1999 to 2006 occurred on days when the entire network was exhibiting high PM. Drivers for network-wide high PM are regional transport, poor atmospheric ventilation across the network, or both of these conditions. Two monitors in particular – Dearborn and Wyandotte – frequently had exceedances on days when the entire network was not exhibiting high PM with 38% of the Dearborn exceedances and 27% of the Wyandotte exceedances occurring on such days. On these days, these sites are significantly influenced by nearby emission sources. Dearborn continues to be influenced by nearby emission sources which drive exceedances (or near exceedances) on days when the network-wide base concentration is as much as 15 µg/m³ below the standard, although the frequency of such days appears to be decreasing. In contrast, at Wyandotte all of the days for which nearby emission sources were disproportionately contributing to exceedances (or near exceedances) of the standard were prior to 2004. At those sites not significantly impacted by plumes from nearby emission sources, high PM days are most often observed when the area is experiencing a multiday high PM episode.

Seasonal variations in surface winds can explain the seasonal differences in frequency of site-specific high PM days between sites in the greater Wayne County area. Sites such as Allen Park, which are relatively free of impacts from nearby sources, exhibit similar seasonal behavior with high PM days most frequent in the summer followed by the winter. The monthly distribution of high PM days is distorted for sites with significant nearby emission sources since there is a strong seasonality to the surface winds distributions. For example, Dearborn has as many high PM days in the winter as in the summer, with the winter season having a stronger contribution from nearby sources located to the southwest of the site.

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Network-wide high PM days have disproportionately high contributions from the major ions (sulfate, nitrate, and ammonium) compared to other days. Summertime events are dominated by sulfate and wintertime events are dominated by ammonium nitrate. However these so-called high nitrate days typically have ammonium sulfate concentrations that are at least 50% of the ammonium nitrate concentration. The role of sulfate on such days should not be overlooked when considering the impact of SO_2 -based regional control strategies. In general, network-wide PM base concentrations above ~20 $\mu g/m^3$ tend to be disproportionately influenced by major ion contributions and are indicative of a disproportionately high influence from regional sources. On network-wide high PM days the sulfate and nitrate concentrations are typically several times higher than the corresponding monthly median concentrations. In contrast, EC is on average only 1.3 times higher. This pattern suggests that network-wide high PM days are primarily influenced by regional transport and are to a lesser extent influenced by increased levels of urban-scale emissions arising from poor atmospheric ventilation. As previously stated, emission sources near the monitoring sites can still exert significant plume impacts regardless of the network-wide PM behavior driven by regional and urban-scale contributions.

Many network-wide high PM days coincide with high nitrate, and in some cases there is substantial variability in PM mass concentration across the network on these days. It is important to determine whether the variability is real or a measurement artifact, because these days can influence the site-specific design values and in general can explain many of the exceedingly high PM levels observed at certain sites that otherwise do not appear to be disproportionately impacted by nearby local emission source plumes on high PM days.

A conceptual model for fine PM over central Southeast Michigan includes four elements. The largest contributor to annual average PM is regionally transported material that is temporally varying but spatially homogeneous over the domain (indeed, this defines the spatial extent of the domain for the conceptual model which in this case excludes the distant sites such as Luna Pier and Port Huron). On some days, regionally transported PM contributions can be sufficiently high to cause exceedances of the daily standard when coupled with average contributions from urban- and finer-scale emission sources and, in relatively rare cases, the regionally transported PM alone can cause exceedances of the daily standard. Urban-scale emission sources (mobile, area, and point sources) create a spatial PM gradient across central Southeast Michigan. For winds from a fixed direction, these contributions create an "urban plume" with decreasing impacts with increasing distance from the zone of highest emissions activity. The spatial gradients can be clearly discerned when binning data from days with common wind patterns and become smeared, to some extent, when averaging over periods with varying wind patterns. At many sites (e.g., Linwood, East 7 Mile) a PM plume emanating from the Detroit commercial / industrial zone, which is nominally Dearborn and areas to its southeast towards the Detroit River, including Zug Island, can be distinguished from the aforementioned urban plume. At sites further downwind, such as New Haven, these two plumes have essentially merged into a single urban plume. Average impacts from these plumes depend on the monitoring site location relative to the Detroit population and commercial/industrial core zones and the frequency that winds from these zones transport the plumes to the monitoring site. Finally, certain monitors in the Detroit commercial/industrial zone are significantly impacted by emissions from nearby sources that ultimately contribute to the distant PM plume emanating from this zone. At

Dearborn and Wyandotte these sources are located southwest of the monitoring sites and historically have contributed to exceedances (or nears exceedances) of the daily standard. These sources also disproportionately influence the annual standard because they are oriented upwind of the sites for prevailing winds from the southwest.

The analyses presented in this report exploited the relatively high spatial and temporal $PM_{2.5}$ FRM measurement density across Southeast Michigan. Each site provided added value to the analysis. Future reductions in either spatial or temporal measurement density would likely affect the ability to update this analysis towards tracking the accountability of emission control programs.

In summary, emission controls that reduce regionally transported PM are needed to reduce the frequency of violations of the 24-hour $PM_{2.5}$ standard. Emission reductions from a few specific point sources, especially near Dearborn, are also needed. While regional and point source controls will decrease annual average $PM_{2.5}$ concentrations for standardized synoptic weather patterns, relatively small but significant decreases could also be realized by controls on other urban emission sources such as mobile, area, and other point sources.

1. MOTIVATION

In 2006, the USEPA promulgated revisions to the National Ambient Air Quality Standard (NAAQS) for particulate matter (PM) which tightened the 24-hour (daily) $PM_{2.5}$ standard from 65 $\mu g/m^3$ to 35 $\mu g/m^3$. This revision profoundly increased the frequency of exceedance days observed in Southeast Michigan. For fifteen monitors over the time period 1999-2006¹, there were 7 and 179 exceedance days (days with one-or-monitors in the area with concentration greater than or equal to 35 $\mu g/m^3$) under the old and new standards, respectively. While none of the monitors violated the old standard, for the period 2004-2006 fourteen of the fifteen monitors violated the new standard. Only one of the eleven sites elsewhere in the state – Grand Rapids – violated the standard for the 2004-2006. This behavior motivates the need for a refined understanding of the drivers for high concentration days in Southeast Michigan. In particular, a detailed description is needed of the emissions, meteorology and atmospheric processes that modulate PM concentrations. This "conceptual model", with explicit emphasis on understanding $PM_{2.5}$ high mass concentration days over the Southeast Michigan, could be utilized towards developing effective control strategies.

The conceptual model presented in this document is focused on the information needed to support the development and implementation of control strategies to meet the 24-hour PM_{2.5} NAAQS. It intentionally excludes a detailed treatment of annual average concentration patterns and instead focuses on the subset of days exhibiting high concentrations. **Figure 1-1** shows the elements that go into the development of a conceptual model which is necessarily dynamic and must be refined as new knowledge becomes available. Content and presentation of the conceptual model for PM_{2.5} over Southeast Michigan loosely follows the framework used by Vickery (2004) for nine North American geographic regions and as revisited by Allen and Turner (2008). A conceptual model for PM_{2.5} over St. Louis was recently developed which included the following elements: annual and seasonal levels of PM_{2.5} in relation to the mass-based standards; compositional analysis of PM_{2.5}; meteorological influences; atmospheric processes contributing to PM_{2.5}; sources and source regions contributing to the principle chemicals of concern; and implications to policy makers (Turner and Garlock, 2007). For this study, the framework has been modified to accommodate the emphasis on those days with high PM concentrations.

An assessment of the drivers for high concentration days, compared to annual average behavior, is complicated by several factors. First, high concentration days are relatively infrequent and thus there is limited data to probe. Second, while the standard defines a bright line for exceedance days, the use of threshold concentration values to censor data can mask some of the key features. The sensitivity of key findings to the operational definition of a high concentration day is important and care has been taken in this study to ensure the results are resistant to the selection of the threshold concentration.

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¹ Some of these monitors did not operate for the entire eight year period, and the monitors collect samples with frequency from every day to every sixth day.

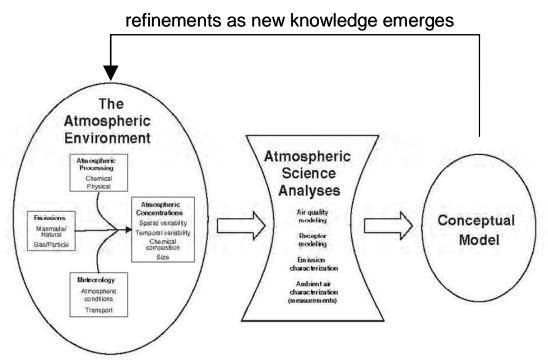


Figure 1-1. Development of a conceptual model (adapted from Vickery, 2004).

This conceptual model is grounded in an analysis of eight years of $PM_{2.5}$ FRM mass concentration data and allied data for particle speciation and meteorology. Nonetheless, it should be treated as a living document, to be periodically updated as additional data becomes available (in particular to capture temporal trends) and revised as more-detailed analysis is performed on the existing data.

2. APPROACH

Extensive monitoring and data analysis has been performed for Southeast Michigan. From a regulatory context, most of the data analysis effort has focused on the annual-average NAAQS. This work focuses on the drivers for exceedances of the daily (24-hour) NAAQS. This metric presents certain challenges to the characterization effort. In contrast to the annual average NAAQS, which requires an understanding of the average contribution of sources to observed burdens, consideration of the daily standard must necessarily focus on a much smaller number of days and with interest in the drivers of the concentrations specifically on those days. Many of the data analysis tools, such as receptor modeling by positive matrix factorization (PMF), are most powerful when considering the average behavior; estimates for high concentration days are less reliable and indeed in many analyses extreme values are down-weighted to improve the stability of the solution. Air quality surveillance is relatively sparse with sustained daily FRM measurements at only one site (Allen Park); this provides a challenge to placing an observed exceedance day in a temporal context. However, the Southeast Michigan FRM network is relatively dense, and this greatly facilitated the ability to the analyses presented in this report. Semicontinuous PM mass and composition measurements certainly provide considerable insight and in the future their deployment should be very beneficial in refining our knowledge of high concentration events.

A significant body of work has been assembled to characterize PM in Southeast Michigan with most-recent emphasis on the annual average PM_{2.5} NAAQS. Indeed, efforts currently underway to address the annual average standard are expected to improve the area's status with regard to the daily standard. However, additional measures might be needed to eliminate violations and bring the area into attainment with the daily standard. Thus, the key findings from this analysis are compared and contrasted with the Weight-of-Evidence (WOE) developed for the annual standard to more fully understand the synergies between the drivers and control strategies.

Many questions can be raised towards characterizing an observed exceedance of the daily standard. Was it driven by local emissions, regional transport, or both? Did it arise from an exceptional event, such as a structural fire or unusual industrial emission, or was it the conventional mix of emission sources with their contributions amplified by poor atmospheric ventilation conditions such as a persistent stagnation? Exceptional events will be experienced that cannot be explained with the typical types and levels of surveillance. For example, documented exceptional events at the St. Louis - Midwest Supersite in East St. Louis, IL, included nearby structural fires that in some cases were captured by the images from an on-site camera and in other cases were visually observed by site operators. The ability to identify exceptional events in the East St. Louis time series decreased as the site operations became routine and staff spent less time at the site. It is possible the observed high concentrations are more susceptible to measurement error (indeed, in some cases even result solely from measurement error) and uncontrollable measurement bias. We will examine this issue in the context of spatial variability in FRM values on high nitrate days. The Regional Haze Rule construct of isolating the 20% worst days – whether visibility or mass concentration – is closer in spirit to the information needed for examining drivers for high concentration days. However, in its most-commonly applied form it is inadequate because the average behavior on the 20% worst days might not represent the condition on an actual day if there are a variety of drivers for high

concentration days. The separation of contributions emanating from different spatial scales (e.g., regional-, urban-, and even finer local-scales) is particularly challenging and the data analysis approaches adopted for this study have been customized to elucidate the contributions from these scales to the extent practicable. In consideration of the above issues, it is apparent that analyses to capture the drivers for high concentration days must focus on <u>distributions</u> of behavior rather than average (central tendency) behavior. The data analysis methods reflect this consideration.

There are many possible drivers for intraurban variability in PM_{2.5} mass, including the following six factors: local sources of primary PM emissions; transient emission events; topographic barriers that isolate subregions of the urban area; meteorological phenomena that vary on spatial scales within the urban area; differences in behavior of semi-volatile components; and measurement error (Pinto *et al.* 2004; Turner and Allen, 2008). While this analysis does not definitively identify which of these factors are responsible for observed differences across the network, it does reflect upon these factors to explain the observed differences.

Figure 2-1 shows a conceptualization of intraurban variability arising from PM emission sources acting on various spatial scales. The air mass is transecting the domain with persistent surface winds from the left. Panel (a) shows the emission sources where the regional-scale contributions are assumed to be homogeneous across the domain (this assumption neglects interactions between regionally transported material and urban area emissions, such as urban plume processing of regionally-transported PM precursor gases). Urban-scale emissions from mobile sources, area sources, and point sources distributed across the metro core contribute to an increase in PM mass concentration (dashed line) which is skewed towards the downwind end of the domain. Neighborhood- and finer-scale emissions create local hot spots which disperse downwind and become part of the urban-scale contributions. The red line in panel (b) shows the concentration profile that would be observed along the transect. Monitor locations will dictate which features of the profile are captured. For example, panel (c) shows the location of five monitors (the "network-wide baseline" will be discussed later in this report). Monitor A is located upwind of the urban area and measures the regional contributions. However, a wind reversal would render it downwind of the urban area and in this case it might be impacted by the urban plume (such as monitor **D**). Monitor **B** is impacted by both a local plume and upwind urban contributions. The plume would miss the site with a wind reversal. Monitor C is located near the core of the urban area (from an urban-scale emissions perspective). For this scenario it is not impacted by local plumes but measures the cumulative contributions from upwind sources, including point sources in the urban area. A wind reversal, however, might render monitor C impacted by a local plume. Monitor **D** is outside the urban area emissions zone but nonetheless is impacted by the "urban plume" as it is advected downwind, while monitor E is far enough downwind that the urban plume has dispersed to the extent that the urban PM mass contributions cannot be distinguished from the underlying regional contributions. A wind reversal would render monitors **D** and **E** upwind of the urban area. This spatial concentration field represents a snapshot in time which becomes smeared for a 24-hour sample as winds shift during the day, and becomes further smeared when averaging results over many days. It is also a 1-D representation of a 2-D surface which adds complexity. That said, it does motivate the use of surface winds as a discriminator for studying site-specific PM mass trends to place each monitor in an emissions field context.

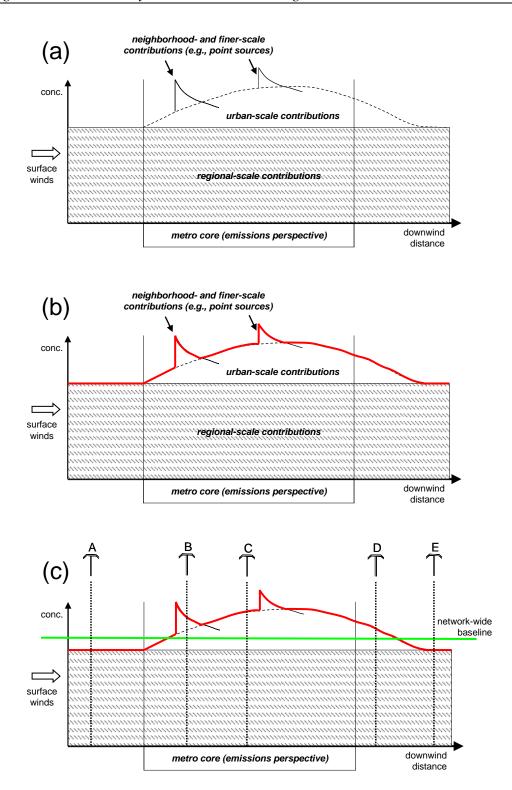


Figure 2-1. The influence of emissions acting on various spatial scales to observed concentrations at monitoring sites.

The report is structured as follows. Section 3 presents an overview of the monitoring network and key descriptive statistics and trends, including but not limited to the regulatory metrics. Section 4 examines the drivers for high concentration days by taking advantage of the relatively dense compliance monitoring network and particle speciation data. Section 5 refines the understanding of local emission source contributions by exploiting spatial and temporal variability within the network. Key findings, policy-relevant implications and recommendations for future work are summarized in Section 6.

3. SOUTHEAST MICHIGAN $PM_{2.5}$ FRM NETWORK AND CONCENTRATION TRENDS

The PM_{2.5} FRM monitoring network used in this analysis is summarized in **Figures 3-1** and **3-2** and **Table 3-1**. For the PM_{2.5} compliance monitoring period of 1999-2006, there were fifteen PM_{2.5} FRM monitoring sites in Southeast Michigan; three sites had collocated PM_{2.5} FRM samplers. All FRM monitoring sites in the Detroit-Warren-Flint CSA are included except Flint. For the purpose of this report, the spatial zone of Wayne County with highest population density (>10 persons/acre) shall be called the <u>Detroit population core</u> and the spatial zone which includes the Detroit central business district and industrialized zones to the south through west (e.g., Dearborn, Zug Island) shall be called the <u>Detroit commercial/industrial core</u>. This latter zone roughly includes the area covered by the five tightly clustered monitors in Figures 3-1 and 3-2 (Dearborn, Southwest High School, FIA/Lafayette, Newberry, and Linwood). <u>Greater Wayne County sites</u> shall refer to the monitoring sites located in Wayne County plus the Oak Park monitor site which is just across the county line in Oakland County.

Daily measurements have been maintained at Allen Park through the eight year period; most other sites operated on a 1-in-3 day schedule although some were cut back to 1-in-6 day in recent years due to budget constraints. There were 2,782 days with valid data reported for one-or-more sites (95.2% of days), with this high temporal data density driven by the daily sampling at Allen Park. There were 14,044 sampling events (neglecting collocated measurements). The historical compliance status of each site with respect to the 24-hour PM_{2.5} NAAQS is presented in **Table 3-2**. Exceedances of the 24-hour standard were observed on 179 days which is 6.1% of the days with one-or-more monitors reporting valid data. There were 546 total exceedances which correspond to 3.9% of all sampling events.

This report reflects on the conditions and drivers for high PM $_{2.5}$ mass concentration days (hereafter called "high PM days"). We are interested in both the frequency and intensity (magnitude) of high concentration days since both factors are important in establishing the design value. **Figure 3-3** shows the site-specific frequencies of high concentration days using thresholds of 25, 30, and 35 μ g/m³. Exceedances were observed on 2-8% of sample days with 5-14%, and 8-24% of sample days having concentrations above 30 μ g/m³ and 25 μ g/m³, respectively. Dearborn exhibits the highest frequency of high concentration days, followed by SW High School and Wyandotte. The remaining monitors in Greater Wayne County exhibit similar behavior. The frequencies for FIA/Lafayette and Newberry sites might be impacted by the relatively short sampling histories at these sites and caution should be used in comparing and contrasting these sites to the rest of the network. Ann Arbor and New Haven exhibit the lowest frequencies of high concentration days; they are 50-60 km from the Detroit commercial / industrial core to the west and northeast, respectively. However, these two sites do not represent the spatial extents of the SE Michigan network with Luna Pier ~70 km to the southwest and Port Huron ~80 km to the northeast.

Figure 3-4 shows the frequency of PM_{2.5} high concentration days by month for Allen Park, Dearborn, Southwest High School and New Haven (frequency plots for all sites are presented in **Figure A-1, Appendix A**). The pattern for Allen Park is generally representative of those Greater Wayne county sites with relatively modest impacts from nearby sources, as will be

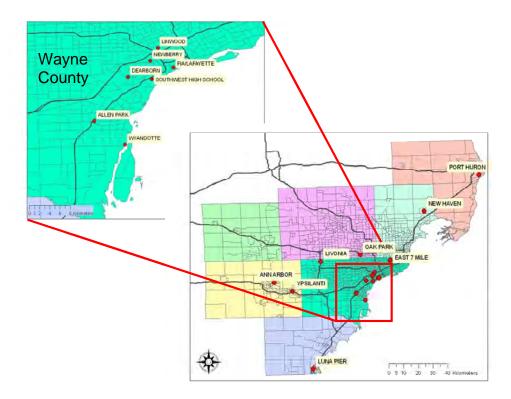


Figure 3-1. Southeast Michigan PM_{2.5} FRM monitoring sites superposed on county boundaries and 2000 census tracts. The red rectangle includes a portion of Wayne County.

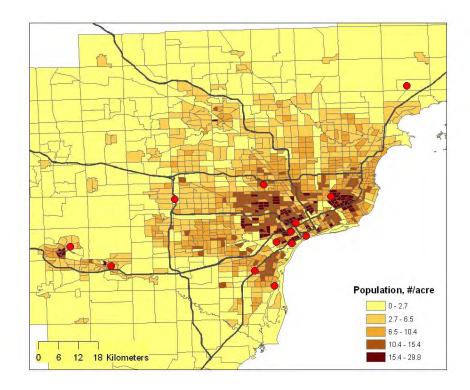


Figure 3-2. Southeast Michigan PM_{2.5} FRM monitoring sites (excluding Luna Pier and Port Huron) superposed on 2000 census tract level population density data.

Table 3-1. Southeast Michigan PM_{2.5} monitoring network as of December 2006, including exceedances days for the NAAOS daily PM_{2.5} standard of 35 µg/m³.

(A)	(B)	(C)	(D)	(E)	(F)	(G)
			Maximum		Number of	
		Sampling	Sampling	Number of	Exceedance	Speciation Site
Site Name	AIRS ID	Dates	Frequency	Sampling Days	Days	(Dates)?
Allen Park	261630001	05/99	daily	2497	92	12/00-
Ann Arbor	261610005	06/99-12/05	1-in-3 day	714	13	no
Dearborn	261630033	02/99	1-in-3 day	879	67	05/02
E 7 Mile	261630019	04/00	1-in-3 day	724	29	12/00-04/02
FIA/Lafayette	261630039	08/05	1-in-3 day	155	1	no
Linwood	261630016	05/99	daily	2273	102	no
Livonia	261630025	08/99	1-in-3 day	777	28	no
Luna Pier	261150005	12/99	1-in-3 day	804	28	05/02
New Haven	260990009	01/99	1-in-3 day	878	22	no
Newberry	261630038	12/04	1-in-3 day	161	6	no
Oak Park	261250001	01/99	1-in-3 day	793	32	no
Port Huron	261470005	01/99	1-in-3 day	841	32	no
SW High Sch.	261630015	02/99	1-in-3 day	881	41	no
Wyandotte	261630036	02/99	1-in-3 day	838	35	no
Ypsilanti	261610008	08/99	1-in-3 day	829	18	06/03

discussed later in this report. High concentration days are most frequent in the summer months followed by the winter months, while high concentration days are relatively infrequent in the spring. Dearborn exhibits higher frequencies of high concentration days for all months with the winter frequencies comparable to the summer frequencies. Southwest High School monthly frequencies fall between Allen Park and Dearborn, while New Haven has relatively few high concentration days but with the same overall pattern as Allen Park – highest frequencies in the summer followed by the winter.

Table 3-2. Southeast Michigan $PM_{2.5}$ monitoring network compliance status for the daily $PM_{2.5}$ NAAQS (POC = 1 samplers only).

		Annual 98th Percentiles					Th	ree-Year	Average:	s of 98th	Percentil	es			
Site	AIRS ID	1999	2000	2001	2002	2003	2004	2005	2006	99-01	00-02	01-03	02-04	03-05	04-06
Allen Park	261630001	43.7	38.6	44.2	39.6	40.5	36.9	43.0	34.1	42	41	41	39	40	38
Ann Arbor	261610005	38.2	33.1	38.5	31.3	33.3	28.4	39.1		37	34	34	31	34	34
Dearborn	261630033	45.1	45.1	43.2	45.7	42.8	39.4	50.2	43.1	44	45	44	43	44	44
E 7 Mile	261630019		42.0	42.0	34.4	37.1	35.0	52.3	36.2	42	39	38	36	41	41
FIA/Lafayette	261630039							43.9	32.4						38
Linwood	261630016	44.5	40.3	40.9	42.7	46.2	38.3	51.8	36.9	42	41	43	42	45	42
Livonia	261630025	38.4	35.9	44.7	32.7	38.1	32.2	40.2	30.4	40	38	39	34	37	34
Luna Pier	261150005	18.1	37.2	39.2	42.7	34.7	35.0	49.3	32.6	32	40	39	37	40	39
New Haven	260990009	31.9	33.2	42.0	35.6	31.8	31.9	41.5	34.4	36	37	36	33	35	36
Newberry	261630038						36.8	57.5	28.6						41
Oak Park	261250001	42.8	40.7	39.4	38.4	36.6	32.5	52.2	33.0	41	40	38	36	40	39
Port Huron	261470005	44.5	33.1	40.5	35.3	37.2	32.2	47.6	37.9	39	36	38	35	39	39
SW High Sch.	261630015	50.2	44.5	42.9	38.2	33.6	36.0	49.7	36.2	46	42	38	36	40	41
Wyandotte	261630036	45.0	42.7	46.6	34.1	34.8	32.3	46.7	33.2	45	41	39	34	38	37
Ypsilanti	261610008	40.6	30.3	39.7	30.9	38.8	31.5	52.1	31.3	37	34	36	34	41	38

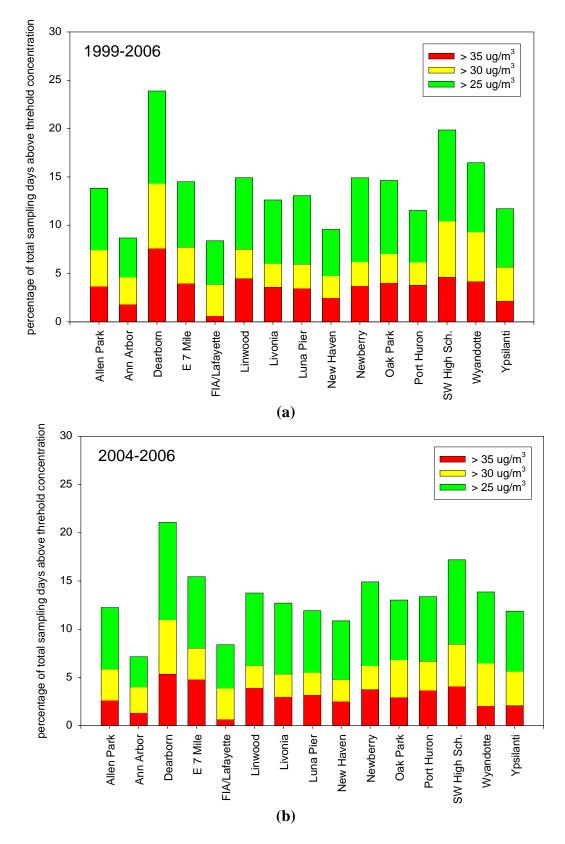


Figure 3-3. Site-specific frequency of total sampling days above the designated concentration for periods: 1999-2006 (a); and 2004-2006 (b),

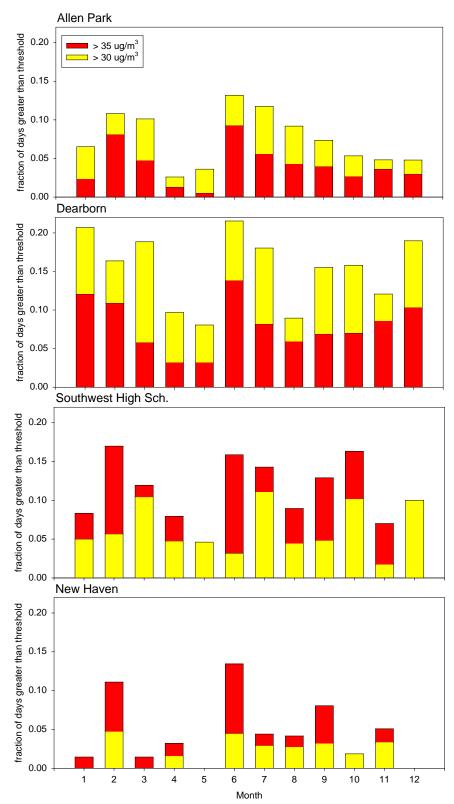


Figure 3-4. Distribution of PM_{2.5} high concentration days by month for Allen Park (top) and Dearborn (second from top), Southwest High School (second from bottom), and New Haven (bottom), 1999-2006.

Surface winds dictate the coupling between local emission sources and the monitoring sites. The wind rose for hourly values from Detroit Metropolitan Wayne County Airport is shown in **Figure 3-5** for the period 1999-2006. Prevailing surface winds are from the southwest with frequent winds also from the west. Winds from the east are relatively infrequent. Hourly surface winds distributions exhibit a distinct seasonality (**Figure 3-6**) albeit with some degree of year-to-year variation (**Figure B-1**, **Appendix B**). Fall and winter months are more advective with a pronounced southwesterly component while spring and summer months have lighter winds and less distinct prevailing features. Seasonal variations in surface winds can explain the seasonal differences in frequency of high PM days between the Greater Wayne County sites. For example, Dearborn has a disproportionately higher frequency of high PM days in the winter compared to Allen Park. This is consistent with the increased frequency of surface winds from the southwest during the winter and the location of nearby emission sources in this direction (and indeed located between Dearborn and Allen Park).

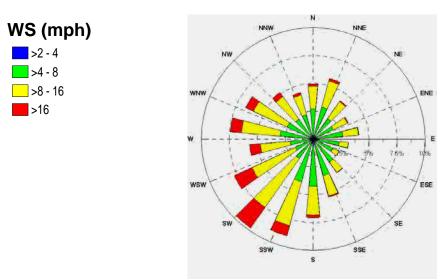


Figure 3-5. Wind rose for non-calm hours, observations at Detroit Metropolitan Wayne County Airport (DTW), 1999-2006. Calms (wind speeds less than 2 mph) were 11% of total hours.

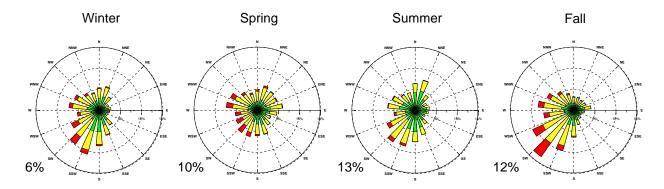


Figure 3-6. Seasonal wind roses for non-calm hours, observations at Detroit Metropolitan Wayne County Airport (DTW), 1999-2006. Calms (wind speeds less than 2 mph) are listed to the lower-left of each wind rose. The wind speed scales are shown in the legend for Figure 3-5.

4. CLIMATOLOGY OF NETWORK-WIDE HIGH PM DAYS

As previously described, a high fine PM mass concentration day at a given site can arise from regional contributions, local contributions, or a combination of these factors. The climatology of network-wide high PM days was examined to understand the drivers for such days. Regionally transported materials might be higher or atmospheric ventilation might be poorer. The extent to which these drivers act synergistically is not clear. For example, a summertime high pressure system advecting regionally transported PM might stall out over the area. However, the lower winds speeds might be partially offset by a large mixing layer depth on such days. While we cannot make definitive statements about such coupling, this section provides a perspective on these issues.

Descriptive analyses, including but not limited to those summarized in the previous section, demonstrate a high coherence in high PM days across the network. Thus, an exploratory analysis was performed to determine the frequency and nature of network-wide high PM days. A subset of sampling days with network-wide high PM mass concentrations was identified by first identifying those days with at least ten of the twelve Southeast Michigan FRM monitor sites reporting a valid PM_{2.5} FRM mass concentration value²; these days are called valid network days. Subsequently, the valid network days were screened to include only those days with at least 2/3 of the reporting monitors having a PM concentration greater than a threshold value (e.g., 30 μg/m³); these days are called <u>network-wide high PM days</u>. **Table 4-1** summarizes the characteristics of the valid network days and network-wide high PM days data sets. For the period 1999-2006 there were 727 sampling days with ten-or-more FRM monitor sites reporting valid PM_{2.5} concentrations (valid network days). 41 of these days (5.6%) had at least 2/3 of the monitors above 30 µg/m³ (network-wide high PM days) and all of these days had one-or-more monitor exceedance. Exceedances were also observed at one-or-more monitors on 35 days (5.1%) that were below the network-wide high PM threshold; these days likely have stronger influences from nearby emission sources impacting one or at most two sites. Figure 4-1 shows the distribution of network-wide high PM days by month. The days are concentrated in the summer followed by the winter, with few days in the seasonal transition months of April, May, and November. This pattern is insensitive to the concentration threshold used to define networkwide high PM days.

Table 4-1. Valid network days and network-wide high PM days by year.

Year	Valid Network Days	Network-Wide High PM Da		
1999	014	01	7%	
2000	084	04	5%	
2001	104	10	10%	
2002	101	04	4%	
2003	113	05	4%	
2004	117	05	4%	
2005	121	09	7%	
2006	073	03	4%	
total	727	41	6%	

² Ann Arbor, FIA/Lafayette and Newberry were excluded form this analysis due to their limited period of operation.

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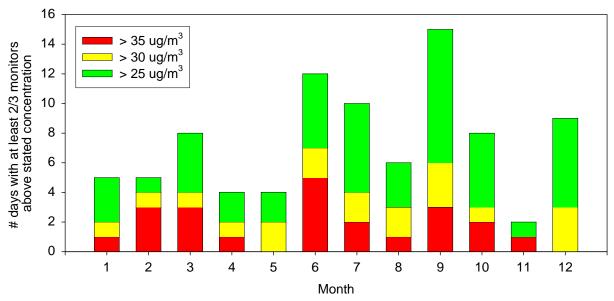


Figure 4-1. Frequency of network-wide high PM days by month using three different thresholds for defining a high PM day: ≥ 25 ; ≥ 30 ; and $\geq 35 \mu g/m^3$.

The relevance of this analysis to understanding drivers for exceedances of the 24-hour standard is demonstrated in **Table 4-2**. Valid network days, as defined above, capture 27-92% of all sampling days (column D). The capture is relatively low at sites with daily sampling (Allen Park, Linwood) because ten-or-more monitors must report valid data for the day to be designated a valid network day. However, the percentage of total exceedances that fall on a valid network day (column E) is nearly identical to the percentage capture of all sampling days by valid network days (column D) and we conclude that the subset of sampling days captured by valid network days is representative of all sampling days. Having established the representativeness of valid network days, we observed that at most sites exceedances occur predominantly – and in some cases exclusively – on days when the entire network is exhibiting high PM (column F). For example, 96% of the exceedances at Allen Park on valid network days occurred on days when the entire network was exhibiting high PM. Assuming the valid network days are representative of all sampling days at Allen Park, we would assign 88 of the 92 total observed exceedances to days with network-wide high PM. Similarly, 100% of the exceedances at Ypsilanti on valid network days occurred on days when the entire network was exhibiting high PM. Assuming the valid network days are representative of all sampling days at Ypsilanti, we would assign all of the 18 total observed exceedances to days with network-wide high PM.³ Dearborn represents the other extreme of behavior with only 62% of exceedances occurring on network-wide high PM days. In summary: (1) For nine of the fifteen monitors, more than 90% of the exceedances occurred on days when the entire network was exhibiting high PM; (2) Four monitors in the core region had a high frequency of exceedances on days when the entire network was not exhibiting high PM (Dearborn – 38%, Wyandotte – 27%, East 7 Mile – 15%, Linwood – 13%); and (3) At Luna Pier and Port Huron, which are located at the geographic extents of the Southeast Michigan

³ In contrast to Allen Park where only 25 of the 92 exceedances were captured by valid network days (27%), at Ypsilanti 16 of the 18 exceedances where captured by valid network days (89%) and the assertion that the valid network days are representative of all sampling days is not even necessary.

Table 4-2.	Site-specific	exceedance i	trends on	valid ne	twork days
I adic T-2.	DITC-SPECIFIC	CACCCGance	u chus on	vanu nc	twork days.

(A)	(A) (B)		(D)	(E)	(F)	
Site Name	Number of Sampling Days	Number of Exceedance Days	% Sampling Days on Valid Network Days	% Exceedances on Valid Network Days	% Network Day Exceedances on Network- Wide High PM Days	
Allen Park	2497	92	27.2%	27.2%	96.0%	
Ann Arbor	714	13	82.9%	92.3%	100.0%	
Dearborn	879	67	77.1%	77.6%	61.5%	
E 7 Mile	724	29	92.1%	89.7%	84.6%	
FIA/Lafayette	155	1	68.4%	100.0%	100.0%	
Linwood	2273	102	29.5%	29.4%	86.7%	
Livonia	777	28	87.1%	89.3%	92.0%	
Luna Pier	804	28	82.8%	85.7%	87.5%	
New Haven	878	22	80.4%	86.4%	100.0%	
Newberry	161	6	71.4%	100.0%	100.0%	
Oak Park	793	32	82.0%	75.0%	91.7%	
Port Huron	841	32	81.8%	81.3%	88.5%	
SW High Sch.	881	41	78.4%	73.2%	93.3%	
Wyandotte	838	35	80.2%	85.7%	73.3%	
Ypsilanti	829	18	82.8%	88.9%	100.0%	

region, 12-13% of the exceedances were on days when the entire network was not exhibiting high PM. It is possible the construct of network-wide high PM days is less robust for these sites since they might not always be bathed in the same regional air mass as the core region.

The threshold concentration used to define a network-wide high PM day does not itself capture the network-wide baseline PM level on that day (it is merely the bright line used to identify the days). Thus, for the purposes of this report a base concentration for each valid network day was defined as the 5th lowest concentration value across the network. Since at least ten monitors must have valid data for the day to be designated a valid network day, the 5th lowest concentration is no higher than the median concentration and ensures the base concentration is not significantly influenced by the monitors at the geographic extents (Luna Pier and Port Huron) which might be bathed in different air masses on certain days, and removes the influence of one-or-two anomalously low concentration values. The base concentration includes regionally transported PM and some portion of the urban-scale contributions. Excess mass at a given site, relative to the base concentration, can be negative for cases such as monitors **A** and **E** in Figure 2-1c.

Figure 4-2 shows the network-wide maximum and second maximum concentrations as a function of the base concentration for valid network days. The maximum concentration is often much greater than the base concentration and on these days there is a large local excess at the maximum concentration site. However, the second maximum concentration is typically within $5 \, \mu \text{g/m}^3$ of the base concentration and thus on a given day there is typically at most one site with a large local excess, regardless of the base concentration. This behavior holds at both high and

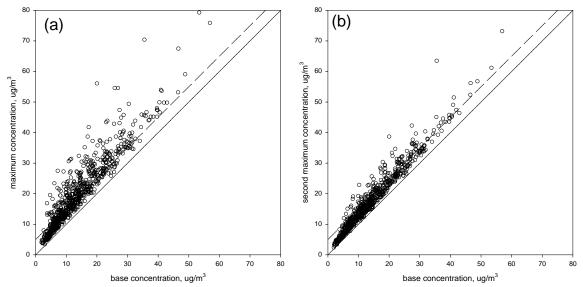


Figure 4-2. Network-wide maximum concentration (a) and second maximum concentration (b) for each valid network day compared to the respective network-wide base concentration. The analysis excludes three sites – Ann Arbor, Lafayette/FIA, and Newberry. The solid line is the base concentration (1:1 line) and the dashed line is $5 \mu g/m^3$ above the base concentration.

low base concentrations; the extreme maximum values possibly arise from nitrate hot spots or measurement artifacts as will be described later in this report.

Day- and site-specific concentrations are compared to the base concentrations in **Figure 4-3** for valid network days. Most sites had a few exceedances days with concentrations in large excess of the base concentration. Assuming poor atmospheric ventilation affects the sites similarly and thus contributes to the base concentration, the extreme excursions above the base concentrations likely arose from nearby emission source plumes, exceptional events (including but not limited to releases from the local emission sources) or measurement error including but not limited to a possible nitrate artifact. With the exception of Dearborn and Wyandotte, most exceedances were clustered about the base concentration. Dearborn and Wyandotte had exceedances on days with base concentrations as low as $20~\mu\text{g/m}^3$ demonstrating significant excess above the base concentration.

The analysis so far has primarily focused on the relative frequency of network-wide high PM days and their associations with exceedances. **Figure 4-4** captures the influence of the extreme values observed at each site (Figure 4-3), showing the 5th highest maximum excess and the median excess compared to the base concentration for exceedance days falling on valid network days. Several sites have low excess for the 5th highest maximum (yellow bars), although this metric is sensitive to sampling frequency. Most sites have median excess below 5 μ g/m³. Excluding Dearborn and Wyandotte, and pooling the site-specific highest three excess concentrations across the remaining ten sites, 23 of the 30 values occurred on only three days – 3/1/2003, 2/3/2005 and 2/6/2005. On all three days the network exhibited high and variable concentrations, and the latter two days were the highest nitrate at Allen Park over all valid network days (no speciation data for 3/1/2003). Further inspection of network-wide behavior on

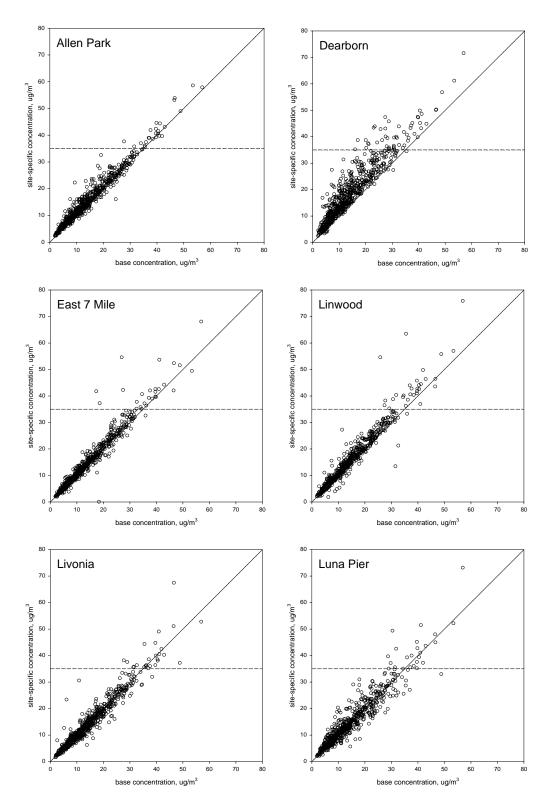


Figure 4-3. Site-specific concentrations versus base concentrations for valid network days. Solid line is the base concentration (1:1 line) and dashed line is $35 \mu g/m^3$.

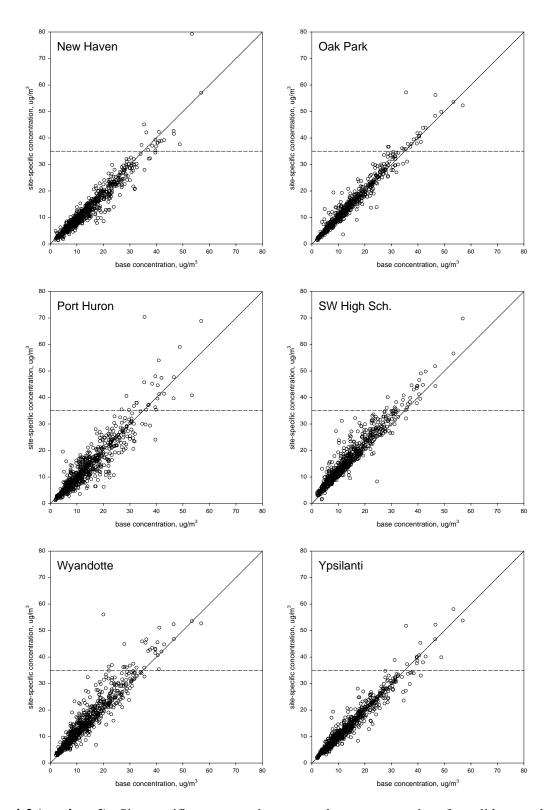


Figure 4-3 (continued). Site-specific concentrations versus base concentrations for valid network days. Solid line is the base concentration (1:1 line) and dashed line is 35 μ g/m³.

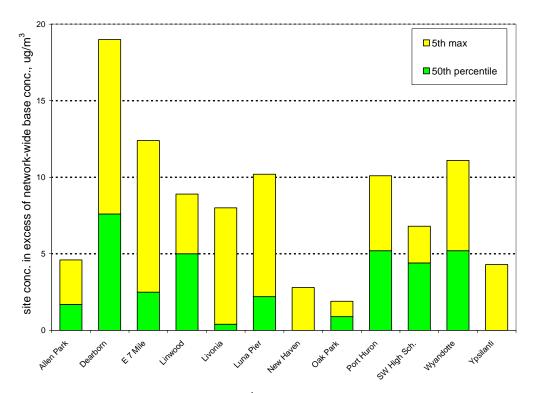


Figure 4-4. Site-specific exceedance days: 5th-highest excess concentration compared to base concentration (yellow); and 50th-precentile excess concentration compared to base concentration (green).

high nitrate days shows the spatial variability can range from nearly homogeneous to quite heterogeneous (such as the above noted days). It is likely that nitrate is driving the variability on such days and is responsible for many of the outlier mass concentrations at high base concentrations in Figure 4-3. There are at least two possible explanations for this behavior. First, there might be nitrate hot spots on high nitrate days. These hot spots are most likely to occur on days with a liquid aerosol due to high humidity. A qualitative inspection of surface weather on high nitrate days suggests they often coincide with fog events. The aerosol state (liquid versus dry), and degree of water content if wet, might vary across the network due to micrometeorology and this could influence nitrate uptake. Spatial variations in crustal material could also influence the pH of the liquid aerosol which in turn influences nitrate uptake. Second, there might be varying nitrate measurement artifact on certain high nitrate days. Even if the ambient nitrate concentration is spatially homogeneous, perhaps the Teflon filters are retaining nitrate to varying degrees and this artificially induces a spatial variability in both nitrate in FRM mass. A better understanding of the drivers for high spatial variability in FRM mass on high nitrate days is needed since only a few such events can dramatically affect the site-specific design values for the 24-hour standard. When such patterns are observed, perhaps the Teflon filters could be analyzed for nitrate by IC to determine the extent of nitrate retention; this would be particularly insightful for days coinciding with speciation sampler operation.

Figure 4-5 shows the same data as Figure 4-3 for Dearborn and Wyandotte but stratified to isolate the period 2004-2006 (the most-recent three-year period for the data set used in this study). Focusing on those days with base concentration below 35 μ g/m³ (to the left of the vertical solid line) but with site-specific concentrations approaching or greater than 35 μ g/m³

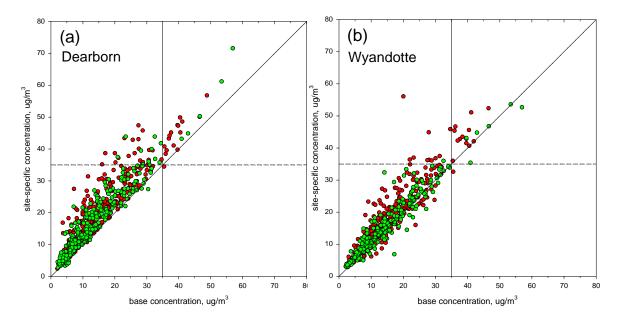


Figure 4-5. Site-specific concentrations versus base concentrations for valid network days at Dearborn (a) and Wyandotte (b) stratified by year: 1999-2003 (red markers); and 2004-2006 (green markers). Solid diagonal line is the base concentration (1:1 line), and the solid vertical line and dashed horizontal line are $35 \mu g/m^3$.

(above the dashed horizontal line), these days correspond to cases where nearby local emission sources are likely significant contributors to an exceedance (or near exceedance) that otherwise would not occur. For Dearborn (Figure 4-4a), there has been a decrease in frequency of such local plume days but they are certainly still present for the 2004-2006 period. For Wyandotte (Figure 4-4b), however, virtually all of the days with local source plumes driving an exceedance (or near exceedance) of the standard were prior to 2004. For the most recent data (2004-2006), Wyandotte does not appear to be disproportionately influenced by local plumes on exceedance (or near exceedance) days. Again, these figures do not represent all sample days but rather the subset of days with ten-or-more reporting monitors such that network-wide behavior could be characterized; however, as previously demonstrated, this subset of days does appear to be representative of the overall data set.

The analysis presented to this point clearly demonstrates that exceedances at most sites correspond to days when the entire network is exhibiting high PM concentrations and the rare extreme values are likely due to exceptional events or possibly a nitrate hot spot or measurement artifact. Some sites, especially Dearborn and Wyandotte, have a significant excess superposed on the base concentration leading to exceedances when the base concentration is as low as $20~\mu\text{g/m}^3$. It would be helpful to determine whether the day-to-day variability in the base concentration is driven by regional transport, poor atmospheric ventilation of urban-scale emissions, or a combination of these factors. There is no direct methodology to partition these influences but insights can be obtained by examining the PM speciation data and surface winds

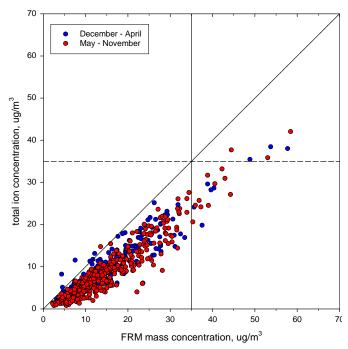


Figure 4-6. Total major ion concentration (sulfate, nitrate and ammonium) versus FRM mass at Allen Park, stratified by season.

distributions.⁴ **Figure 4-6** shows the contribution of major ions (sulfate, nitrate, and ammonium) to FRM mass at Allen Park for the period December 2002 through December 2006. There were valid FRM and speciation data for 21 of the 71 exceedances observed at Allen Park during this time period. Six (29%) of these 21 exceedances days exhibited major ion concentrations greater than 35 μ g/m³ and thus the major ions alone were sufficient to cause the exceedance.⁵ Over all 21 exceedance days the major ion contribution was 56-112% of the standard (median 84%) and 53-85% of the FRM PM_{2.5} mass concentration (median 71%). At Allen Park the major ions are more than half of the PM on all exceedance days and, since Allen Park is representative of the network-wide base concentration (Figure 4-3), the major ions are deemed the dominant PM component at all sites with small local plume impacts.

Aerosol composition at Allen Park was examined in more detail for the network-wide high PM days. Speciation data was available for 31 of the 41 days. **Figure 4-7** is a scattergram for sulfate and nitrate on these days with all sulfate assumed to be ammonium sulfate and all nitrate assumed to be ammonium nitrate. Summertime events typically exhibit high sulfate and relatively low nitrate. Wintertime events exhibit high nitrate but sulfate is typically ~50% or more of the nitrate concentration. Thus, while the wintertime events are often called high nitrate days, it should be noted that sulfate is also quite significant on these days. Overall, 55% of these

⁴ Typically synoptic winds would be analyzed to identify days conducive to regional transport but sulfate and nitrate will be used as a surrogate for regional transport in this work since the sulfate at Allen Park is overwhelmingly from regional transport and the nitrate is dominated by regional transport.

⁵ This assumes nitrate does not appreciably volatile from the FRM Teflon filter; in practice, the major ion concentration will be lower due to nitrate volatilization.

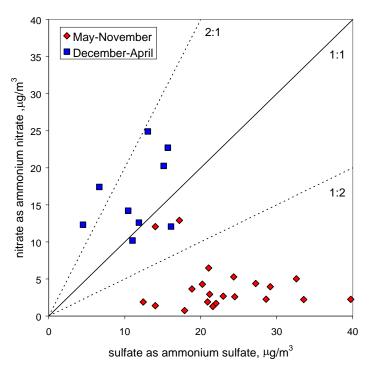


Figure 4-7. Ammonium nitrate and ammonium sulfate concentrations at Allen Park on network-wide high PM days.

31 days have a combined ammonium sulfate plus ammonium nitrate concentration exceeding $25 \,\mu\text{g/m}^3$ although, as previously mentioned, the contribution of these ions will be less on certain days after correcting for ammonium nitrate loss from the FRM Teflon filters.

Previous work has demonstrated that sulfate at Allen Park is overwhelmingly from regional transport; nitrate is likely dominated by regional transport but the precise split between regional and local sources remains unknown. Assuming the sulfate and nitrate are regionally transported, the total contribution from regional sources would likely be larger than the sum of the major ions (after correction for nitrate loss), but cannot be further quantified because organic matter likely has significant regional and local contributions and at this time the organic matter concentrations in Southeast Michigan cannot be apportioned to regional and local contributions on an average basis, no less a day-to-day basis. A multivariable linear regression was performed to explain the observed organic carbon (OC) in terms of observed sulfate and elemental carbon (EC). The unconstrained regression yielded ($R^2 = 0.50$, all units in $\mu g/m^3$);

$$OC = (1.53 \pm 0.22) + (0.176 \pm 0.029)SO_4 + (2.49 \pm 0.26)EC$$

where the 95% confidence interval for each regression parameter is reported. There are many caveats to this very simple approach to apportioning the OC. For example OC associated with sulfate would likely vary by season due to differences in secondary formation rates. The approach is merely intended as a crude way to assign OC to a component that associates with

local, primary emissions (OC associating with EC) and a component that scales with sulfate. The results should not be over-interpreted. Assuming an OM/OC ratio of 1.8, the scaling factor for organic matter associated with sulfate would be $0.32\times SO_4$. **Figure 4-8** shows the contribution of major ions measured at Allen Park to the base concentration on valid network days (Figure 4-8a) and the contribution of the major ions plus this crude estimate of the OM associated with sulfate (Figure 4-8b). The inclusion of associated OM substantially closes the mass balance on the base concentration. High base concentrations can largely be explained by regionally transported material alone and any concomitant increase in urban- and finer-scale contributions (for example, due to poorer atmospheric ventilation) might be relatively modest. From Figure 4-8a it can also be discerned that the major ion contribution deviates positively from the median contribution at base concentrations above ~ $20 \mu g/m^3$; it is above this value that major ions – and thus presumably regional transport – disproportionately influences the observed $PM_{2.5}$ mass concentrations.

Concentration enhancements in surrogates for regional transport (sulfate and nitrate as surrogates) and urban-scale local emissions (EC as surrogate) were examined for the network-wide high PM days. For each valid network day with speciation data at Allen Park, concentrations for the major aerosol components were compared to the monthly median values obtained by pooling over all valid network days and over all years. Sulfate was on average 4.5 times higher and nitrate was on average 2.8 times higher on network-wide high concentration days compared to the corresponding monthly median value. In contrast, EC was on average only 1.3 times higher on network-wide high concentration days compared to the corresponding monthly median EC concentration. Using the EC enhancement as an indicator for the atmospheric ventilation of urban-sale emissions, this analysis suggests reduced atmospheric ventilation on network-wide high PM days does indeed increase the urban- and-finer scale contributions but, in the absence of direct plume hits from near-field sources, the enhancement is much smaller than observed for the major ions which are associated with regional transport. OC was on average 1.6 times higher on network-wide high PM days compared respective monthly median concentration (1.8 times higher if the OC data is blank-corrected). This enhancement is between the values ascribed to regionally transported material and urban-scale emissions, consistent with OC contributions from a combination of local (urban or finer-scale) primary sources and secondary formation (whether local or regional in origin).

Lastly, the daily FRM sampling at Allen Park is exploited to determine whether high PM days at each site correspond to a spatially- and temporally- isolated excursion (high PM not observed at Allen Park which, as shown in Figure 4-3, largely captures the network-wide PM base concentration), a temporally-isolated excursion (high PM also observed at Allen Park on that day but not the preceding or following day), or a multiday PM episode (high PM observed at Allen Park on adjacent days). Multiday PM episodes are stratified into two groups – two successive high PM days at Allen Park, and three-or-more successive high PM days at Allen Park. Two successive high PM days could arise from a brief event that straddles midnight, while three-or-more more successive high PM days clearly indicate an episode with temporal persistence, whether caused by regional transport or sustained areawide air mass stagnations.

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⁶ EC has a day-of-week trend at Allen Park (higher on weekdays) so the analysis was repeated by first stratifying days as weekends and weekdays and then calculating the day-specific enhancement compared to the respective weekend or weekday monthly median value. The same average enhancement – a factor of 1.3 – was obtained.

High PM conditions were defined as $>30~\mu g/m^3$ for the interrogated site and $>25~\mu g/m^3$ for the reference site (Allen Park). **Figure 4-9** shows the site-specific distributions. Dearborn has the highest frequency of spatially- and temporally-isolated high PM days (red bars; dates for all sites listed in **Appendix C**). As previously described, nearby emission sources are most important at this site and can drive high PM days even when the network-wide base concentration is as low as $20~\mu g/m^3$. Relatively high frequencies of spatially- and temporally-isolated high PM days were also observed at Wyandotte, Southwest High School, and East 7 Mile. At all sites except Dearborn, high PM days most frequently correspond to multiday PM episodes (blue plus green bars) and most frequently to longer-persistence episodes (three- or-more days with high PM at Allen Park).

In summary -

- Network-wide high PM days are observed primarily in the summertime but are also observed in the wintertime.
- Network-wide high PM days in the summer typically have high sulfate and relatively low ambient nitrate. The nitrate contributions to FRM mass will be even lower after accounting from ammonium nitrate volatilization from the Teflon filter.
- Network-wide high PM days in the winter typically have high nitrate but ammonium sulfate is typically at least 50% of the ammonium nitrate mass so these events are actually high nitrate and high sulfate.
- Network-wide high PM days are primarily driven by regional transport with intensification of urban-scale emissions due to low atmospheric ventilation being relatively less important. Plume impacts from nearby emission sources are superposed on the network-wide baseline.
- Site-specific high PM days are most often observed when the area is experiencing a multiday high PM episode.

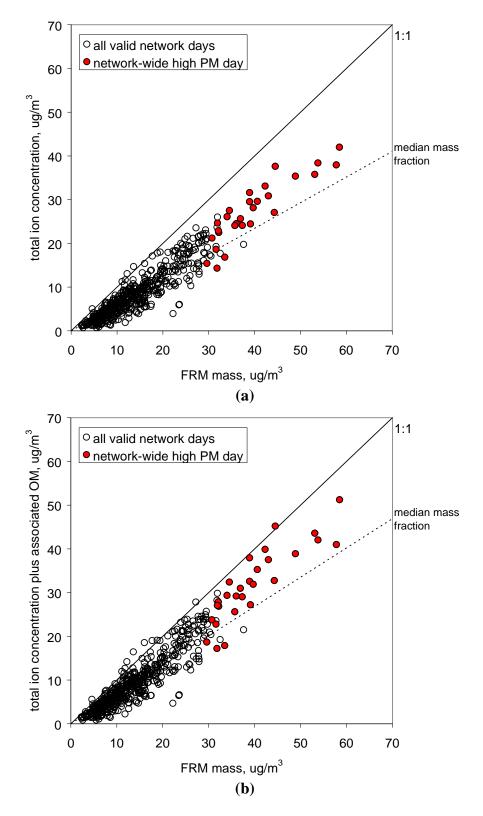


Figure 4-8. Allen Park species contributions to the base concentration on valid network days: major ions only (a); and major ions plus associated organic matter (b).

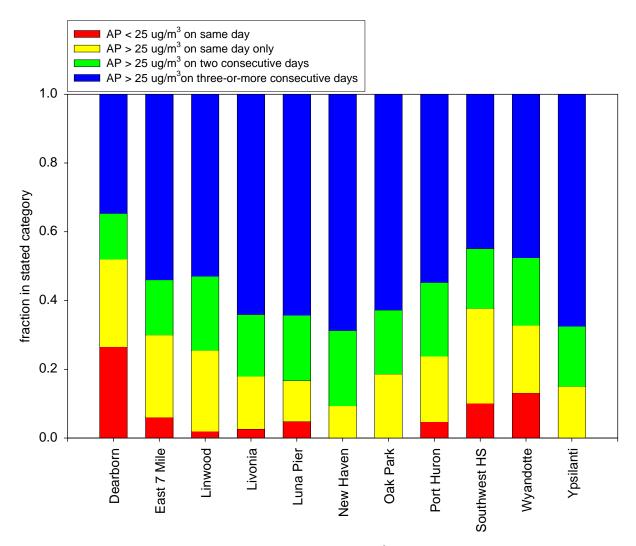


Figure 4-9. Classification of high PM days ($PM_{2.5} > 30 \mu g/m^3$) at each site in Southeast Michigan by the time series behavior of daily $PM_{2.5}$ at Allen Park.

5. RELATIONSHIPS BETWEEN SITE-SPECIFIC EXCESS MASS AND SURFACE WINDS

The previous section demonstrates that exceedances of the 24-hour PM_{2.5} NAAQS at most sites in Southeast Michigan are primarily driven by an enhancement of regionally transported material which elevates the concentration baseline upon which local contributions are superposed. There is typically a modest enhancement in the urban-scale contributions on high PM days, presumably due to coincident air mass stagnations. This does not imply that urban-scale contributions are small, but rather they are not significantly modulated on high PM days. At a few sites, plumes from nearby emission sources can substantially contribute to exceedances and near exceedances.

This section presents relationships between the excess mass (relative a reference concentration that should capture regional- and larger-scale influences, and to some extent urban-scale influences) and surface winds for each site. This analysis does not focus on excess mass observed on high PM days exclusively, but rather excess mass over the entire concentration distribution. While in principle there might be different local sources contributing on high PM days compared to all other days, the previous analyses do not provide any evidence for such preferential behavior and there is much statistical power to be gained from using the entire data set. Allen Park was initially chosen as a reference site for generating the times series of excess mass. Figure 4-3 demonstrates that Allen Park is indeed highly correlated with the network-wide base concentration on most days, although it is modestly influenced by nearby emissions for winds from the northeast (Dearborn) (Wade et al. 2008). Thus, the calculated excess mass concentrations at other sites might be biased low for winds from the northeast and sources in this direction might be damped or even missed. This limitation does not significantly affect the results for other bearings. Certain analyses were repeated using the network-wide base concentration as the reference for calculating excess mass. This reduces the number of sample days used in the analyses but removes any issues with directional bias introduced by using a single site as the reference. Finally, this methodology also relies on having representative surface winds at the site. It is not clear that hourly winds from Detroit Metropolitan Wayne County Airport will be representative for sites such as Luna Pier and Port Huron; however, the excess mass construct using Allen Park or the entire Southeast Michigan network to define a base concentration is perhaps an equally limiting factor when examining these distant sites.

Relationships between excess mass and surface winds were probed using Conditional Probability Function (CPF) and 1-D Nonparametric Wind Regression (NWR) plots. A brief summary of the methods and their utility is presented in **Appendix D**. The stability of CPF and NWR results was examined in various ways. Many analyses were run with the entire data set (1999-2006) and the most recent three years (2004-2006) since greatest interest is in current conditions. The former includes substantially more data and is less susceptible to spurious results. The threshold for high excess mass in the CPF analysis was chosen as the top quartile and each analysis was repeated using the top decile. Finally, sensitivity to days with high excess mass at the reference site (Allen Park) was examined by running most of the analyses on the full data set and then repeating the analyses with the data censored to exclude days where the excess mass was less than -2 μ g/m³. This threshold is based on two times the propagated collocated precision for the concentration difference using a collocated precision of 0.7 μ g/m³for a single measurement. Since the excess mass concentration is a 24-hour integrated parameter but the surface winds data

are hourly, care must be taken in handling calm hours. Calms days (operationally defined as six-or-more calm hours in the day) were retained but calm hours (wind speeds less than 2 mph) were removed.⁷

Figure 5-1 shows the CPF analysis results for each site using the top decile of the excess mass distribution as the threshold for a high concentration and for the period 1999-2006. Plots for Luna Pier, New Haven and Oak Park are excluded because they were either sensitive to the methodology or indistinguishable from the CPF plot for the absolute mass concentration at the site. The plots were nearly identical when using 1999-2006 data and 2004-2006 data only, which means the bearings of sources driving the excess mass have not changed over time. Dearborn, Newberry, and Wyandotte CPF plots all implicate sources to the southwest. These are likely nearby sources and, recalling that the prevailing wintertime surface winds are from the southwest (Figures 3-4 and 3-5) it is not surprising that the analyses in Section 4 implicated local sources as significant contributors to the excess mass at these sites given the favorable wind directions for advecting emissions from the sources to these sites. East 7 Mile, Linwood, and Southwest High School CPF plots exhibit higher frequency of excess mass with winds from the south/southwest, albeit weaker signatures than observed for Dearborn / Newberry / Wyandotte. Ann Arbor and Ypsilanti have a higher frequency of high excess mass when the winds are from the east and Livonia has a higher frequency of high excess mass when the winds are from the southeast. All three of these cases correspond to bearings in the direction of the Detroit population and/or commercial/industrial cores. However, surface winds from the east are not common and thus these sites are rarely downwind of the urban core zones. The CPF plot for Port Huron implicates sources at bearings east through south of the site which is consistent with the industrial zones along the U.S./Canada border including but not limited to petroleum refineries. In summary, the CPF plots are consistent with the local source impacts identified in Section 4 and consistent with known emission source regions. It appears this analysis can pick up the urban plume advected from Detroit to sites that are outside the urban emissions zone.

NWR plots show the expected excess concentration as a function of wind direction. Several analyses were performed using Allen Park data for the reference concentrations (see footnote #11). Subsequently, NWR plots were constructed using the network-wide base concentration estimates for the reference concentrations and those results, which were generally consistent with the results that were obtained using Allen Park as a reference, are presented here. Yearly box plots were generated for the excess PM_{2.5} mass at each site compared to the network-wide base concentration. FRM data for 2007 was also included in these time series plots but have not been examined in sufficient detail for incorporation into wind regression analyses. Given the seasonal and year-to-year variability in surface winds, the representativeness of valid network days was assessed by comparing to the annual surface winds for all days. **Figure B-2** shows annual wind roses for all days and valid network days.

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⁷ The following analyses were performed with results available upon request: CPF for top quartile of absolute concentration distribution (no subtraction of Allen Park concentration values), 1999-2006; CPF for top decile of entire excess mass distribution, 1999-2006; CPF for top quartile of entire excess mass distribution, 1999-2006; CPF for top decile of entire excess mass distribution, 2004-2006; CPF for top quartile of entire excess mass distribution, 2004-2006; NWR for entire excess mass distribution, 1999-2006; NWR for entire excess mass distribution, 2004-2006; and NWR for censored excess mass distribution, 1999-2006.

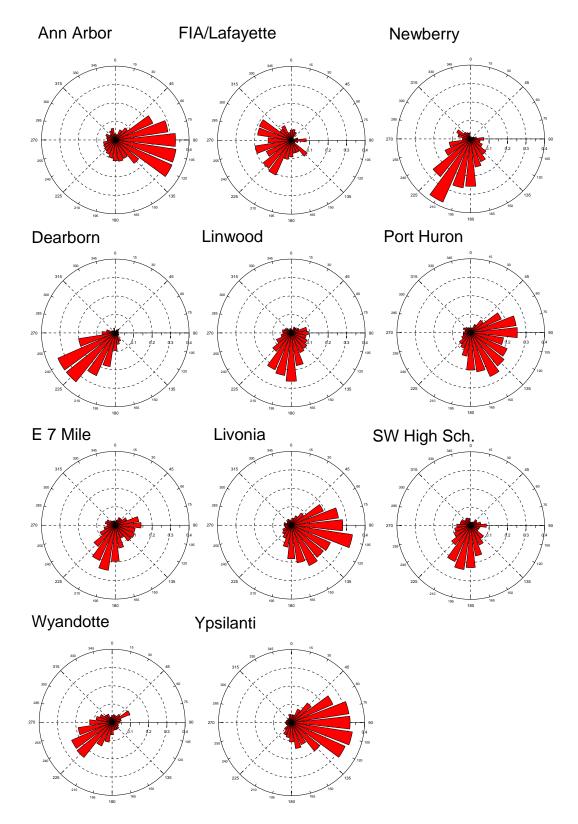


Figure 5-1. Conditional probability function plots for excess mass at each site relative to Allen Park, 1999-2006. Probabilities are for the top decile of the excess mass distribution.

Data for 1999 were excluded from the concentration time series because the network was being phased in during that year; there were only 14 valid network days and the valid network days winds distribution was markedly different from the 1999 annual surface winds distribution. For the remaining years the annual wind roses for valid network days showed good agreement with the annual wind roses for all days (Figure B-2), including 2006 which had a relatively low number of valid network days (also seasonally biased towards the first quarter) due to a reduction in sampling frequency at certain sites. These favorable comparisons demonstrate the representativeness in using valid network days to capture annual average behavior for the site-specific excess mass.

Figure 5-2 shows annual distributions of the network-wide base concentration. Similar distributions were observed for the first four years (2000-2003) with substantial year-to-year variability for the subsequent four years (2004-2007). The lowest median value was in 2004, followed by a relatively high mean value in 2005.

Excess mass annual time series and NWR plots were generated for each site with selected results presented in this report. NWR was performed after stratifying the data into two time periods: 2000-2003 and 2004-2006. Hourly surface winds distributions, **Figure 5-3**, exhibit a lower frequency of winds from the southwest for the 2004-2006 period compared to the 2000-2003 period. However, the NWR results are presented as the expected concentration value for each wind direction and thus are not influenced by the frequency distribution of wind direction. An apportionment of the average excess mass to wind sectors (and thus emission sources in that sector) would require a weighting of the sector-specific expected concentration by the sector-specific frequency of wind direction. This step is discussed in Section 6 as a recommendation for future work. For the purpose of identifying whether emission source strengths have changed over time, the expected concentration as presented in this section is appropriate since it is not influenced by year-to-year differences in the distribution of surface wind directions.

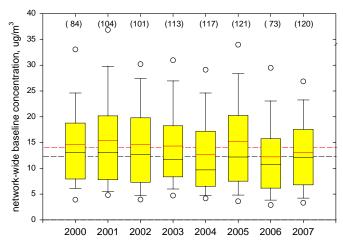


Figure 5-2. Annual distributions of the network-wide base concentration. Open circles are 5th and 95th percentile values; the red and black lines are mean and median values (solid within box for yearly, dashed for entire data set); and the number of samples is in parenthesis at the top of each plot.

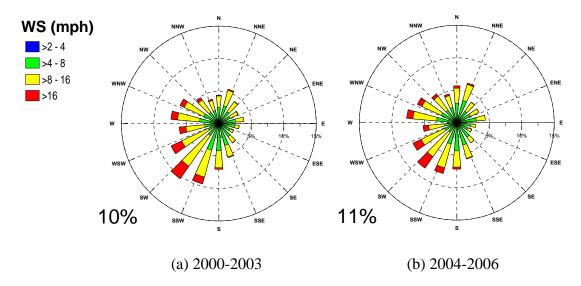


Figure 5-3. Wind rose for non-calm hours, observations at Detroit Metropolitan Wayne County Airport (DTW): (a) 2000-2003; and (b) 2004-2006. Calms (wind speeds less than 2 mph) are listed to the lower-left of each wind rose.

Ann Arbor, FIA/Lafayette, Newberry were excluded because sampling was not sustained over the 2000-2006 time period. Luna Pier and Port Huron were excluded because they are at the spatial extents of the geographic domain and both the base concentration estimates and surface winds might not be representative. **Figures 5-4** through **5-7** show the time series and NWR plots for Dearborn, Wyandotte, Southwest High School and Linwood, respectively. NWR results as polar plots are presented with the radial concentration units in $\mu g/m^3$; the red line is the average contribution and the pink lines are the 95% confidence intervals. NWR results were also generated after first censoring the data to remove extreme values and thereby stabilize the expected concentration estimates. The bottom plot in each panel of Figure 5-4 shows the NWR results for censored data, and includes the difference in expected concentration values between the two time periods (blue line). **Figure E-1** (**Appendix E**) presents NWR results for all sites as Cartesian plots for all data and censored data.

Dearborn (Figure 5-4) exhibits the average highest average excess mass. Both the mean and median excess mass decreased beginning in 2005 despite an increase that year in the mean base concentration (Figure 5-2). The time series for the annual base concentration and excess mass concentration are decoupled which adds confidence that the decreases in excess mass are real. Expected mass concentrations of at least 2 $\mu g/m^3$ were observed for all wind directions (center and bottom plots in Figure 5-4) with elevated excess mass for bearings 120-300°N and a maximum in the expected concentration at ~225°N of with concentrations 9 and 7 $\mu g/m^3$ for 2000-2003 and 2004-2006, respectively. The excess mass floor of ~2 $\mu g/m^3$ might be real since the Detroit population core, perhaps a surrogate for mobile source and area emissions, is to the

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⁸ Extreme values were defined as those values exceeding the upper outer fences (UOF) and lower outer fences (LOF) of the nonparametric distribution of excess mass: $C_{UOF} = C_{75} + 3 \times C_{IQR}$; and $C_{LOF} = C_{25} - 3 \times C_{IQR}$ where C_i is concentration i, subscripts 25 and 75 denote 25th and 75th percentiles, and IQR is the interquartile range ($C_{75} - C_{25}$).

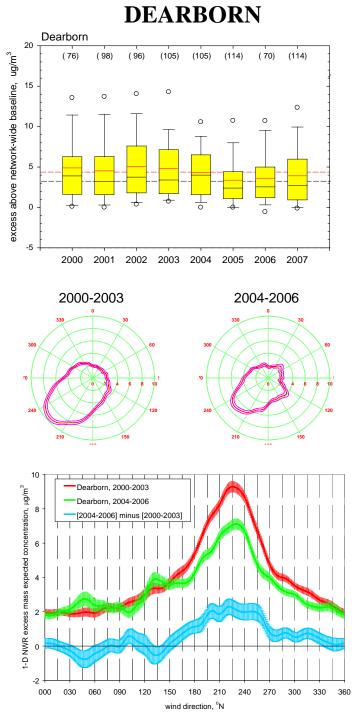


Figure 5-4. Excess PM_{2.5} mass compared to the network-wide base concentration. **Top:** box plots of annual excess mass concentration distributions. Open circles are 5^{th} and 95^{th} percentile values; the red and black lines are mean and median values (solid within box for yearly, dashed for entire data set); and the number of samples is in parenthesis. **Center:** 1-D NWR for excess mass at each site relative to the network-wide baseline concentration for 2000-2003 (left) and 2004-2006 (right). Average concentrations and 95% confidence intervals are red and pink lines, respectively, and radial units are $\mu g/m^3$. **Bottom:** 1-D NWR expected excess mass concentrations after censoring the data to remove extreme values.

WYANDOTTE Wyandotte (100) (107) (112) (69) (116) excess above network-wide baseline, ug/m³ 0 2000 2001 2002 2003 2004 2005 2006 2007 2000-2003 2004-2006 Nyandotte, 2000-2003 1-D NWR excess mass expected concentration, µg/m³ [2004-2006] minus [2000-2003] 6 270 030 060 090 120 210 240 180 wind direction, °N

Figure 5-5. Excess PM_{2.5} mass compared to the network-wide base concentration. **Top:** box plots of annual excess mass concentration distributions. Open circles are 5^{th} and 95^{th} percentile values; the red and black lines are mean and median values (solid within box for yearly, dashed for entire data set); and the number of samples is in parenthesis. **Center:** 1-D NWR for excess mass at each site relative to the network-wide baseline concentration for 2000-2003 (left) and 2004-2006 (right). Average concentrations and 95% confidence intervals are red and pink lines, respectively, and radial units are $\mu g/m^3$. **Bottom:** 1-D NWR expected excess mass concentrations after censoring the data to remove extreme values.

SOUTHWEST HIGH SCHOOL

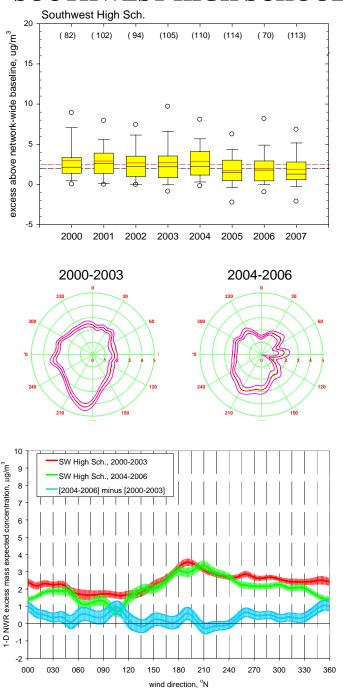


Figure 5-6. Excess PM_{2.5} mass compared to the network-wide base concentration. **Top:** box plots of annual excess mass concentration distributions. Open circles are 5th and 95th percentile values; the red and black lines are mean and median values (solid within box for yearly, dashed for entire data set); and the number of samples is in parenthesis. **Center:** 1-D NWR for excess mass at each site relative to the network-wide baseline concentration for 2000-2003 (left) and 2004-2006 (right). Average concentrations and 95% confidence intervals are red and pink lines, respectively, and radial units are μg/m³. **Bottom:** 1-D NWR expected excess mass concentrations after censoring the data to remove extreme values.

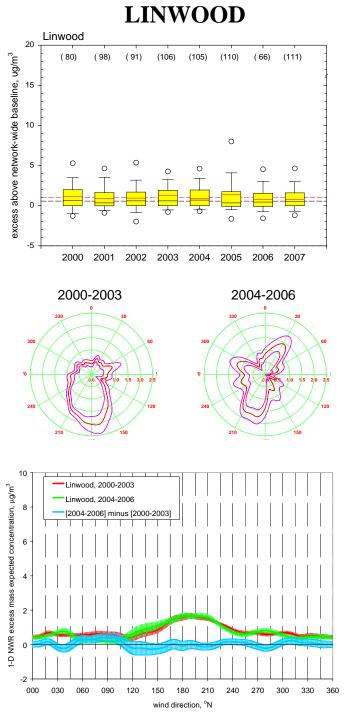


Figure 5-7 Excess $PM_{2.5}$ mass compared to the network-wide base concentration. **Top:** box plots of annual excess mass concentration distributions. Open circles are 5^{th} and 95^{th} percentile values; the red and black lines are mean and median values (solid within box for yearly, dashed for entire data set); and the number of samples is in parenthesis. **Center:** 1-D NWR for excess mass at each site relative to the network-wide baseline concentration for 2000-2003 (left) and 2004-2006 (right). Average concentrations and 95% confidence intervals are red and pink lines, respectively, and radial units are $\mu g/m^3$. **Bottom:** 1-D NWR expected excess mass concentrations after censoring the data to remove extreme values.

north and east and the site is located in the Detroit commercial/industrial core. It is possible, however, that the excess mass floor reflects contamination in the NWR analysis from using daily-integrated FRM mass together with hourly winds which might vary over the course of the day and thus smear short duration, high intensity contributions from the local emission sources across other wind directions. The period 2004-2006 does indeed exhibit lower excess mass concentrations compared to 2000-2003, consistent with Figure 4-5, with maximum reductions of ~2 μ g/m³ from ~195-255°N. The Dearborn NWR results were insensitive to the censoring of data to remove extreme values (Figure E-1). In summary, Dearborn is significantly influenced by local emission sources to the southwest of the site. This is also the direction of the prevailing wind and thus emission controls on these sources would have a dramatic impact on observed concentrations at the Dearborn site. There is also a significant urban scale contribution regardless of wind direction with expected concentration greater than 2 μ g/m³ (the excess mass floor is 2 μ g/m³ plus there are urban-scale contributions to the base concentration) although the relative contributions of mobile, area, and dispersed point source emissions is not clear.

Wyandotte and Southwest High School have similar mean and median excess mass, second after Dearborn. Let us first consider Wyandotte. The top plot in Figure 5-5 shows a dramatic and steady reduction in mean and median excess mass starting in 2004. Expected mass concentrations were lowest for wind directions from the east/southeast (indeed, ~0 µg/m³ for 2004-2006) which is consistent with the orientation of the site relative to local sources and the Detroit population and commercial / industrial cores. There are two sectors with high expected excess mass concentrations. One sector is broadly to the north/northwest with expected excess mass concentrations ~2 µg/m³ for 2000-2003 and with ~50% decrease for 2004-2006. This bearing is consistent with the location of the Detroit population and commercial / industrial cores which are north of Wyandotte. The other sector is to the southwest with a maximum excess mass concentration of nearly 4 μ g/m³ for 2000-2003 and with ~50% decrease for 2004-2006. Given both the alignment of this sector with the prevailing wind direction and the magnitude of the expected concentrations, the emission reductions achieved from this sector are most responsible for the observed decrease in the excess mass concentration over time. Local emission sources in this sector include a large quarry. Like Dearborn, the NWR results are relatively insensitive to the censoring of extreme values (Figure E-1). In summary, the wind direction dependence of expected excess mass concentration at Wyandotte is consistent with known emission sources / source regions. Expected concentrations have decreased over the past few years for two wind distinct bearings – north and southwest of the site – which suggest that emission reductions have taken place in both sectors. The reductions from the southwest are more significant to both annual average concentrations and high PM days.

Annual mean and median excess mass concentrations at Southwest High School also decreased starting in 2005 (Figure 5-6). NWR polar plots suggest the excess mass decrease was driven by emission reductions to the south/southwest. However, NWR results for all data and censored data (Figure E-1) demonstrate that a few extreme events were driving the higher expected mass from the south/southwest for 2000-2003 compared to 2004-2006. Indeed, the NWR plot for censored data (Figure 5-6, bottom) shows essentially no difference in expected excess mass at the location of the maximum (\sim 190°N). Emission reductions might be driving small differences in expected excess concentration between 2000-2003 and 2004-2006 at bearings of \sim 105, 250-300, and 355°N. The expected excess mass concentration floor was \sim 2 μ g/m³ for

2000-2003 and was modestly lower for 2004-2006. This floor is similar to the excess mass concentration floor observed at Dearborn which is consistent with their close proximity. In summary, in contrast to Dearborn and Wyandotte, individual local sources contributing to excess mass at Southwest High School appear to have smaller average impact and are potentially more numerous. Highest expected concentrations are from bearing of ~190°N and with extreme events realized primarily in the period 2000-2003. Based on Figure 4-3 these events typically occurred on relatively clean days (base concentrations 10-20 $\mu g/m^3$) and were not sufficiently high to cause exceedances. Like Dearborn, there is a relatively high floor for the excess mass over all wind directions but the relative contributions of mobile, area, and dispersed point source emissions is not known.

Linwood is located near the northeast corner of the Detroit industrial core. There are no discernible changes in the excess mass distribution over time (Figure 5-7). NWR polar plots show different features for the 2000-2003 and 2004-2006 time periods; the differences are driven by extreme values and NWR on the censored data yields very consistent concentrations profiles for the two time periods (Figure 5-7, bottom plot). The excess mass concentration floor is $\sim 0.5 \ \mu g/m^3$ with perhaps modestly lower values for bearings to the east/northeast (60-105°N). Increased excess mass is observed over the range 120-240°N with a broad maximum at 150-210°N. This behavior suggests that Linwood is at the fringe of the high emissions zone in the Detroit commercial/industrial core. Excess mass concentrations when the winds are from the southwest approach 1.5-2 $\mu g/m^3$, consistent with the floors at Dearborn and Southwest High School. For other wind directions, the expected concentrations are lower.

The above detailed analysis of four sites – Dearborn ,Wyandotte, Southwest High School, and Linwood – contributes to our understanding of local and urban-scale emissions contributions to observed PM mass. Local point source impacts are historically large at both Dearborn and Wyandotte but have been decreasing. In both cases, the largest contributors are located southwest of the sites, which is consistent with the prevailing winds. Dearborn and Southwest High School have expected excess mass concentrations of a least 1.5-2 $\mu g/m^3$ from all wind directions, while the minimum expected mass concentrations are lower at Wyandotte and Linwood. Thus, Dearborn and Southwest High School appear to be within the high emissions zone with contributions from multiple point sources and possibly significant contributions from area and mobile sources. The plume from this high emissions zone is advected downwind, consistent with the higher expected excess mass concentrations at Wyandotte and Linwood for winds from the north and south/southwest, respectively. Thus plume is superposed on the broader urban-scale emissions contributions from mobile, area, and possibly other point sources, which presumably also disperse as advected downwind.

This conceptual model is tested by examining excess mass patterns at sites outside the commercial/industrial core. **Figure 5-8** shows the Allen Park, Dearborn, Linwood, East 7 Mile, and New Haven NWR results for 2004-2006 censored data (note the concentration scale range is different than Figures 5-4 to 5-7). These five sites fall roughly along a transect oriented ~225°N and thus can be examined using the framework of Figure 2-1. The right figure shows that Allen Park (black line) and Linwood (red line) concentration profiles are nearly mirror images spanning the range $0.25-2 \,\mu\text{g/m}^3$ with a Linwood maximum and Allen Park minimum at $180-220^\circ\text{N}$. These sites are on opposite ends, and nearly equidistant, from Dearborn (clearly a

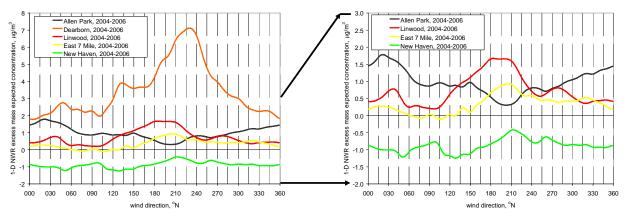


Figure 5-8. NWR expected concentrations for excess PM_{2.5} mass compared to the network-wide base concentration, 2004-2006 data censored to remove extreme values as described in footnote #12.

hot spot, left figure) and the industrialized zone to its southeast. This suggests there is an emissions plume emanating from Dearborn and industrial areas to its southeast, extending to the Detroit River, which impact Allen Park and Linwood similarly in terms of expected concentrations. Average mass contributions to these sites will differ with the frequency distribution of wind directions. The expected concentration from this plume decreases at more distant sites. It is still evident at East 7 Mile and is barely discernible from the baseline at New Haven. In addition to this plume from the Detroit commercial/industrial core, there appears to be a broader urban scale emissions profile as evidenced by the decreasing excess mass concentration floors with increasing distance from the Detroit population core. Expected excess mass concentrations for bearings outside the commercial / industrial core plume are $\sim 0.5~\mu g/m^3$ at Allen Park and Linwood, $\sim 0~\mu g/m^3$ at East 7 Mile, and about $\sim 1~\mu g/m^3$ at New Haven. Recall that the base concentration includes regional and some urban-scale contributions and the negative excess concentrations at New Haven suggest a maximum of at least $\sim 1.5~\mu g/m^3$ (based on $\sim 1~\mu g/m^3$ at New Haven and $\sim 1.5~\mu g/m^3$ at Allen Park/Linwood) for the urban-scale contributions within the metro area

Six sites – Allen Park, East 7 Mile, Livonia, New Haven, Oak Park, and Ypsilanti – are considered from a complementary perspective. Figure 4-3 demonstrates that large excess mass excursions above the network-wide base concentration are relatively rare at these sites. There are occasional extreme excess values which are both positive and negative with respect to the base concentration, so the excess mass distributions were stabilized by censoring to remove extreme values. NWR results for all these sites using both all data and censored data are presented in Figure E-1. For each site, all expected concentrations were scaled so the lowest expected concentration as a function of wind direction was zero (concentrations are negative when the expected concentration is below the base concentration) and the normalized to a maximum expected concentration of unity. **Figure 5-9** shows the scaled NWR concentration plots are superposed on 2000 census tract level population density data. The plots are ordered clockwise about Detroit area starting with Ypsilanti and ending with Allen Park. The following features are observed.

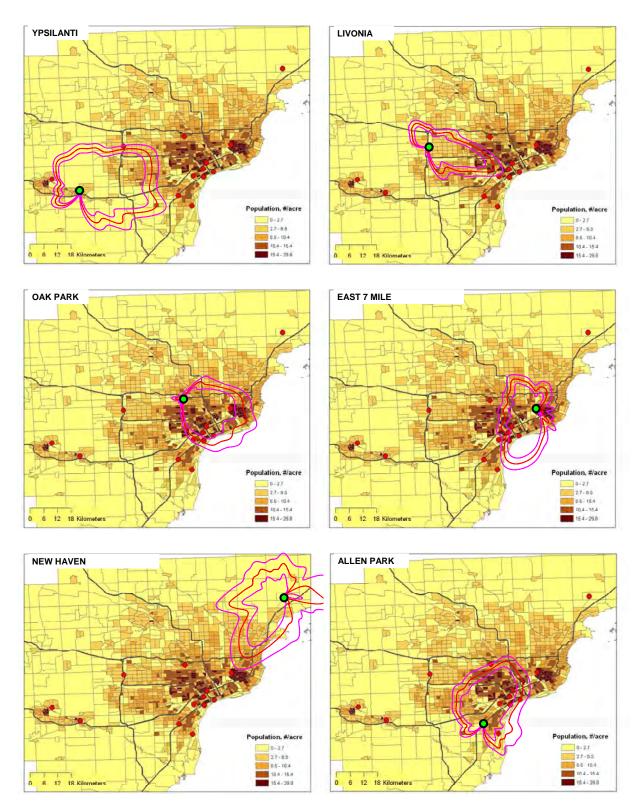


Figure 5-5. 1-D NWR for excess mass at each site (green circle) relative to the network-wide baseline concentration for 2004-2006, censored to remove extreme values according to footnote #12. NWR results are superposed on 2000 census tract population density data.

- *Ypsilanti*. The expected excess mass concentration ranged from -1 to +1 μ g/m³. Bearings of highest excess mass concentration correspond to an urban plume emanating from the east, consistent with the Detroit population and commercial/industrial cores.
- *Livonia*. The expected excess mass concentration ranged from -1 to +1 μg/m³. Bearings of highest excess mass concentration correspond to an urban plume emanating from the Detroit population and commercial/industrial cores with potentially larger impacts from the latter core.
- Oak Park. The expected excess mass concentration ranged from -0.5 to +1 μg/m³. The higher floor is consistent with this site being closer to the Detroit population and commercial/industrial cores than Ypsilanti and Livonia. Bearings of highest excess mass concentration range from due east to due south, consistent with the Detroit population and commercial/industrial cores.
- East 7 Mile. The expected excess mass concentration ranged from 0 to +1 µg/m³. Highest concentration bearings are to southwest with elevated expected concentrations also to the west, consistent with the Detroit commercial/industrial and population cores, respectively.
- New Haven. The expected excess mass concentration is nearly constant at -1 μg/m³. A bearing of modestly elevated expected excess mass concentrations is to the southwest, aligned with the Detroit commercial/industrial and population core.
- Allen Park. The expected excess mass concentration ranged from 0.5 to +2 μg/m³. High concentration bearings are to the north through east of the site, consistent with the Detroit population and commercial/industrial cores.

These six sites completely surround the Detroit population and commercial / industrial cores and in each case demonstrate highest expected excess mass for winds from the core zones. This analysis clearly demonstrates the ability to decouple excess mass concentrations from the network-wide base and to draw a consistent interpretation from the results across all sites. Ypsilanti, Livonia and New Haven all have expected excess mass concentrations of about $1~\mu g/m^3$ when the sites are upwind of the Detroit population and commercial/industrial cores. Thus a reasonable estimate for the average urban-scale contribution to the average network-wide base is about $1~\mu g/m^3$. The 2004-2006 average base concentration of $13.5~\mu g/m^3$ could be apportioned as $12.5~\mu g/m^3$ from regional sources and a $1~\mu g/m^3$ portion of the urban-scale contributions. Urban-scale emissions contribute, on average, up to $3~\mu g/m^3$ depending on location within the Greater Wayne County area and the specified wind pattern. They appear to be greatest in the Detroit commercial/industrial core and might include significant contributions from point sources that are either far enough from the monitor and/or low enough in emissions to not be distinguished from the urban-scale contributions. Large local emission sources can exert significant influence on near-field monitors as demonstrated for Dearborn and Wyandotte.

In summary, analyses of $PM_{2.5}$ mass concentrations – separated into network-wide base and site-specific excess mass components – together with surface winds provide the following model for $PM_{2.5}$ mass concentrations over Southeast Michigan (excluding the distant sites of Ann Arbor, Luna Pier, and Port Huron):

- At most sites, high PM days coincide with network-wide high PM levels and typically occur when the area is experiencing a multiday high PM episode (Section 4).
- Hot spots of high PM concentration arise near large emission sources and, in the cases of Dearborn and Wyandotte, can drive exceedances (or near exceedances) of the daily standard. In both cases, the high emissions zones are located to the southwest of the monitoring sites. This is consistent with the prevailing winds from the southwest exacerbating the frequency of high impact days. Expected concentration contributions from these zones have decreased in recent years, presumably due to emission reductions.
- An idealized conceptual model for PM behavior, that explains, both daily and annual PM mass trends, consists of four layers: (1) regionally transported PM with average concentration of about 12.5 μg/m³ over the 2004-2006 period; (2) urban-scale emissions from mobile, area, and point sources that contribute up to 3 μg/m³ for specific surface winds patterns, depending on location, with less impact when averaged over the distribution of wind directions impacting a given site; (3) a PM plume from the Detroit commercial/industrial core that remains distinct from the other urban-scale contributions for sites as distant as East 7 Mile and possible New Haven; and (4) localized hot spots for cases when certain monitoring sites are directly impacted by emission plumes from nearby sources with high potentially impacts on a given day and less impact when averaged of the distribution of wind directions impacting such sites.

6. SUMMARY, POLICY-RELEVANT IMPLICATIONS, AND RECOMMENDATIONS

Summary

At most sites in Southeast Michigan, exceedances occur predominantly – and in some cases exclusively – on days when the entire network is exhibiting high PM. For thirteen of the fifteen monitors, at least 85% of exceedances occurred on days when the entire network was exhibiting high PM. Drivers for network-wide high PM are regional transport, poor atmospheric ventilation across the network, or both of these conditions. Two monitors in particular – Dearborn and Wyandotte – frequently had exceedances on days when the entire network was not exhibiting high PM with 38% of the Dearborn exceedances and 27% of the Wyandotte exceedances occurring on such days. On these days, these sites are significantly influenced by nearby emission sources. Dearborn continues to be influenced by nearby emission sources which drive exceedances (or near exceedances) on days when the network-wide base concentration is as much as 15 μ g/m³ below the standard, although the frequency of such days appears to be decreasing. In contrast, at Wyandotte all of the days for which nearby emission sources were disproportionately contributing to exceedances (or near exceedances) of the standard were prior to 2004. At those sites not significantly impacted by plumes from nearby emission sources, high PM days are most often observed when the area is experiencing a multiday high PM episode.

Seasonal variations in surface winds can explain the seasonal differences in frequency of high PM days between sites in the Greater Wayne County area. Sites such as Allen Park, which are relatively free of impacts from nearby sources, exhibit similar seasonal behavior with high PM days most frequent in the summer followed by the winter. The monthly distribution of high PM days is distorted for sites with significant nearby emission sources since there is a strong seasonality to the surface winds distributions. For example, Dearborn has as many high PM days in the winter as in the summer, with the winter season having a stronger contribution from nearby sources located to the southwest of the site.

Network-wide high PM days have disproportionately high contributions from the major ions (sulfate, nitrate, and ammonium) compared to other days. Summertime events are dominated by sulfate and wintertime events are dominated by ammonium nitrate. However these so-called high nitrate days typically have ammonium sulfate concentrations that are at least 50% of the ammonium nitrate concentration. The role of sulfate on such days should not be overlooked when considering the impact of SO_2 -based regional control strategies. In general, network-wide PM base concentrations above ~20 $\mu g/m^3$ tend to be disproportionately influenced by major ion contributions and are indicative of a disproportionately high influence from regional sources. On network-wide high PM days the sulfate and nitrate concentrations are typically several times higher than the corresponding monthly median concentrations. In contrast, EC is on average only 1.3 times higher. This pattern suggests that network-wide high PM days are primarily influenced by regional transport and are to a much lesser extent influenced by a increased levels of urban-scale emissions arising from poor atmospheric ventilation. As previously stated, emission sources near the monitoring sites can still exert significant plume impacts regardless of the network-wide PM behavior driven by regional and urban-scale contributions.

Many network-wide high PM days coincide with high nitrate, and in some cases there is substantial variability in PM mass concentration across the network on these days. It is important to determine whether the variability is real or a measurement artifact, because these days can influence into the site-specific design values and in general can explain many of the exceedingly high PM levels observed at certain sites that otherwise do not appear to be disproportionately impacted by nearby local emission source plumes on high PM days.

A conceptual model for fine PM over central Southeast Michigan includes four elements. The largest contributor to annual average PM is regionally transported material that is temporally varying but spatially homogeneous over the domain (indeed, this defines the spatial extent of the domain for the conceptual model which in this case excludes the distant sites such as Luna Pier and Port Huron). On some days, regionally transported PM contributions can be sufficiently high to cause exceedances of the daily standard when coupled with average contributions from urban- and finer-scale emission sources and, in relatively rare cases, the regionally transported PM alone can cause exceedances of the daily standard. Urban-scale emission sources (mobile, area, and point sources) create a spatial PM gradient across central Southeast Michigan. For winds from a fixed direction, these contributions create an "urban plume" with decreasing impacts with increasing distance from the zone of highest emissions activity. The spatial gradients can be clearly discerned when binning data from days with common wind patterns and become smeared, to some extent, when averaging over periods with varying wind patterns. At many sites (e.g., Linwood, East 7 Mile) a PM plume emanating from the Detroit commercial / industrial zone, which is nominally Dearborn and areas to its southeast towards the Detroit River, including Zug Island, can be distinguished from the aforementioned urban plume. At sites further downwind, such as New Haven, these two plumes have essentially merged into a single urban plume. Average impacts from these plumes depend on the monitoring site location relative to the Detroit population and commercial/industrial core zones and the frequency that winds from these zones transport the plumes to the monitoring site. Finally, certain monitors in the Detroit commercial/industrial zone are significantly impacted by emissions from nearby sources that ultimately contribute to the distant PM plume emanating from this zone. At Dearborn and Wyandotte these sources are located southwest of the monitoring sites and historically have contributed to exceedances (or nears exceedances) of the daily standard. These sources also disproportionately influence the annual standard because they are oriented upwind of the sites for prevailing winds from the southwest.

In summary, emission controls that reduce regionally transported PM are needed to reduce the frequency of violations of the 24-hour PM_{2.5} standard. Emission reductions from a few specific point sources, especially near Dearborn, are also needed. While regional and point source controls will decrease annual average PM_{2.5} concentrations for standardized synoptic weather patterns, relatively small but significant decreases could also be realized by controls on other urban emission sources such as mobile, area, and other point sources.

Policy-Relevant Implications

The policy-relevant implications are framed using the Weight-of-Evidence (WOE)⁹ prepared in support of the control strategy package for the attaining the $PM_{2.5}$ annual average standard. The WOE elements are in regular typeface with comments, based on this work, added in italics.

I. Inventory

- I(c)... National stationary source controls, including CAIR and the NO_x SIP call, are expected to reduce point source NO_x emissions by 40% and SO₂ emissions by 15% between 2002 and 2009. These reductions are very important to the daily standard control strategy because exceedances of the daily standard tend to occur on days with network-wide high PM from regional transport of species including but likely not limited to ammonium sulfate and ammonium nitrate.
- I(c)... [There] is compelling evidence that areas in Southeast Michigan that are currently attaining the [annual] standard will remain in compliance. For the period 2004-2006, only one monitor in Southeast Michigan was in compliance with the 24-hour standard. While reductions in local emissions should prevent backsliding, daily exceedances are coupled to the frequency and severity of regional transport episodes which can vary from year to year. For example, 2005 was severe in terms of regional transport and this is reflected in the elevated 98th percentile concentrations across all sites in Southeast Michigan.
- I(d)... While these [programmed] reductions are already having a significant, positive impact in Southeast Michigan and will continue to do so in future, we cannot assume that they will result in attainment at Dearborn and Southwestern High School (SWHS), the two monitoring sites that are still exceeding the annual standard. Additional reductions in the vicinity of these sites are needed. Dearborn is currently the design value site for the daily standard. This study has demonstrated that contributions from certain local emission source have decreased in recent years. In contrast, the magnitude of the urban plume, which includes a broader range of emission sources, has not significantly decreased in recent years as evidenced by expected concentration patterns observed at sites such as East 7 Mile and Linwood.
- I(e)... The area surrounding the two non-attaining monitors in Southeast Michigan contains a complex array of emission sources. Some of these sources may be significant contributors because their emissions occur closer to ground level and/or because of their proximity to a monitor. However, many of these are area sources that are exempt from MDEQ's emissions inventory reporting requirements so their exact contribution is unknown. In addition, many of these smaller sources have little or no emission controls while larger sources already have controls in place. At these sites and a few other sites, the analysis in Section 5 point towards the bearings where efforts should be focused to refine emission estimates in particular, zones southwest of the monitoring sites due to prevailing wintertime surface winds lead to lead to higher PM than the rest of the Southeast Michigan network.
- I(f)... A number of industrial facilities in the area surrounding the Dearborn, Southwestern High School, and Wyandotte monitors have either closed or scaled back their operations since 2002. These changes are likely contributing to the more rapid decrease in PM_{2.5} levels observed at industrial monitoring sites. While we have indeed identified decreased local emission source contributions at some sites.
- I(g)... In addition to changes that have already taken place, significant local PM_{2.5} reductions will be achieved from controls that are currently being phased in at the Severstal and U.S Steel facilities as well as the Marathon oil refinery. All three of these facilities are less than three miles from the

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⁹ "Weight of Evidence for the Southeast Michigan PM2.5 Attainment Strategy", 8/28/2007 draft, prepared by SEMCOG and MDEQ.

monitors measuring the highest PM_{2.5} concentrations in the region - Dearborn and Southwestern High School. Based on a recent EPA study as well as permit application data, MDEQ estimates these controls will provide a combined PM_{2.5} emission reduction of 330 tons per year. These reductions should reduce the frequency of high PM days that occur at these sites when the network-wide baseline concentration is relatively low, thereby reducing the frequency of spatially isolate exceedances and near exceedances. Conditions on network-wide high PM days will also be improved. Note that in these cases the sources are located at bearings where prevailing winds can favor plume impacts at the monitoring sites, especially in the wintertime. Impacts can be tracked by periodically updating the NWR plots and the evaluating shifts in the distribution of site-specific high PM days by month. For example, if local source impacts at Dearborn are indeed reduced, we would expect the distribution of high PM days by month for Dearborn to shift to more-closely resembling Allen Park. Intersite differences between Allen Park and Dearborn should also decrease for the relevant species in the CSN data.

• I(h)... There are a significant number of storage piles, unpaved lots, and parcels of barren land in the vicinity of the Dearborn and SWHS monitors. While most emissions from these sources are larger than 2.5 microns, their collective impact is cause for concern. As part of a previous particulate SIP, a number of facilities in the area do have fugitive dust plans. However, many others do not. *The contribution of fugitive dust to high PM days was not explicitly examined.*

II. Monitoring:

- II(a)...PM_{2.5} in Southeast Michigan is comprised largely of sulfates, nitrates, and organic carbon. At the Dearborn monitoring site, there is also a significant "crustal" component, which is largely iron. The excess iron at Dearborn is largely related to steelmaking operations and efforts to reduce these emissions will reduce the local source impacts on exceedance and near exceedance days at Dearborn.
- II(b)... Southeast Michigan's current nonattainment designation pertains to the annual standard, not the daily standard. Developing a control strategy to address an annual standard is complicated because sources may be significant contributors on certain days or during certain times of the year but not during others. However, recent source apportionment studies show that the source contributions to PM_{2.5} on an annual average basis are similar to those on high PM_{2.5} concentration days. This suggests that a strategy designed to reduce annual average PM_{2.5} concentrations will also be effective in reducing high daily PM_{2.5} concentrations. Strategies targeting regional sulfate and nitrates will be important towards meeting both standards; in the absence of significant sulfate reduction it might be very challenging to meet the daily standard, even with aggressive local source controls. SO₂ reductions might not be fully realized as PM reductions if ammonium nitrate formation is ammonia limited.
- II(c)... The entire Southeast Michigan area has been designated nonattainment. However, the only monitors measuring violations of the standard are located in a small portion of eastern Wayne County. For 2004-2006 all Southeast Michigan sites except Ann Arbor violated the daily standard. However, based on 2005-2007 data several sites were in compliance. This demonstrates the year-to-year variations that can be induced by synoptic weather patterns which in turn include the frequency and severity of regional transport events.
- II(f)... Since 2000, PM_{2.5} concentrations at all sites in the region have steadily declined. The 3-year average concentration dropped 1.6 µg/m³ between 2002 and 2006.. The largest decreases have occurred at the sites with the highest concentrations: Dearborn (2.69 µg/m³), SWHS (2.16 µg/m³), and Wyandotte (3.04 µg/m³). Dramatic year-to-year swings in the 98th percentile concentration are observed throughout the Southeast Michigan network due to varying impacts from regional transport. In this sense the daily stand is more "volatile" and it is harder to discern long term trends.
- II(g)... PM_{2.5} concentrations at monitoring sites in the industrial core of Southeast Michigan's nonattainment area (Dearborn, SWHS & Wyandotte) have been decreasing faster than other sites. This is likely due to changes in emissions in the industrial area. *Reduced impacts from local emission*

- sources at Dearborn and especially Wyandotte have indeed been demonstrated in this study; average reductions are smaller at SWHS and are coupled to a lower frequency of extreme events which might have been caused by upsets at industrial facilities. Of course, any future industrial activity expansions that increase emissions could at least partially reverse these trends.
- II(k)... At the time Southeast Michigan was designated nonattainment, monitoring data showed the Luna Pier monitor in Monroe County was violating the annual standard. This monitor is located in the southeastern corner of the county, one mile north of the Ohio border. In its February 2004 PM2.5 nonattainment designation recommendation to EPA, MDEQ argued strongly that Monroe and Wayne counties should be designated as separate nonattainment areas because PM_{2.5} levels at the Luna Pier monitor tracked far more closely with those in Toledo. Many regional events operate on a large enough spatial scale to similarly impact Luna Pier and the Wayne County area. Indeed, the behavior at these two areas appears coupled on high PM days. That said, Figure 4-3 demonstrates that Luna Pier and Port Huron have the greatest dispersion about the network-wide base concentration including many days that are lower than the base concentration and thus they are not fully coupled.
- II(1)... MDEQ and SEMCOG also showed that levels at the site had been decreasing in recent years and trend data indicated the monitor would likely measure attainment by the end of 2004. This was an accurate prediction. Levels at the site have continued to track those in Toledo and monitors in both areas have measured attainment of the standard since 2004. In 2005, EPA redesignated the Toledo area as attainment. Luna Pier continues to violate the daily standard (2004-2006 data).
- II(m)... In addition, as of 2006, monitors at Allen Park, Linwood and Wyandotte are now measuring attainment [of the annual average NAAQS]. *Allen Park, Linwood and Wyandotte continue to violate the daily standard* (2004-2006 data).
- II(n)... The area where the two remaining violating monitors (Dearborn and SWHS) are located is one with a history of particulate matter problems, associated with local industrial sources. [...] the areas are nearly identical. The primary source of the former PM₁₀ problem was determined to be a few local industrial sources. Emissions from these sources were reduced and the region came into compliance in 1996. The overlap of the Total Suspended Particulate (TSP) and PM₁₀ nonattainment areas with the PM2.5 nonattainment area, and the successful attainment of those standards after the application of local controls, suggests that the most effective attainment strategy is to focus on local emission reductions from sources in this area. Reductions in local emissions near Dearborn have decreased the frequency of high PM days resulting from local emission sources (Figure 4-5) and the programmed emission reductions should further help to lower the local emission source influences. Local emissions near SWHS frequently lead to near exceedances when the network-wide PM levels are otherwise low but, unlike Dearborn and Wyandotte, the local source contributions appear to be just low enough to avoid exceedances on otherwise relatively clean days.
- II(o)... Various analyses of both local and regional monitoring data all indicate that Southeast Michigan's nonattainment problem is caused by a combination of regional transport and local emissions from sources in the vicinity of the violating monitors. This has also been demonstrated for the daily standard in this study with local source impacts driving the frequency of exceedances at a few sites and regional transport contributing to exceedances at all sites.
- II(o)(3)... Lake Michigan Air Directors Consortium (LADCO) analysis of rural background concentrations versus urban excess in the Midwest shows that the vast majority of PM_{2.5} measured in our region is coming from outside Southeast Michigan. This pattern is certainly the case on networkwide high PM days that coincide with the most frequent exceedances of the daily standard.
- II(o)(6)... A wind rose for the iron component of PM_{2.5} at Dearborn points directly to the southwest. Conversely, the iron wind rose for Allen Park, while measuring much lower levels, points to the northeast. The Allen Park monitor is approximately five miles southwest of Dearborn. Additional wind direction analysis shows that, when winds are from the southwest average crustal concentrations at Dearborn are over 2.5 μg/m³ higher than those at Allen Park and are sometimes as much as 6 μg/m³ higher. This clearly indicates a significant local iron source directly between these

two sites (which are approximately five miles apart) and closer to the Dearborn monitor. The importance of local emission sources to the southwest of Dearborn has been demonstrated in this study. These sources likely lead to the high frequency of wintertime high PM days at Dearborn compared to the Allen Park and the rest of the network since the prevailing surface winds are from the southwest during the winter.

- II(p)... The localized nature of Southeast Michigan's nonattainment problem makes broad-based application of control measures throughout the official seven-county nonattainment area an ineffective and unproductive strategy for bringing the region into compliance. All available data show that targeted local organic carbon emission reductions, coupled with the iron reductions resulting from planned steel mill controls, will be the most cost-effective way to bring the region into attainment. Similarly, while the relative roles of regional- and urban-scale contributions have not been quantified in this study, it has been shown that regionally transported species concentrations are enhanced much more than urban-scale tracer species on high PM days.
- II(y)... The difference in PM_{2.5} is highest from the southwest and west wind directions when nearby monitors are subtracted out of the Dearborn concentration. This would indicate that there is a large local source [or sources] between the Dearborn and "background" monitors (Allen Park, Luna Pier & Ypsilanti). *This was clearly demonstrated in Section 5 of this study*.

Recommendations for Future Work

Projects currently funded by LADCO will likely provide considerable refinements to this analysis. For example, sampling for organics speciation and the planned data analysis – including source apportionment – should inform the assignment of organic carbon to local and regional sources. . MDEQ is deploying semicontinuous monitors for particle speciation and high time resolution data can be particularly powerful for identifying source contributions. Results from the summer 2007 USEPA/ORD measurement intensive should be particularly insightful since PM elements were measured at high time resolution. Outdoor measurement data from USEPA's DEARS project could also be exploited.

A background monitoring site would be helpful, but care is needed to ensure it is sufficiently coupled to the Detroit area in terms of air mass behavior. The site needs to be far enough away to not be influenced by the urban area, but close enough to still be similarly impacted by regional transport. This is even more critical if measurements will be conducted for 24-hour periods once every few days as the "snapshots" at each monitor must be synchronized across the domain. For example, the background site effectively used for St. Louis is about 100 km to the south which places it upwind of the city for the prevailing surface winds. Perhaps more importantly, however, the line connecting this site to the urban area is nearly perpendicular the direction of summertime synoptic air mass flows which lead to high PM in St. Louis (St. Louis is often on the backside of a high pressure system located over the Great Lakes, which lead to regional transport from the east/southeast). This orientation might actually be preferred as an upwind site – from the surface winds perspective – because the sites will be impacted at the same time for regional air masses with spatial extents greater than the site separation.

A better understanding of the drivers for high spatial variability in FRM mass on high nitrate days is needed since only a few such events can dramatically affect the site-specific design values for the 24-hour standard. When such patterns are observed, perhaps the Teflon filters could be analyzed for nitrate by IC to determine the extent of nitrate retention; this would be

particularly insightful for days coinciding with speciation sampler operation to place the Teflon filter nitrate levels in context of the ambient concentrations.

It might be possible to better characterize the spatial pattern of urban-scale emission source contributions by performing reflectrometry measurements on archived $PM_{2.5}$ FRM samples. The parameter from these measurements is typically highly correlated with thermo-optical EC. Thus, it provides an inexpensive approach to characterizing EC spatial profiles which might be a reasonable surrogate for urban-scale emission sources.

FIA/Lafayette and Newberry sites were excluded from this analysis because they commenced operation relatively recently. The analysis used concentration percentiles across the network to define network-wide behavior and thus it was necessary to use a temporally consistent subset of the network. The base concentrations generated from this work could be applied to the FIA/Lafayette and Newberry data to split observed concentrations into base and excess mass components, and then nonparametric wind regression (NWR) could be used to determine expected excess mass concentrations as a function of wind direction. This work, which is relatively modest in level of effort, could significantly contribute to our understanding of PM behavior in the Detroit commercial/industrial core.

Relatively little attention has been given to the behavior at Luna Pier and Port Huron on high concentration days. The analysis approach taken in this report has limitations when applied to sites at such large distances from the Greater Wayne County area. Locally-collected surface winds should be used to examine behavior at these sites. Luna Pier and Port Huron must also be considered within the context of emissions from Toledo and Sarnia, respectively.

NWR results are presented in Section 5 in terms of the expected concentration of excess mass observed at a site when the winds are from a given direction. This has proven to be insightful because the results do not depend on the frequency distribution of wind directions which can vary from year-to-year and confound the identification of temporal changes in emissions. From this analysis we can state that emissions from a certain wind sector have been modulated by a certain amount over time. However, we are also interested in an apportionment of the contributions of such emission sources to the PM burden at the monitoring site. This would require weighting the expected concentrations by the frequency of winds from each direction sector. We have developed a conceptual model based on the network behavior for various surface wind patterns. The next step is to incorporate the observed distribution of wind patterns to quantify how the observed PM levels are being modulated. We have shown that a PM plume from the Detroit commercial/industrial core impacts Linwood. By incorporating the observed distribution of surface wind directions we could quantify the annual-average PM mass from this plume impacting the site. This analysis would extend the focus of this report on high PM days to more robustly characterize annual average behavior and could be instrumental in providing a framework for assessing the effectiveness of emission control programs.

7. REFERENCES

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APPENDIX A: PM_{2.5} HIGH CONCENTRATION DAYS BY MONTH

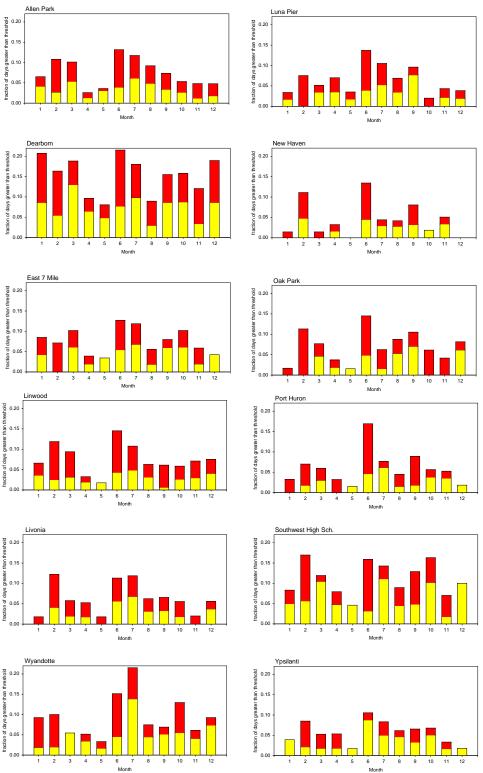


Figure A-1. Distribution of $PM_{2.5}$ high concentration days by month, 1999-2006. Ann Arbor, FIA/Lafayette, and Newberry are excluded because FRM data was not collected at these sites over the entire time period.

APPENDIX B: HOURLY WIND ROSES

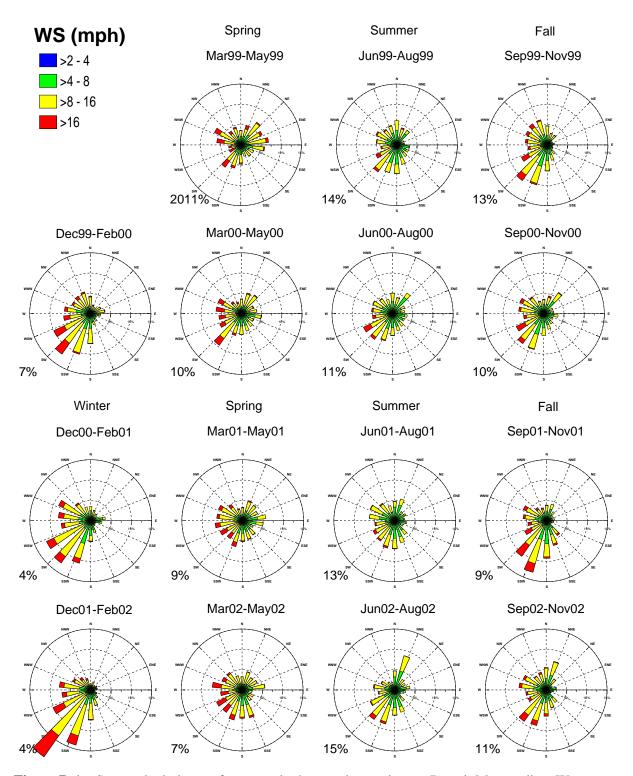


Figure B-1. Seasonal wind roses for non-calm hours, observations at Detroit Metropolitan Wayne County Airport (DTW). Calms (wind speeds less than 2 mph) are listed to the lower-left of each wind rose.

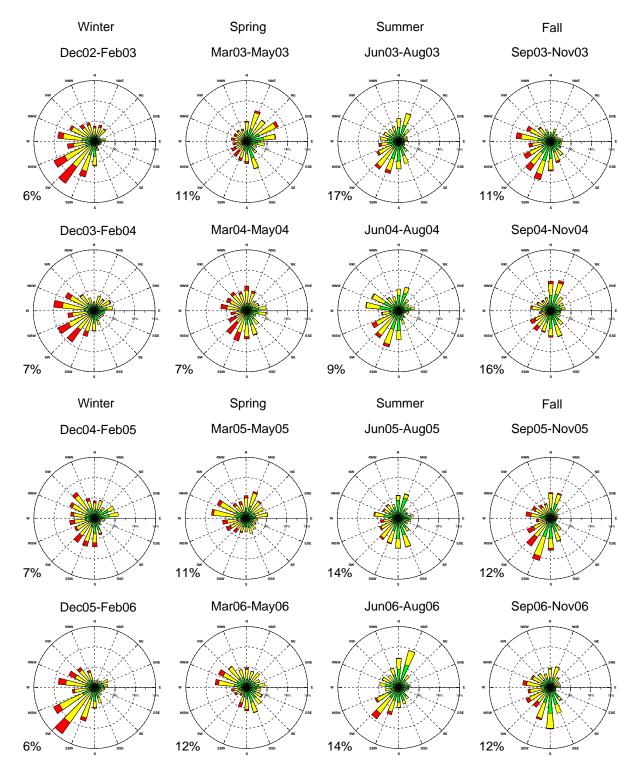


Figure B-1 (continued). Seasonal wind roses for non-calm hours, observations at Detroit Metropolitan Wayne County Airport (DTW). Calms (wind speeds less than 2 mph) are listed to the lower-left of each wind rose.

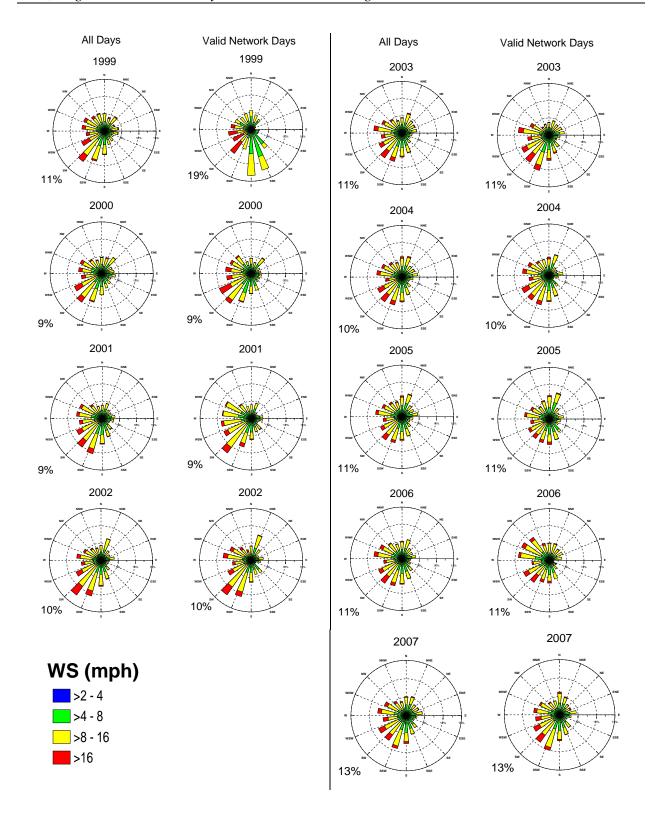


Figure B-2. Annual wind roses for non-calm hours, observations at Detroit Metropolitan Wayne County Airport (DTW) for all days and valid network days. Calms (wind speeds less than 2 mph) are listed to the lower-left of each wind rose.

APPENDIX C: SITE-SPECIFIC HIGH PM DAYS NOT OBSERVED THROUGHOUT THE NETWORK, 1999-2006

Dearborn		E 7 Mile		Linwood		Livonia		Luna Pier		Oak Park		Port Huron	SWHS		Wyandotte	
07/05/1999	(2)	05/12/2000	(1)	11/18/1999	(2)	12/17/2000	(1,2)	05/10/2001	(1,2)	11/25/2006	(1)	05/07/2005 (1,2)	09/27/1999	(2)	07/05/1999	(2)
01/03/1333	(2)	04/18/2004	(1,2)	12/08/1999	(2)	12/11/2000	(1,2)	03/12/2002	(2)	11/20/2000	(1)	07/18/2005 (1,2)	07/17/2000	(1,2)	05/06/2000	(1)
01/31/2000	(2)	01/28/2005	(1,2)	01/02/2000	(2)			04/02/2002	(1,2)			07/10/2000 (1,2)	02/09/2001	(1,2)	05/12/2000	(1)
02/24/2000	(1,2)	05/07/2005	(1,2)	02/24/2000	(1,2)			11/02/2003	(1)				05/10/2001	(1,2)	12/26/2000	(1,2)
03/07/2000	(1,2)	03/01/2003	(1,2)	02/24/2000	(1,2)			11/02/2003	(1)				08/23/2001	(1,2)	01/10/2001	(1,2)
05/06/2000	(1,2)												02/14/2003	(1)	01/10/2001	(1)
05/12/2000	(1)												08/25/2003	(1)	07/09/2001	(1)
06/08/2000	(1,2)												11/02/2003	(1)	08/23/2001	(1)
09/09/2000	(1)												12/29/2003	(1,2)	11/28/2002	(1,2)
12/23/2000	(1,2)												03/25/2004	(1,2)	03/19/2003	(1,2)
01/10/2001	(1,2)												04/30/2004	(1,2)	08/25/2003	(1)
01/28/2001	(1)												02/16/2006	(1)	02/06/2004	(1)
03/02/2001	(1,2)												12/13/2006	(1)	07/18/2005	(1)
08/23/2001	(1)												12/13/2000	(1)	12/06/2005	(1,2)
12/27/2001	(1,2)														12/21/2005	(1,2)
01/05/2002	(2)														02/10/2006	(1)
01/03/2002	(1,2)														02/10/2000	(1)
02/07/2002	(1,2)															
03/06/2002	(1)															
11/13/2002	(1,2)															
11/28/2002	(1,2)															
01/18/2003	(1,2)															
02/08/2003	(1,2)															
02/14/2003	(1)															
02/20/2003	(1,2)															
04/15/2003	(1,2)															
11/11/2003	(1,2)															
03/25/2004	(1,2)															
04/30/2004	(1)															
10/15/2004	(1)															
12/02/2004	(1,2)															
03/29/2005	(1,2)															
04/19/2005	(1,2)															
07/18/2005	(1,2)															
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12/06/2005	(1,2)															
02/16/2006	(1)															
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12/13/2006	(1,2)															
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^{*} Ann Arbor, FIA/Lafayette and Newberry excluded from the analysis.

^{*} No high PM days meeting these criteria for New Haven and Ypsilanti.

⁽¹⁾ denotes site concentration >30 $\mu g/m^3$ while Allen Park <25 $\mu g/m^3$.

⁽²⁾ denotes site concentration >30 μ g/m³ while network-wide base <25 μ g/m³ (limited to those days which are valid network days) (red bars in Figure 4-9).

APPENDIX D: CPF AND NWR PLOTS¹⁰

PM_{2.5} mass concentrations (both absolute and excess with respect to a reference site) were examined using Conditional Probability Function (CPF) plots and 1-D Nonparametric Wind Regression (NWR) plots. Kim and Hopke (2004) compared and contrasted the CPF and NWR methodologies and demonstrated that they provide complementary and largely consistent information. CPF plots provide insights into the bearing of emission sources impacting the monitoring site from a perspective that is not biased by the prevailing wind patterns. The plots address the question: "for the ensemble of hourly winds from a given direction, what fraction of these hours corresponds to high concentration days?". The threshold for defining a "high concentration day" is typically chosen as the top 25th percentile of the concentration distribution (either absolute concentrations or mass fractions). For example, Figure D-1(a) shows the CPF plot for the PMF-resolved copper factor at the Springfield site in Chicago. Nearly 70% of the hours with winds from 180°N coincided with a daily contribution in the top quartile of the concentration distribution for the copper factor contributions. This pattern suggests a copper source south of the monitoring site but it does not address the magnitude of the impacts which depend on source strength, dispersion characteristics and the frequency of winds from this direction.

One dimensional nonparametric wind regression estimates the expected value of concentration as a function of wind direction (Kim and Hopke, 2004). It is similar to a pollution rose but with more robust mathematical support. NWR results can be multiplied by the wind direction frequency distribution to determine the contribution of each wind direction to average PM mass. Figure D-1(b) shows the NWR plot for the PMF-resolved copper factor at the Springfield site in Chicago. Factor contributions (red line) for winds from 180°N are three times greater than the study-average factor contribution (pink circle). In aggregate the CPF and NWR plots provide insights into both the location of emission sources and their impacts.

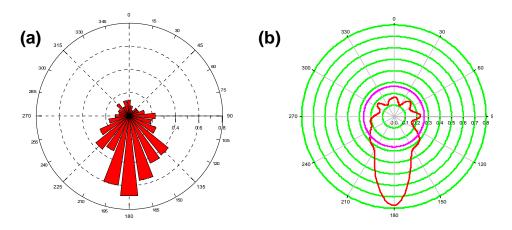
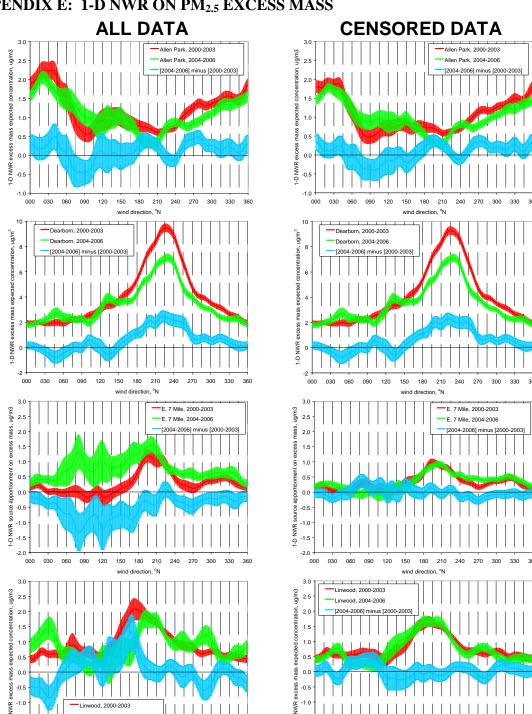


Figure D-1. Surface wind analyses for the PMF-resolved copper factor at the Springfield site in Chicago: (a) conditional probability function plot for the top quartile of absolute contributions; and (b) 1-D nonparametric wind regression plot (radial units in $\mu g/m^3$) where the red line denotes the observed contributions from each bearing and the pink circle denotes the study-average contribution.

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¹⁰ Adapted from Wade et al. (2008).



APPENDIX E: 1-D NWR ON PM_{2.5} EXCESS MASS

Figure E-1 (continued). 1-D NWR results for PM_{2.5} excess mass relative to the network-wide base for all network days (left column) and censored to remove extreme values as defined in footnote #12 (right column). The blue line is the change between 2004-2006 and 2000-2003 and the error bands are 95% confidence intervals.

060

180 210

150

060 090 120

[2004-2006] minus [2000-2003

150 180 210

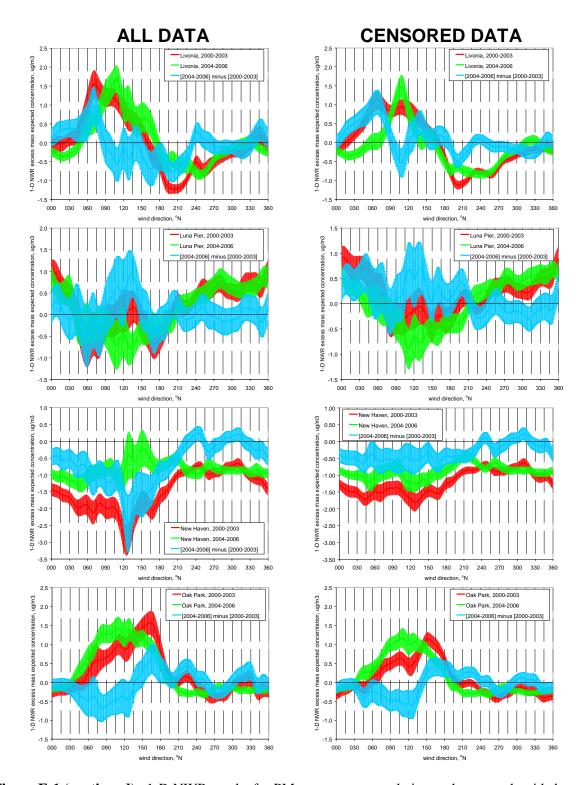


Figure E-1 (continued). 1-D NWR results for $PM_{2.5}$ excess mass relative to the network-wide base for all network days (left column) and censored to remove extreme values as defined in footnote #12 (right column). The blue line is the change between 2004-2006 and 2000-2003 and the error bands are 95% confidence intervals.

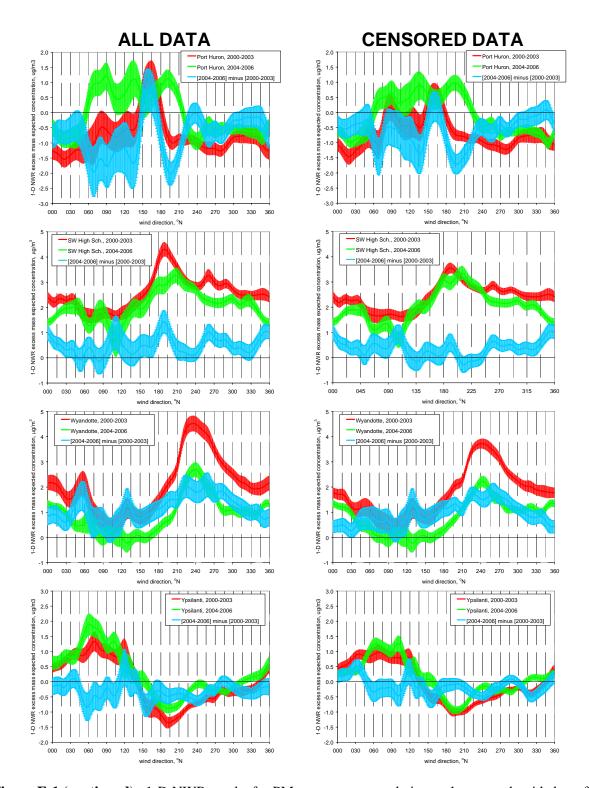


Figure E-1 (continued). 1-D NWR results for $PM_{2.5}$ excess mass relative to the network-wide base for all network days (left column) and censored to remove extreme values as defined in footnote #12 (right column). The blue line is the change between 2004-2006 and 2000-2003 and the error bands are 95% confidence intervals.