UW Biostatistics Working Paper Series

12-16-2013

Issues Related to Combining Multiple Speciated PM2.5 Data Sources in Spatio-Temporal Exposure Models for Epidemiology: The NPACT Case Study

Sun-Young Kim
University of Washington - Seattle Campus, puha0@u.washington.edu

Lianne Sheppard
University of Washington, sheppard@u.washington.edu

Timothy V. Larson
University of Washington-Seattle Campus, tlarson@u.washington.edu

Joel Kaufman
University of Washington, joelk@u.washington.edu

Sverre Vedal
University of Washington - Seattle Campus, svedal@u.washington.edu

INTRODUCTION

Evidence of the association between long-term exposure to ambient $PM_{2.5}$ and human health continues to accumulate (Dockery et al 1993; Miller et al 2007; Pope et al 1995; Puett et al 2009), and has spurred recent research to understand the role of specific $PM_{2.5}$ chemical components in adverse health effects (Ostro et al. 2010; Ostro et al. 2011 [erratum]; Vedal et al. 2013). Recent cohort studies have relied on predictions of long-term average $PM_{2.5}$ or $PM_{2.5}$ component concentrations at participant homes, based on models developed from monitoring data (Eeftens et al. 2012; Paciorek et al. 2009; Sampson et al. 2011; Szpiro et al. 2010; Yanosky et al. 2009). Parallel research in the statistics literature suggests that features of the monitoring data can affect the quality of the prediction models (Diggle et al. 2010; Gelfand et al. 2012) and the resulting health effect estimates (Szpiro et al. 2011; Szpiro and Paciorek 2013). Regulatory monitoring data collected and managed by governments are a common and useful resource for this application. For study of health effects of $PM_{2.5}$ chemical components in U.S., existing data are primarily from two networks: the U.S. Environmental Protection Agency (EPA) Chemical Speciation Network (CSN) and the Interagency Monitoring of Protected Visual Environment (IMPROVE) sponsored by EPA and other agencies (Bergen et al. 2013; Ostro et al. 2010; Pope et al. 1995). However, because these monitoring networks were designed for regulatory purposes, they may not be completely compatible with epidemiological applications.

The University of Washington National Particle Component and Toxicity (NPACT) study was designed to investigate the associations between long-term exposure to $PM_{2.5}$ chemical components and cardiovascular health based on the Multi-Ethnic Study of Atherosclerosis (MESA) cohort. This study conducted a dedicated and extensive monitoring campaign targeting the study cohort. In the original plan, the NPACT monitoring data were intended to be combined

with the regulatory monitoring data in models to produce predicted exposures, similar to the approach of combining all available data from multiple sources that has been used previously for predicting $PM_{2.5}$ (Paciorek et al. 2009; Sampson et al. 2011; Yanosky et al. 2009). In order to meet this objective, first we needed to assess various features of the $PM_{2.5}$ component data from the three sources in order to ensure basic compatibility in our models.

This paper compares and contrasts the compatibility of the two sets of regulatory monitoring network data with the NPACT monitoring data within the context of the NPACT study goals. In particular, we discuss the spatial coverage of exposure monitoring, filter analysis methods, and sampling protocols. NPACT analyses focused on four primary pollutants: EC, OC, silicon, and sulfur. Here we focus on EC and silicon to highlight similarities and differences in important features.

METHODS

Population

The NPACT study was based on the subjects who were originally recruited in MESA and consented to the Multi-Ethnic Study of Atherosclerosis and Air Pollution (MESA Air) study. The cohort includes approximately 7,000 participants residing in six metropolitan U.S. cities: Baltimore, Chicago, Los Angeles, Minneapolis-St. Paul, New York, and Winston-Salem (Bild et al. 2002; Kaufman et al. 2012).

Data

National Particle Component and Toxicity (NPACT) monitoring data

In order to characterize spatial variability of exposures across participant residences, the NPACT study measured $PM_{2.5}$ components by exploiting the MESA Air monitoring campaign (Vedal et al. 2013). MESA Air carried out an extensive exposure monitoring campaign focusing

on participant residence locations and the measurement of $PM_{2.5}$ mass and gaseous pollutant concentrations. The MESA Air monitoring campaign included three to seven fixed sites measuring pollutants in 2-week samples over multiple years, and approximately fifty rotating home-outdoor sites providing two to three 2-week samples in each city (Cohen et al. 2009). One fixed site was co-located with one CSN site in each of six cities. The two-week sampling schedule targeted the objective of obtaining estimates of long-term average exposure in the context of logistical and resource constraints. Whereas the NPACT-MESA Air sampling for trace elements was carried out over four years (August 2005 through August 2009), carbon data were collected for a shorter period (March 2007 through August 2008). Two-week samples for trace elements and carbon were collected on Teflon and quartz filters, respectively, placed into the Harvard Personal Environmental Monitors (HPEMs) with a 2.5 um cut size when operated at 1.8 L/min. Trace elements were quantified using X-ray Fluorescence (XRF) in the Cooper Environmental Services of Portland, Oregon. EC and OC were blank-corrected and quantified using the IMPROVE_A TOR method, which is currently employed in the EPA CSN network and comparable to the method used in the IMPROVE network, in the Sunset Laboratory Inc. of Tigard, Oregon. See Vedal et al (2013) for additional details.

Regulatory monitoring data

CSN and IMPROVE networks have collected $PM_{2.5}$ component measurements across the U.S. over 24-hour periods every 3rd or 6th day since 2000 and 1988, respectively (Hand et al. 2005; Rao et al. 2005; U.S. EPA 2004; U.S. EPA 2005a). From more than 300 monitoring sites in both networks, we selected 91 monitoring sites within 200 kilometers from the centers of the six MESA cities, and downloaded measurements collected between 1999 and 2009 from the EPA Air Quality System database. We chose 1999 as the starting year because it is one year

prior to the baseline screening of MESA participants in 2000. In CSN and IMPROVE, $PM_{2.5}$ components were sampled by various compliance samplers (U.S. EPA 1998). Two networks measured trace elements, including silicon, by XRF. In the CSN network, EC and OC were measured by NIOSH TOT and changed to IMPROVE_A TOR without blank correction for both methods. In contrast, IMPROVE has only used IMPROVE_A TOR with blank correction.

Data processing

We focus on silicon and EC in this paper because they represent groups of pollutants with contrasting spatial and temporal characteristics and their sampling and analysis protocols are distinct. See Vedal et al (2013) for data description and exploratory analysis results for the other components. To align with 2-week averages in NPACT, we computed corresponding averages of daily CSN/IMPROVE data for the 2-week periods centered on every other Wednesday. The two-week averages were log transformed after adding one to approximate a normal distribution. Whereas the units for EC were microgram per cubic meter, we used nanograms per cubic meter for silicon given the small observed values.

Three monitoring features affecting data comparability between networks

We focused on spatial coverage, filter analysis protocol, and sampling protocol as factors which may influence on data comparability between CSN, IMPROVE, and NPACT networks. First, monitoring sites in the CSN and IMPROVE networks are located far from each other and typically have only one or a few sites in a city, whereas the NPACT monitoring sties were densely located within each MESA city. The spatial sparseness of the regulatory monitoring data limits our ability to model $PM_{2.5}$ component concentrations over space (Lippmann. 2009). Second, analytical methods for fine particle elemental and organic carbon (EC and OC) differ within and between networks. In particular, CSN has historically used the National Institute for

Occupational Safety and Health (NIOSH) Total Optical Transmittance (TOT) method, whereas IMPROVE chose the IMPROVE_A Total Optical Reflectance (TOR) method. The two methods use different time/temperature analytical protocols to measure fractions of EC and OC on quartz filters. Data discrepancies resulting from these method differences have been documented (Chow et al. 2001; Malm et al. 2011). Consequently, EPA decided to change the laboratory method for CSN sites to the IMPROVE_A TOR method beginning in May 2007 (U.S. EPA. 2005b; U.S. EPA. 2006). NPACT also adopted the IMPROVE_A TOR method. Finally, the NPACT, CSN, and IMPROVE networks operated on different sampling schedules and use different sampling hardware. Whereas NPACT collected 2-week average samples, CSN/IMPROVE sites have collected daily average samples; these have been operated every 3rd day at most core CSN and all IMPROVE sites and every 6th day at supplemental CSN sites. Different sampling devices across networks may also contribute to data inconsistencies.

Exploratory data analysis for data comparability

Sparse coverage in urban space

We investigated the impact of spatial sparseness on the spatio-temporal prediction model by assessing spatial distributions of monitors and homogeneity of smoothed temporal trends across networks in each city. Temporal trends of pollutants can vary over space and across monitoring networks. When we rely on only a small number of monitoring locations, it is difficult to discern the presence of spatially-varying temporal trends. When temporal trends do not vary across locations, we can develop a simplified model that does not require many locations with long time-series of measurements.

Different filter analysis methods

We compared the two filter analysis methods for EC between CSN and IMPROVE networks as well as within the CSN network. All core CSN sites simultaneously changed from the NIOSH TOT to IMPROVE_A TOR method in May 2007, while the method change was phased in over time at supplemental CSN sites after that date. First, to confirm the inconsistent measurements between networks that have been reported in previous studies (Chow et al. 2001; Malm et al. 2011), we compared pairs of daily average measurements of EC at four co-located sites from CSN and IMPROVE networks between January 2000 and July 2007. For the within-network comparison, we investigated the consistency of daily averages of EC measured by the two methods at the six core CSN sites co-located with NPACT sites during the overlapping time period from May 2007 through July 2007.

Different sampling protocols

Given that NPACT collected 2-week average measurements and CSN and IMPROVE collected 24 hour samples every 3rd or 6th day, it was not clear whether these latter measurements could reliably estimate 2-week averages and temporal trends. Because there are a few core CSN sites in MESA cities and IMPROVE sites are mostly far away from city centers, most CSN/IMPROVE data available for NPACT were collected on a 6 day schedule. Thus we investigated the importance of sampling frequency by making within-site comparisons at four CSN sites co-located with NPACT fixed sites. Specifically, we compared the smoothed temporal trends of 2-week average silicon estimates using data obtained from every 3rd day samples vs. a reduced subset of every 6th day samples. The impact of differences in sampling hardware systems was compared using pairs of 2-week averages for EC and silicon from CSN and NPACT at six co-located sites. The EC analysis was restricted to the period after the filter analysis method change.

Exposure prediction model

The NPACT exposure prediction model aimed to predict 2-week average concentrations of $PM_{2.5}$ components at participant addresses by adopting the spatio-temporal modeling framework developed for the MESA Air study. While the NPACT-MESA Air monitoring design provided reasonable spatial coverage, logistical constraints resulted in data that were highly imbalanced: there were very few fixed site locations with long time series and a rich set of temporally sparse and unbalanced home-outdoor site measurements (Figure 2). The spatio-temporal model was designed to effectively utilize such highly imbalanced monitoring data. Applications of the spatio-temporal model for $PM_{2.5}$ and NOx in MESA Air have been described previously (Sampson et al. 2011; Szipro et al. 2010) and is available for implementation as the R package "SpatioTemporal" (Lindstrom et al. 2013a; Lindstrom et al. 2013b). In brief, this model assumes that 2-week average concentrations over space and time consist of spatially-correlated site-specific long-term means, site-specific temporal trends, and spatio-temporal residuals. Long-term means and temporal trends vary over space as characterized by geographical predictors and spatial correlation structures. Temporal trends are derived from a singular value decomposition of the data at sites with long time series. Space-time residuals are assumed to be temporally independent and spatially dependent.

Exploration of possible spatio-temporal modeling approaches

We explored three approaches to develop spatio-temporal prediction models for silicon and EC based on our experience developing the MESA Air spatio-temporal model for $PM_{2.5}$ (Sampson et al, 2011). First, we attempted to fit the full spatio-temporal model directly using all available $PM_{2.5}$ component data from regulatory and NPACT monitoring networks as in Sampson et al (2011) (Approach 1). In the $PM_{2.5}$ spatio-temporal modeling work, the regulatory

data were highly correlated with the MESA Air data. The regulatory $PM_{2.5}$ monitoring data were spatially dispersed and allowed a much larger spatial region to be modeled than was possible from MESA Air data alone. Given these characteristics, the long-time series of regulatory data provided most of the data for the trend function estimates, while the MESA Air monitoring data enhanced the model spatially. This rich data set allowed final models with multiple temporal trends and spatial models for the long-term mean, each temporal trend, and spatio-temporal residuals. If the $PM_{2.5}$ component data are insufficiently compatible to combine, NPACT data alone are not sufficiently rich to support the full spatio-temporal model. Thus we consider a second approach of fitting a simplified version of the spatio-temporal model assuming one temporal trend without spatial dependence structure (Approach 2). To assess the feasibility of the second modeling approach, we investigated whether a single trend is appropriate by comparing smoothed temporal trends across fixed sites with measurements at home sites in each city. Finally, we considered substituting temporal trend functions for $PM_{2.5}$ components with those estimated from other pollutant time series, such as $PM_{2.5}$ and NOx (Approach 3), because these pollutants have longer time series of data from a larger number of monitoring sites. We obtained $PM_{2.5}$ and NOx data measured at EPA monitoring sites located within 200 kilometers from the six MESA cities, computed 2-week averages, and compared temporal trends to those of EC and silicon in NPACT.

RESULTS

Table 1 summarizes important characteristics of the monitoring data across the NPACT-MESA Air, CSN, and IMPROVE networks. The table highlights three aspects of the regulatory monitoring data that may make it difficult to combine with those specially collected for NPACT

in one unified spatio-temporal model of $PM_{2.5}$ components: spatial sparseness, analysis method differences for carbon data, and different sampling protocols.

Data compatibility between CSN, IMPROVE and NPACT networks

Figure 1 displays maps of NPACT and regulatory monitoring site locations by city. Included are the locations of all selected regulatory monitoring sites from the CSN and IMPROVE networks along with the NPACT monitoring sites and MESA Air participant homes (locations jittered). As was seen by Sampson et al (2011), the regulatory monitoring sites cover a much larger spatial region than the area represented by the NPACT monitoring. Figure 2 shows a conceptual representation of the space-time sampling design for silicon and EC data in Los Angeles. Note that the longest time series of measurements are only available at the CSN and IMPROVE sites. While the specialized NPACT sampling provides data at many more locations, most of these have limited temporal representation.

Sparse coverage in urban space

The numbers of CSN and IMPROVE monitoring sites within 200 km ranged from six to twenty-seven and from one to eight, respectively, depending on city (Figure 1 and Table 2). However, when we restricted attention to the MESA city areas where most participants live in Figure 1, very few sites remained. Most IMPROVE sites are located in rural areas far away from participants making them less useful in prediction models. Differences between the urban vs. rural siting are manifest in their temporal trends. Figure 3 shows estimated smoothed temporal trends for the CSN and IMPROVE sites in Los Angeles. Temporal trends for EC at IMPROVE sites are strikingly different from those observed at CSN sites, representing the contrast between urban-focused CSN sites and rural-focused IMPROVE sites. There are also differences between the temporal trends for silicon across networks, but these are less striking. In the other five cities,

the temporal trends for EC are more or less heterogeneous depending on city, whereas those for silicon are more consistent across cities although there are some differences between sites in magnitude (Supplemental Figure 1).

Different filter analysis methods

Figure 4 shows that, as expected, daily average measurements of EC between CSN and IMPROVE at four co-located sites collected before the method change in May 2007 did not agree well. Figure 5 compares 24-hour average measurements of EC between the NIOSH TOT and IMPROVE_A TOR filter analysis methods for the two-month period of overlap from May 2007 to July 2007 at one CSN site in each MESA city. In Chicago and New York, the two methods showed constant differences and high correlations (estimates were 0.94 and 0.97, in part thanks to the larger variability between measurements in these cities). In contrast, the other cities displayed less systematic differences and had moderate correlations between 0.71 and 0.84.

Different sampling protocols

Table 2 gives tallies of CSN and IMPROVE sites by sampling schedule. Less than half of CSN sites (the core CSN sites) and all IMPROVE sites sampled $PM_{2.5}$ components every 3rd day, while more than half of CSN sites (the supplemental sites) sampled every 6th day. Figure 6 shows smoothed temporal trends for 2-week averages of silicon at four CSN sites co-located with NPACT fixed sites. The trends generally do not vary strongly by sampling schedule although a few local differences are evident. In the comparisons of sampling hardware, Figure 7 shows that 2-week averages of EC measured by the IMPROVE_A TOR method at CSN sites are not comparable to those at co-located NPACT fixed sites during the May 2007 through August 2008 period of overlap. In addition to NPACT measurements being consistently higher than CSN measurements in all cities, there are additional non-systematic differences between the two

networks. Time series plots with smoothed trends for the same data used in Figure 7 show local differences over time (Supplemental Figure 2). Silicon measurements were more comparable than EC but also manifested some non-systematic differences (Supplemental Figures 3 and 4).

Possible exposure modeling approaches

Approach 1: Full spatio-temporal model combining the CSN/IMPROVE and NPACT data

The monitoring data for $PM_{2.5}$ components were much more limited than those for other pollutants such as $PM_{2.5}$ (Supplemental Table 1). Because the $PM_{2.5}$ component data are not compatible across networks (see Figures 4 and 7 and Supplemental Figures 1, 2, and 3), we concluded that we cannot fit the full spatio-temporal modeling on the combined CSN, IMPROVE, and NPACT data.

Approach 2: Simplified spatio-temporal model based on the NPACT data only

If we estimate temporal trends of $PM_{2.5}$ components based only on the NPACT monitoring data, the time series of EC from fixed site monitors is short (approximately one-and-a-half years) (Figure 2). Figure 8 displays one temporal trend estimated using all fixed sites along with measurements across home-outdoor sites over time in Los Angeles and Chicago. Although one homogenous temporal trend in each city is a strong assumption and the variation in the home sites reflects spatial as well as temporal variability, the smooth trends generally capture the temporal variability across home sites.

Approach 3: Full spatio-temporal model using another pollutant

$PM_{2.5}$ and NOx have been collected since 1990's at a large number of sites relative to other pollutants in the regulatory monitoring network. However, the estimated temporal trends for $PM_{2.5}$ and NOx were not sufficiently consistent with the temporal trends estimated from the

$PM_{2.5}$ components. As an example, Figure 9 shows dramatically different temporal trends for the $PM_{2.5}$ components compared to $PM_{2.5}$ and NOx in the Minneapolis and St. Paul area.

DISCUSSION

We explored the features of regulatory and NPACT monitoring data for EC and silicon relevant to our goal of combining all available data for constructing spatio-temporal models to investigate health effects of long-term exposures to $PM_{2.5}$ chemical components in the NPACT study. The small number of regulatory monitoring sites deployed in urban areas limited the amount of data available for modeling in the NPACT study areas. In addition, we found insufficient between- and within-network consistency to combine CSN, IMPROVE and NPACT data in one spatio-temporal model. These findings led us to conclude that we should develop spatio-temporal models using NPACT monitoring data only. Given the limited space-time data in NPACT, the resulting spatio-temporal models needed to be simplified compared with those used in other MESA Air applications.

We found inconsistency between NPACT and regulatory monitoring networks for measurements of both EC and silicon despite the fact that they both used the same filter analysis methods. This inconsistency seems to be due to different sampling protocols such as sampling frequency and equipment. EC was measured for a sampling period of 2 weeks in NPACT versus daily sampling in the regulatory networks. NPACT EC tended to be higher than CSN, while OC measurements were lower than the corresponding CSN measurements (Vedal et al. 2013). It is possible that the more reactive OC components, which would have been contributed to OC measurements in the lab filter analysis if stayed on filters, oxidized over 2 weeks, thus resulting in decreased OC and increased EC concentrations in NPACT. In addition to the sampling schedule, other differences in carbon sampling between NPACT and CSN/IMPROVE shown in

Table 1 could have also affected inconsistencies in the data. NPACT used the HPEM sampler with lower pump flow rate and the blank correction protocol based on backup quartz filters. However, our observation that there was good agreement between total carbon measurements in the CSN and NPACT networks (Vedal et al. 2013) suggests that the inconsistency of EC and OC measures is more likely driven by the split of EC and OC rather than the sampling and blank correction protocols. Differences between silicon measurements could be driven by silicon grease of the HPEM sampler in NPACT. If grease contamination occurred during filter handling, silicon on grease might have reached the filters and resulted in increased silicon concentrations. However, grease contamination usually appears as a very large spike in the contaminated sample compared to other samples, which was not observed in our data. Another possible explanation may be local dust plumes. The co-located NPACT monitor was placed a few meters away from the CSN monitor. The consistent $PM_{2.5}$ and sulfur concentrations at these co-located sites indicate that the Teflon filters of the two monitors generally sampled the same fine particles. However, local dust plume gradients could exist resulting in concentration differences between the two monitoring locations.

Some studies have developed calibration models to allow EC and OC data from the CSN and IMPROVE networks to be combined. White et al (2008) and Malm et al (2011) used elemental, organic, and total carbon data at about 10 co-located urban CSN and IMPROVE sites in 2005 and 2006 to estimate relationships of EC and OC between the two networks. Their IMPROVE-adjusted EC and OC at CSN sties was highly correlated with EC and OC at co-located IMPROVE sites. However, these calibration methods were based on fairly limited data collected at relatively small number of co-located sites during a short time period. More research is needed to determine whether these calibration methods can be generalized to other areas or

years. In addition, we observed different relationships by city (Figures 7 and 8), suggesting area-specific calibration models may be needed. The NPACT study also developed city-specific calibration models to estimate CSN-adjusted EC and OC based on the consistency of total carbon between CSN and NPACT as well as EC in CSN and black carbon in NPACT (Vedal et al. 2013). However, the city specific data were limited, particularly for New York, and this limitation might have affected the calibration approach.

Based on the work described in this paper, we concluded that the use of only NPACT monitoring data without including regulatory data is the preferred choice for our spatio-temporal prediction model for EC and silicon. In contrast, other published studies of health effects of long-term average $PM_{2.5}$ component concentrations relied exclusively on regulatory monitoring data. Ostro et al (2010) used the CSN data and assigned $PM_{2.5}$ components at the nearest monitors to participant homes in California. Bergen et al (2013) used CSN and IMPROVE data to build a universal kriging model across the U.S. Both studies used long-term averages and developed pure spatial models thus avoiding the need to model the temporal data structure. In order to take advantage of the extensive project-based monitoring campaign focused on the target cohort, the NPACT options were to either use the NPACT data alone or to combine the NPACT data with regulatory monitoring data. We found, however, these data obtained from different sources were not sufficiently comparable to allow us to use all available data in our spatio-temporal model.

We focused on EC and silicon out of the four components in the NPACT study in order to illustrate the features of monitoring networks which result in data inconsistency and then affect exposure prediction modeling approaches. OC performed similarly to EC; in contrast sulfur measurements agreed well across networks. Thus for sulfur it may be reasonable to combine all available data and develop a more sophisticated exposure model. Future work is

needed to examine the data consistency for other $PM_{2.5}$ components for use in exposure prediction models.

It is questionable whether the existing spatio-temporal prediction modeling approaches for $PM_{2.5}$ can be transferred directly to modeling $PM_{2.5}$ components. Recent cohort studies have developed advanced exposure prediction approaches in space-time frameworks for estimating $PM_{2.5}$ long-term average concentrations (Paciorek et al.2009; Sampson et al. 2011; Yanosky et al. 2009). These advanced modeling approaches were developed using $PM_{2.5}$ monitoring data collected by governments and project-based campaigns. In comparison with the component data, the regulatory $PM_{2.5}$ monitoring data have been collected under consistent protocols over a relatively long time period and across a fairly dense set of monitoring locations in U.S. (U.S. EPA 2004). Furthermore, there is reasonable agreement between these regulatory monitoring data and the data collected by MESA Air (Cohen et al. 2009). Thus Sampson et al (2011) were able to combine EPA Federal Reference Method and MESA Air monitoring data in single city-specific predictive models for $PM_{2.5}$. In contrast, the sampling periods and coverage areas for $PM_{2.5}$ component data are limited and, as shown in this paper, there are important discrepancies between measurements collected under different protocols. We have shown the importance of understanding the $PM_{2.5}$ component data prior to developing exposure prediction models.

Given widespread scientific interest in understanding the associations between long-term air pollution and health for multiple pollutants, an undertaking that is feasible only after development and application of appropriate exposure prediction models, it is crucial that we also acquire sufficient understanding of monitoring network features which may in turn affect exposure predictions and the resulting health effect estimates. Methodological research has shown that features of the underlying exposure surface, exposure assessment design, and

approaches to exposure modeling may all impact health effect estimates (Gryparis et al. 2009; Kim et al. 2009; Szpiro et al. 2011; Szpiro and Paciorek 2013). This study adds monitoring data from multiple sources as another feature affecting exposure modeling for estimating health effects. We demonstrated the importance of evaluating the consistency of the monitoring data from the perspective of how they will be incorporated into the exposure models.

CONCLUSIONS

U.S. regulatory monitoring data for $PM_{2.5}$ components measured at CSN and IMPROVE sites are a potentially rich data resource to be used solely or jointly with project-based monitoring data for the study of health effects of $PM_{2.5}$ components. However, the sparse spatial coverage of these networks and differences across networks in their analysis and sampling protocols for some $PM_{2.5}$ components can affect their utility in epidemiological studies, particularly for inclusion in spatio-temporal prediction models for $PM_{2.5}$ component concentrations. Future studies of long-term concentrations of $PM_{2.5}$ components and health need to assess exposure data characteristics before developing exposure prediction models.

REFERENCES

Bergen S. Szpiro AA, Sheppard L, Sampson PD, Kim SY, Richards M, et al. 2013. A national prediction model for $PM_{2.5}$ component exposures and measurement error-corrected health effect inference. Environ Health Perspect (in press).

Bild DE, Bluemke DA, Burke GL, Detrano R, Diez Roux AV, Folsom AR, et al. 2002. Multi-ethnic study of atherosclerosis: objectives and design. Am J Epidemiol 156:871-881.

Chow JC, Watson JG , Crow D, Lowenthal DH, Merrifield T. 2001. Comparison of IMPROVE and NIOSH carbon measurements. Aerosol Sci and Technol 34:23-34.

Cohen MA, Adar SD, Allen RW, Avol E, Curl CL, Gould T, et al. 2009. Approach to estimating participant pollutant exposures in the Multi-Ethnic Study of Atherosclerosis and Air Pollution (MESA Air). Environ Sci Technol 43:4687-4693.

Dockery DW, Pope 3rd CA, Xu X, Spengler JD, Ware JH, Fay ME, et al. 1993. An association between air pollution and mortality in six U.S. cities. N Engl J Med. 329:1753-1759.

Diggle PJ, Menezes R, Su T. 2010. Geostatistical inference under preferential sampling. J R Stat Soc Ser C Appl Stat 59:191-232.

Eeftens M, Beelen R, de Hoogh K, Bellander T, Cesaroni G, Cirach M, et al. 2012. Development of land use regression models for PM(2.5), PM(2.5) absorbance, PM(10) and PM(coarse) in 20 European study areas; results of the ESCAPE project. Environ Sci Technol 46:11195-11205.

Gelfand AE, Sahu SK, Holland DM. 2012. On the effect of preferential sampling in spatial prediction. Environmetrics 23:565-578.

Gryparis A, Paciorek CJ, Zeka A, Schwartz J, Coull BA. 2009. Measurement error caused by spatial misalignment in environmental epidemiology. Biostatistics 10:258-274.

Hand JL, Copeland SA, Day DE, Dillner AM, Indresand H, Malm WC, et al. 2011. Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States: Report V. June (http://vista.cira.colostate.edu/improve/publications/Reports/2011/PDF/Cover_TOC.pdf).

Kaufman JD, Adar SD, Allen RW, Barr RG, Budoff MJ, Burke GL, et al. 2012. Prospective Study of Particulate Air Pollution Exposures, Subclinical Atherosclerosis, and Clinical Cardiovascular Disease: The Multi-Ethnic Study of Atherosclerosis and Air Pollution (MESA Air). Am J Epidemiol 176:825-837.

Kim SY, Sheppard L, H Kim. 2009. Health effects of long-term air pollution: influence of exposure prediction methods. Epidemiology 20:442-450.

Lindstrom J, Szpiro AA, Sampson PD, Sheppard L, Oron A, Richards M, et al. 2013a. A flexible spatio-temporal model for air pollution with spatial and spatio-temporal covariates. Environ Ecol Stat (in press).

Lindstrom J, Szpiro AA, Sampson PD, Bergen S, Oron A. 2013b. SpatioTemporal: An R package for spatio-temporal modelling of air-pollution. J stat softw (in press) (http://cran.r-project.org/web/packages/SpatioTemporal/index.html).

Lippmann M. 2009. Semi-continuous speciation analyses for ambient air particulate matter: An urgent need for health effect studies. J Exp Sci Environ Epidemiol 19:235-247.

Malm WC, Schichtel BA, Pitchford ML. 2011. Uncertainties in $PM_{2.5}$ gravimetric and speciation measurements and what we can learn from them. J. Air & Waste Manage Assoc 61:1131-1149.

Miller KA, Siscovick DS, Sheppard L, Shepherd K, Sullivan JH, Anderson GL, et al. 2007. Long-term exposure to air pollution and incidence of cardiovascular events in women. N Engl J Med 356:447-458.

Ostro B, Lipsett M, Reynolds P, Goldberg D, Hertz A, Garcia C, et al. 2010. Assessing Long-Term Exposure in the California Teachers Study. Environ Health Perspect 118:363-369.

Ostro B, Burnett RT, Shin H, Hughes E, Garcia C, Henderson KD, et al. 2011. Assessing Long-Term Exposure in the California Teachers Study. Environ Health Perspect 119:A242–A243 Erratum.

Paciorek CJ, Yanosky JD, Puett RC, Laden F, Suh HH. 2009. Practical large-scale spatio-temporal modeling of particulate matter concentrations. Ann Appl Stat 3:370–397.

Pope 3rd CA, Thun MJ, Namboodiri MM, Dockery DW, Evans JS, Speizer FE, et al. 1995. Particulate air pollution as a predictor of mortality in a prospective study of U.S. adults. Am J Respir Crit Car Med 151:669-674.

Puett RC, Hart JE, Yanosky JD, Paciorek C, Schwartz J, Suh H, et al. 2009. Chronic fine and coarse particulate exposure, mortality, and coronary heart disease in the Nurses' Health Study. Environ Health Perspect 117:1697-1701.

Rao V, Frank N, Rush A, Dimmick F. 2003. Chemical Speciation of $PM_{2.5}$ in Urban and Rural Areas in National Air Quality and Emissions Trends Report (http://www.epa.gov/air/airtrends/aqtrnd03/pdfs/2_chemspecofpm25.pdf).

Sampson PD, Szpiro AA, Sheppard L, Lindström J, Kaufman JD. 2011. Pragmatic Estimation of a Spatio-Temporal Air Quality Model with Irregular Monitoring Data. Atmos Environ 45:6593-6606.

Szpiro AA, Sampson PD, Sheppard L, Lumley T, Adar SD, Kaufman JD. 2010. Predicting intraurban variation in air pollution concentrations with complex spatio-temporal interactions. Environmetrics 21:606-631.

Szpiro AA, Paciorek CJ, Sheppard L. 2011. Does more accurate exposure prediction improve health effect estimates? Epidemiology 22:680-685.

Szpiro AA and Paciorek CJ. 2013. Measurement error in two-stage analyses, with application to air pollution epidemiology. Environmetrics (in press).

U.S. EPA. 1998. Guideline on Speciated Particulate Monitoring. Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency. RTP, NC (http://epa.gov/ttnamti1/files/ambient/pm25/spec/drispec.pdf).

U.S. EPA. 2004. Air quality criteria for particulate matter (Report No. EPA 600/P-99/002aF-bF): Volume 1. U.S. Environmental Protection Agency. Washington, DC.

U.S. EPA. 2005a. Office of Inspector General Report, EPA needs to direct more attention, efforts, and funding to enhance its speciation monitoring program for measuring fine particulate matter (Report No. 2005-P-00004). U.S. Environmental Protection Agency. Washington, DC.

U.S. EPA. 2005b. EPA's final draft national ambient air monitoring strategy: An advisory by the ambient air monitoring and methods subcommittee of the EPA Clean Air Scientific Advisory Committee (Report No. EPA-SAB-CASAC-05-006). U.S. Environmental Protection Agency. Washington, DC (http://www.epa.gov/sab/pdf/casac-05-006.pdf).

U.S. EPA. 2006. Modification of carbon procedures in the speciation network: Overview and frequently asked questions (FAQs) (http://www.epa.gov/ttn/amtic/files/ambient/pm25/spec/faqcarbon.pdf).

Vedal S, Kim SY, Miller KA, Fox JR, Bergen S, Gould T, et al. NPACT epidemiologic study of components of fine particulate matter and cardiovascular disease in the MESA and WHI-OS cohorts. Research Report 178. Health Effects Institute, Boston, MA (in press).

White WH. 2008. Interim Approach for Relating the Two Datasets: Discussion and Action Plan Development. Presented at the IMPROVE–CSN PM Monitoring Workshop, Davis, CA (vista.cira.colostate.edu/improve/Publications/Workshops/Carbon_Jan2008/IMP-CSN_White.ppt).

Yanosky JD, Paciorek CJ, Suh HH. 2009. Predicting chronic fine and coarse particulate exposures using spatiotemporal models for the Northeastern and Midwestern United States. Environ Health Perspect 117:522-529.

Table 1. Major contrasting characteristics between NPACT, CSN, and IMPROVE networks

Characteristics		Network		
		NPACT-MESA Air	CSN	IMPROVE
Sampling design	Location of sites	Urban	Urban	Rural
	Spatial density in MESA city areas	Dense	Sparse	Sparse
	Monitoring period	2005-2009	Since 1999	Since 1987
	Sampling schedule	2-week average	24-hour average: 1 in 3 or 6 day	24-hour average: 1 in 3 day
Filter analysis method	Analysis method for elements	XRF*	XRF	XRF
	Analysis method for carbon*	IMPROVE_A TOR*	NIOSH TOT IMPROVE_A TOR+	IMPROVE_A TOR
	Blank correction using backup quartz filter	Yes	No	Yes
Sampling protocol	Sampler type for elements	HPEMs	Met One and others	IMPROVE
	Sampler type for carbon	HPEMs	Met One and 4 others URG+	IMPROVE
	Pump flow rate	1.8 L/min	6.7 ~ 16.7 L/min 22.8 L/min+	22.7 L/min

* XRF analysis was performed in Cooper Environmental Services of Portland, Oregon and IMPROVE_A TOR analysis was performed in Sunset laboratory Inc. of Tigard, Oregon

+ New carbon sampling and analysis protocols have been implemented from core CSN sites since May 2007

Table 2. Number of sites with long-term monitoring data available within 200 km of six MESA city areas between 1999 and 2009

Area		Regulatory				NPACT/MESA Air	
		CSN			IMPROVE	Fixed	Home
	Total*	Total	3-day	6-day	Total (3-day)	Total (14 day avg)	Total (14 day avg)
Los Angeles	21 (141)+	6	3	2	8	7	120
Chicago	23 (166)	15	4	11	1	7	143
Minneapolis-St. Paul	11 (145)	6	2	4	1	4	134
Baltimore#	37 (125)	27	8	19	5	5	88
New York#	31 (152)	25	14	11	3	3	121
Winston-Salem	19 (137)	12	2	10	3	4	118

* Co-located sites are counted once in the grand total and also as appropriate in each network category

+ Number of sites excluding NPACT-MESA Air home sites (Number of sites including home sites)

13 sites appear in both Baltimore and New York due to overlap of regions: 12 CSN and 1 IMPROVE

Figure Legends

Figure 1. Locations of CSN, IMPROVE, and NPACT monitoring sites for $PM_{2.5}$ components within 200 km from city centers in six MESA city areas.

Figure 2. Temporal and spatial sampling for silicon and EC by CSN, IMPROVE, and NPACT monitors in Los Angeles.

Figure 3. Temporal trends of 2-week averages of silicon and EC measured by CSN and IMPROVE sites in Los Angeles from 1999 to 2009.

Figure 4. Scatter plots of every 3rd day measurements of EC between CSN and IMPROVE from January 2000 through July 2007 at four co-located in six MESA city areas.

Figure 5. Scatter plots of every 3rd day measurements of EC between pre- and post- filter analysis method change for the overlapping 2 months from May 2007 through July 2007 at six CSN sites co-located with NPACT sites in six MESA city areas.

Figure 6. Time series plots of 2-week averages of silicon between every 3rd day and 6th day measurements at four CSN site co-located with four NPACT fixed sites in Chicago, Minneapolis-St. Paul, Baltimore, and New York from 1999 to 2009.

Figure 7. Scatter plots of 2-week averages of EC for the overlapping period from May 2007 through August 2008 between co-located CSN and NPACT fixed sites in each of six MESA city areas.

Figure 8.Time series of 2-week averages of silicon and EC across home-outdoor sites along with one temporal trend estimated using NPACT fixed sites in Los Angeles and Chicago.

Figure 9. Temporal trends of 2-week averages of silicon (top) and EC (bottom) across NPACT fixed sites along with trends of $PM_{2.5}$ and NOx across EPA sites in the Minneapolis-St. Paul area.

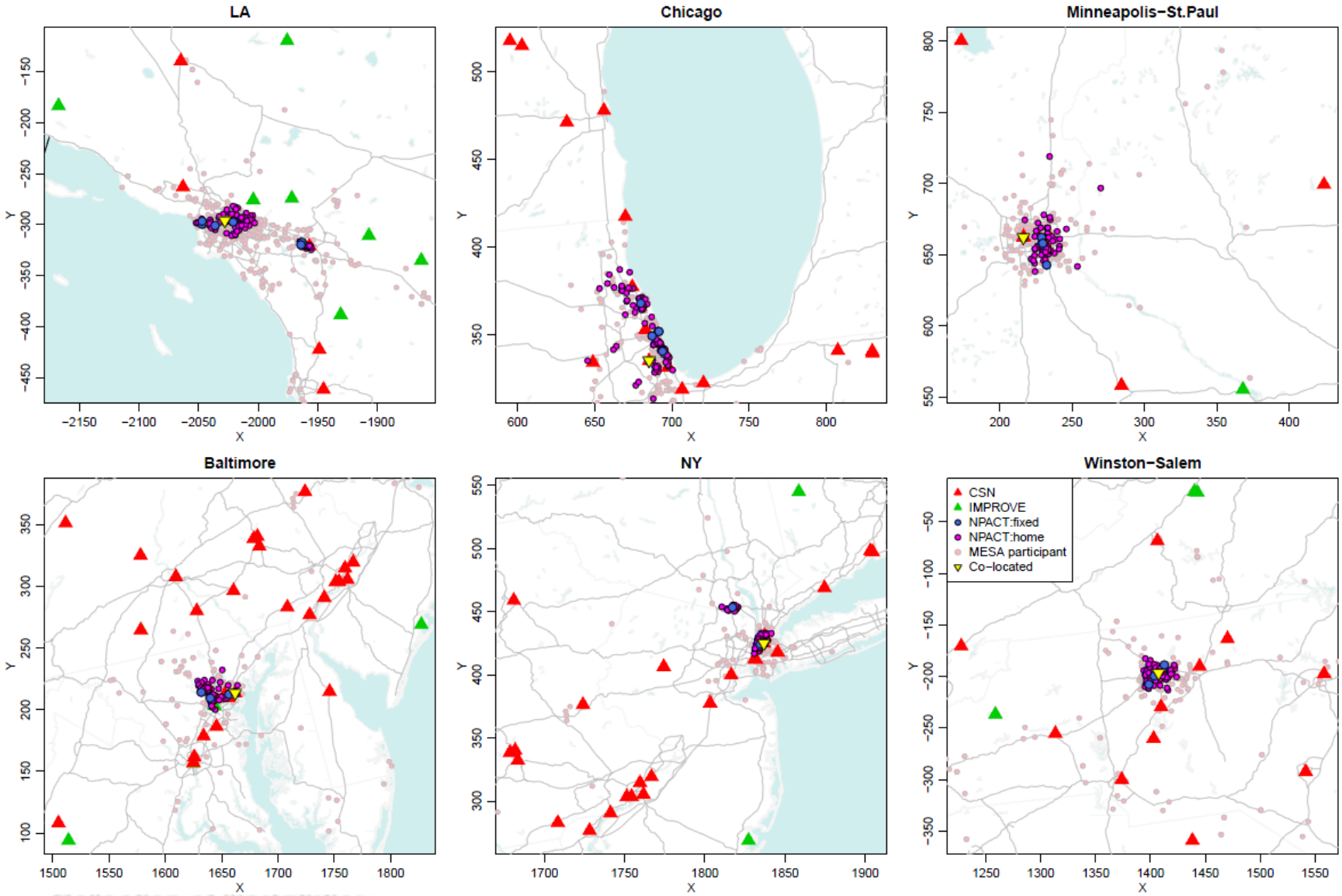


Figure 1. Locations of CSN, IMPROVE, and NPACT monitoring sites for $PM_{2.5}$ components within 200 km from city centers in six MESA city areas

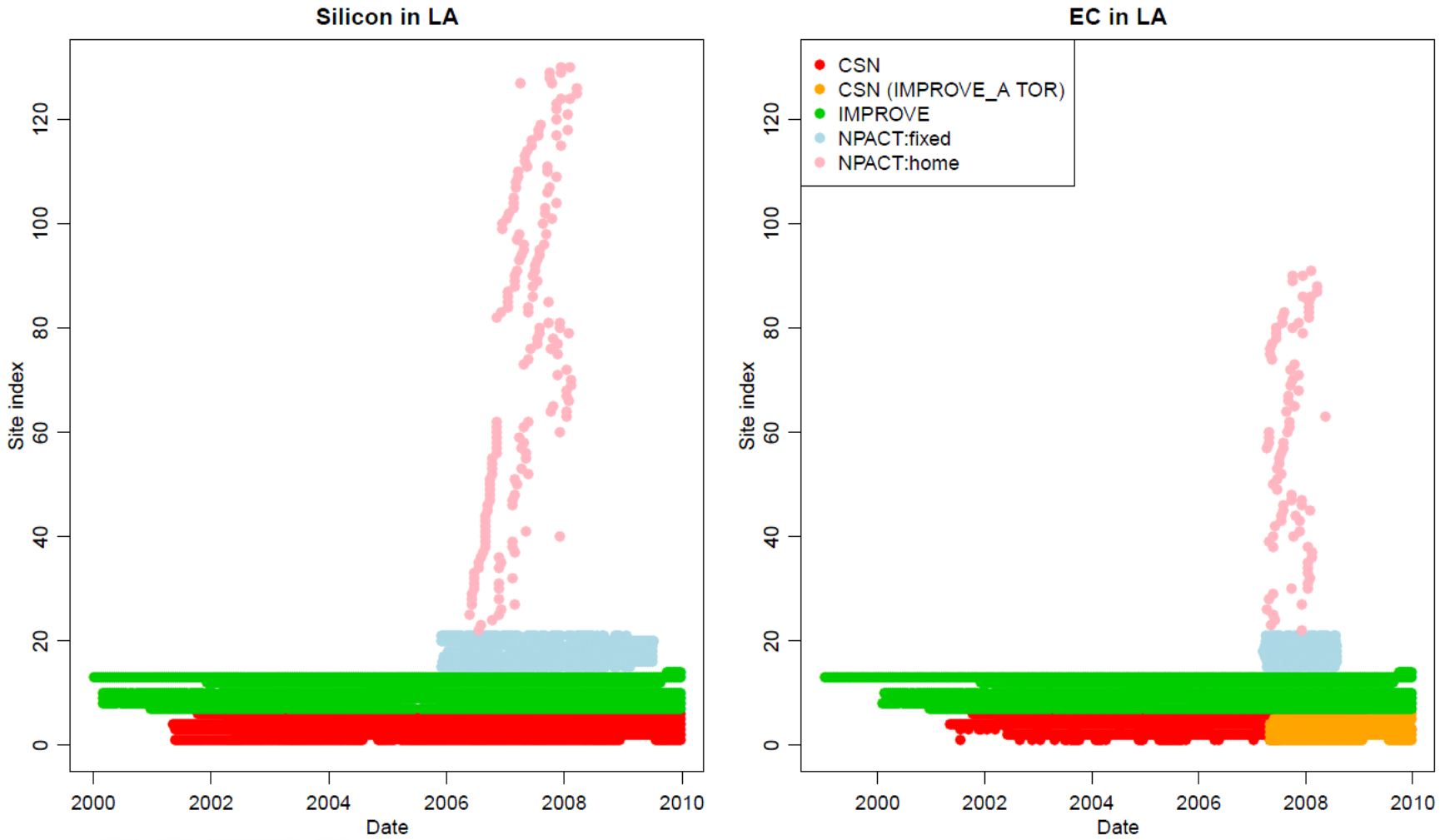


Figure 2. Temporal and spatial sampling for silicon and EC by CSN, IMPROVE, and NPACT monitors in Los Angeles

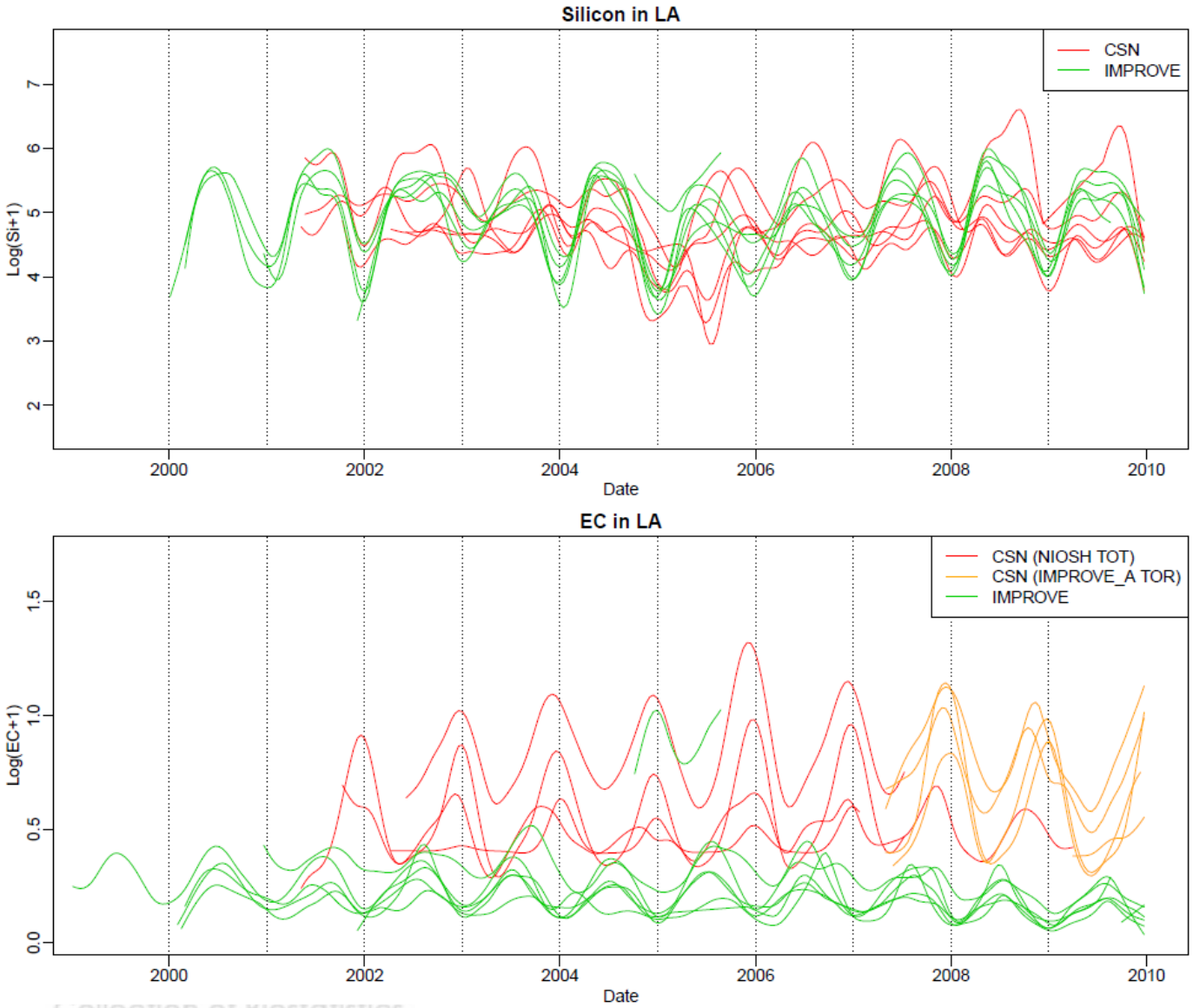


Figure 3. Temporal trends of 2-week averages of silicon and EC measured by CSN and IMPROVE sites in Los Angeles from 1999 to 2009

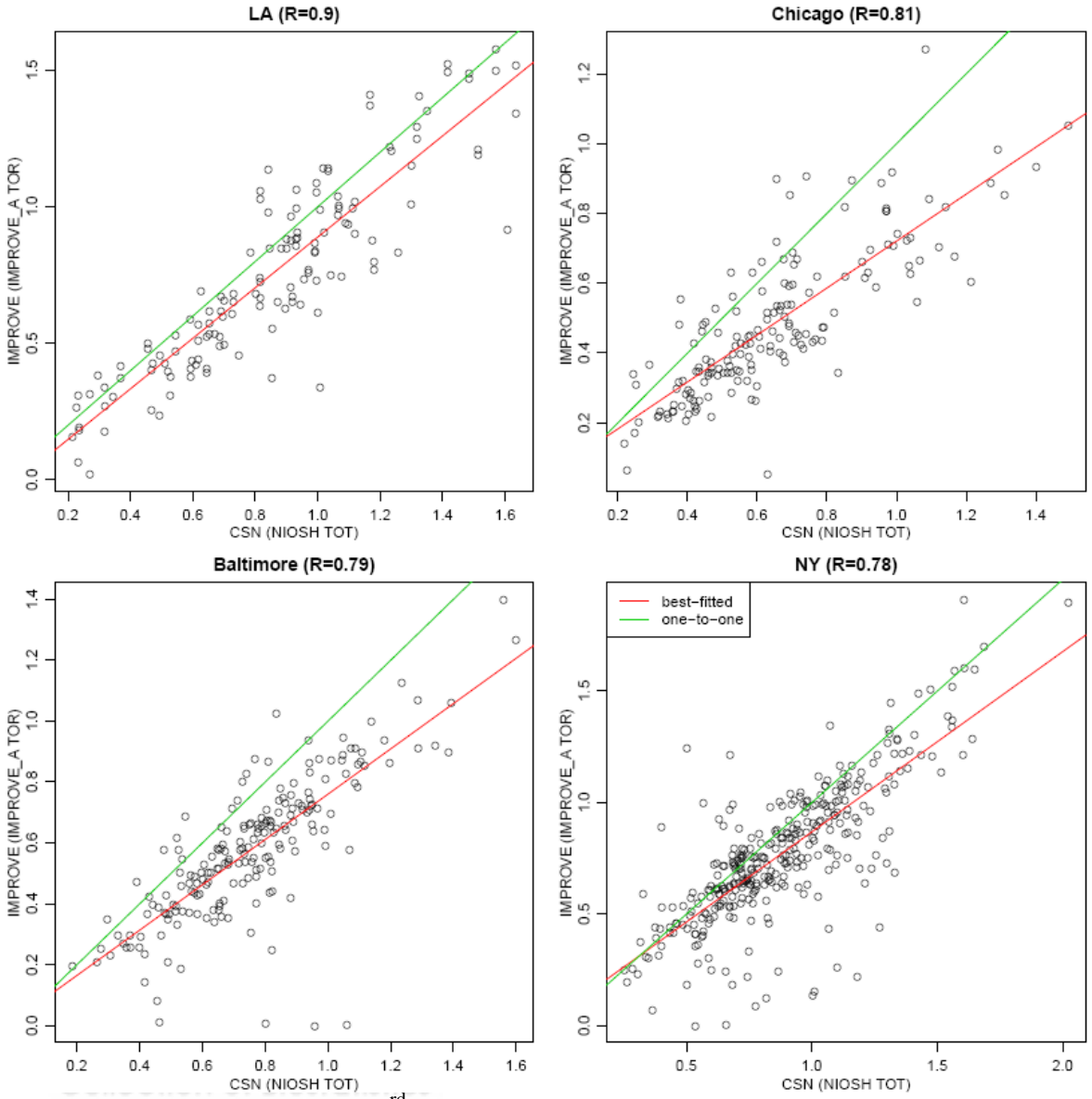


Figure 4. Scatter plots of every 3rd day measurements of EC between CSN and IMPROVE from January 2000 through July 2007 at four co-located in six MESA city areas

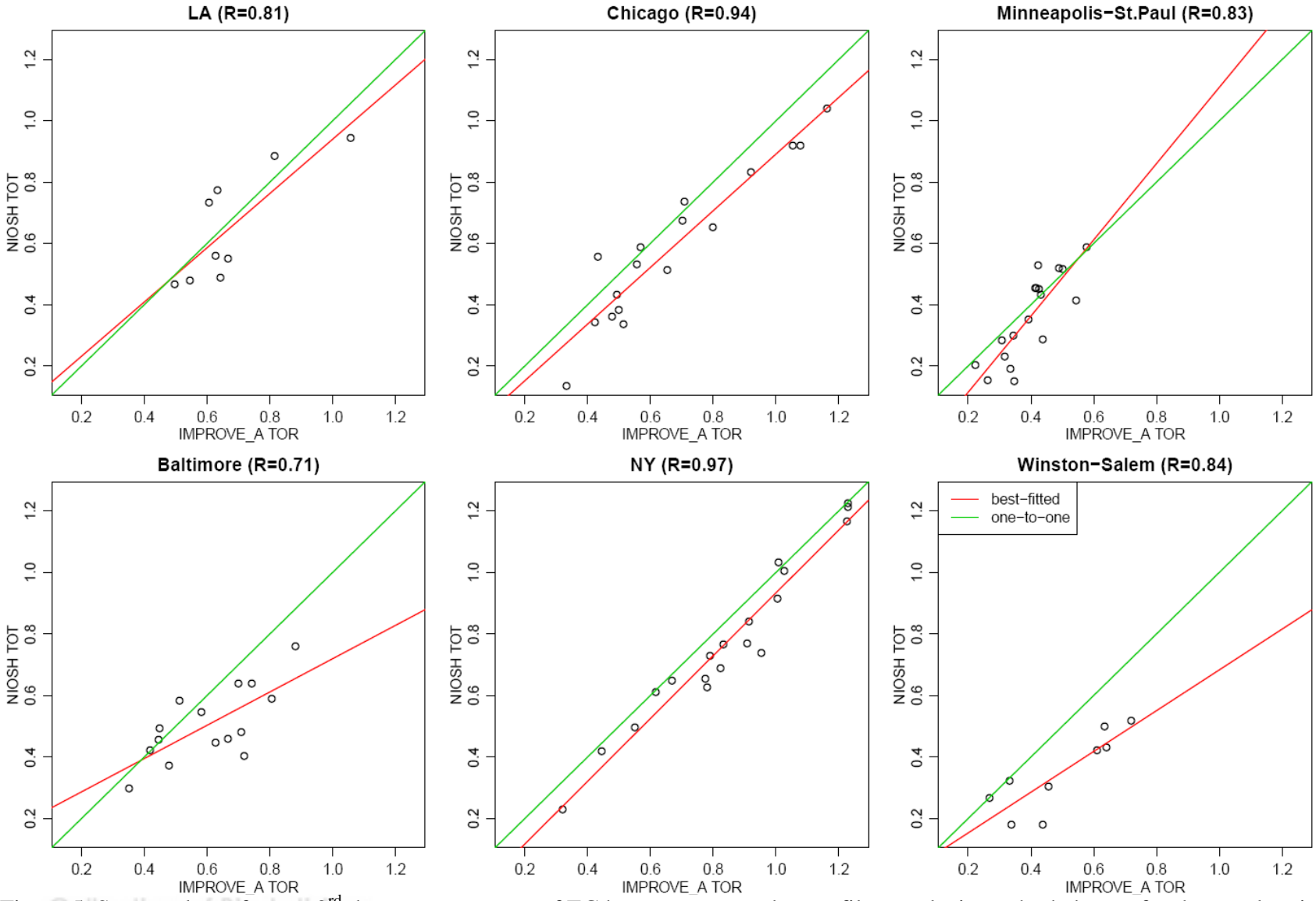


Figure 5. Scatter plots of every 3rd day measurements of EC between pre- and post- filter analysis method change for the overlapping 2 months from May 2007 through July 2007 at six CSN sites co-located with NPACT sites in six MESA city areas

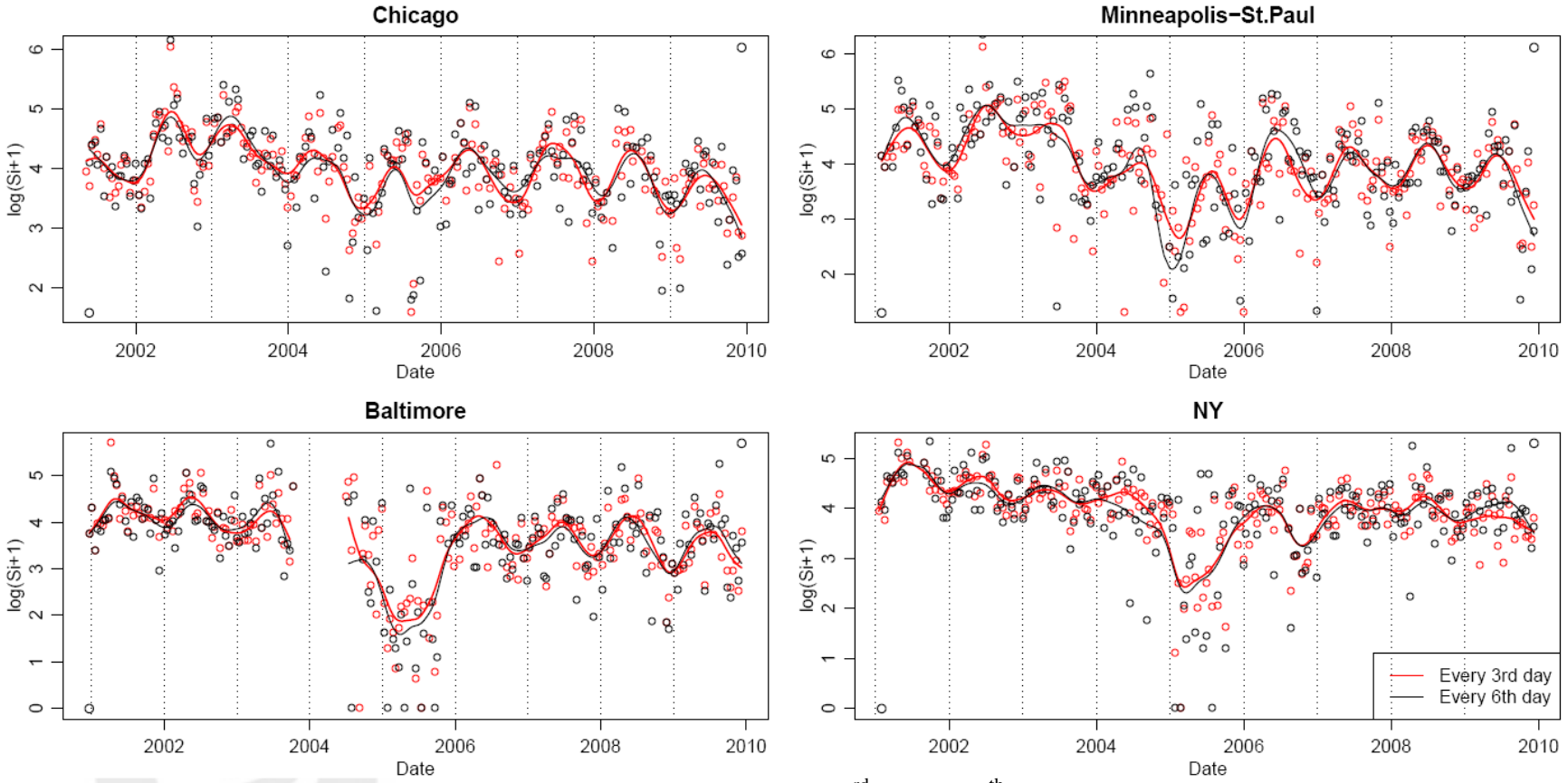


Figure 6. Time series plots of 2-week averages of silicon between every 3rd day and 6th day measurements at four CSN site co-located with four NPACT fixed sites in Chicago, Minneapolis-St. Paul, Baltimore, and New York from 1999 to 2009

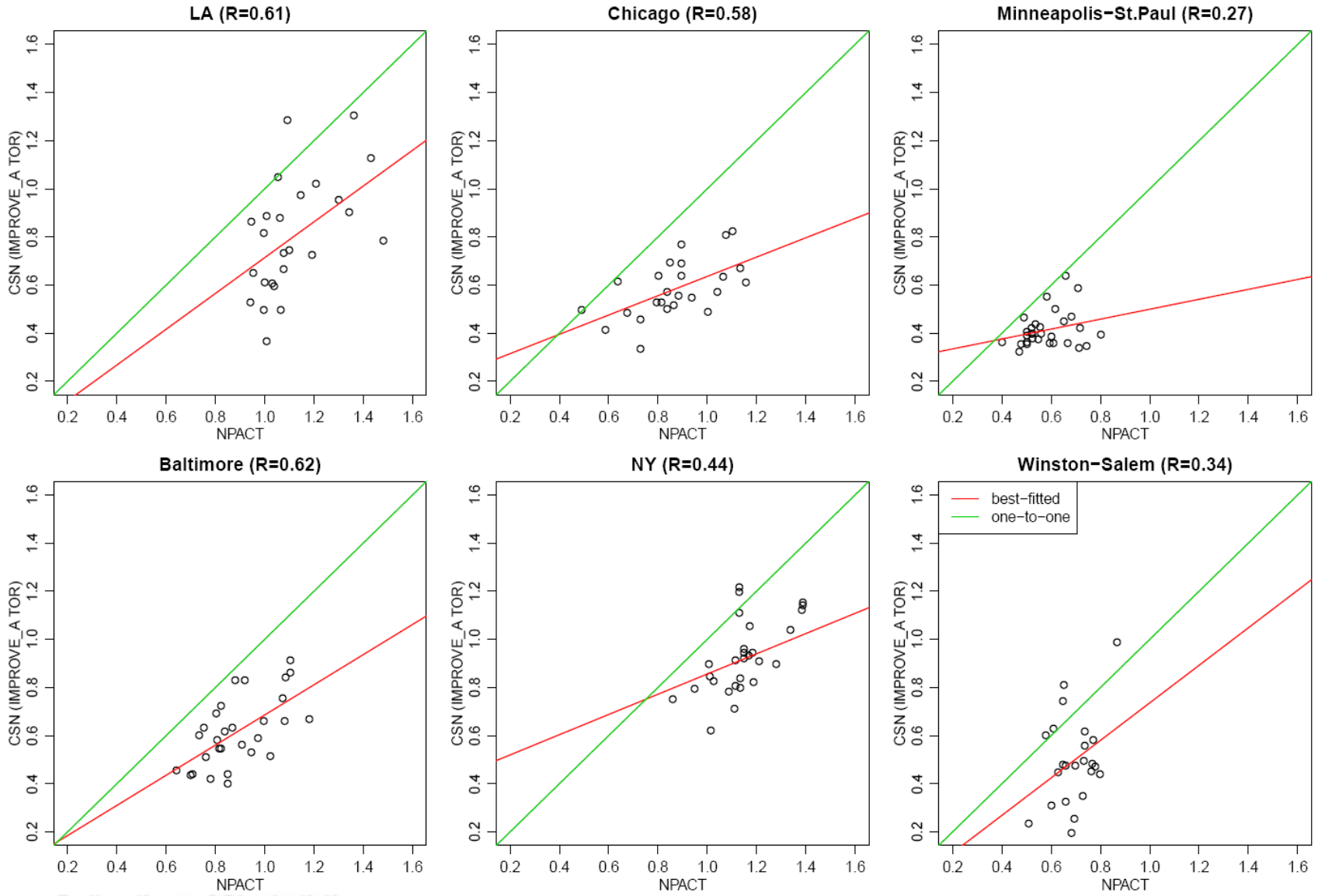


Figure 7. Scatter plots of 2-week averages of EC for the overlapping period from May 2007 through August 2008 between co-located CSN and NPACT fixed sites in each of six MESA city areas

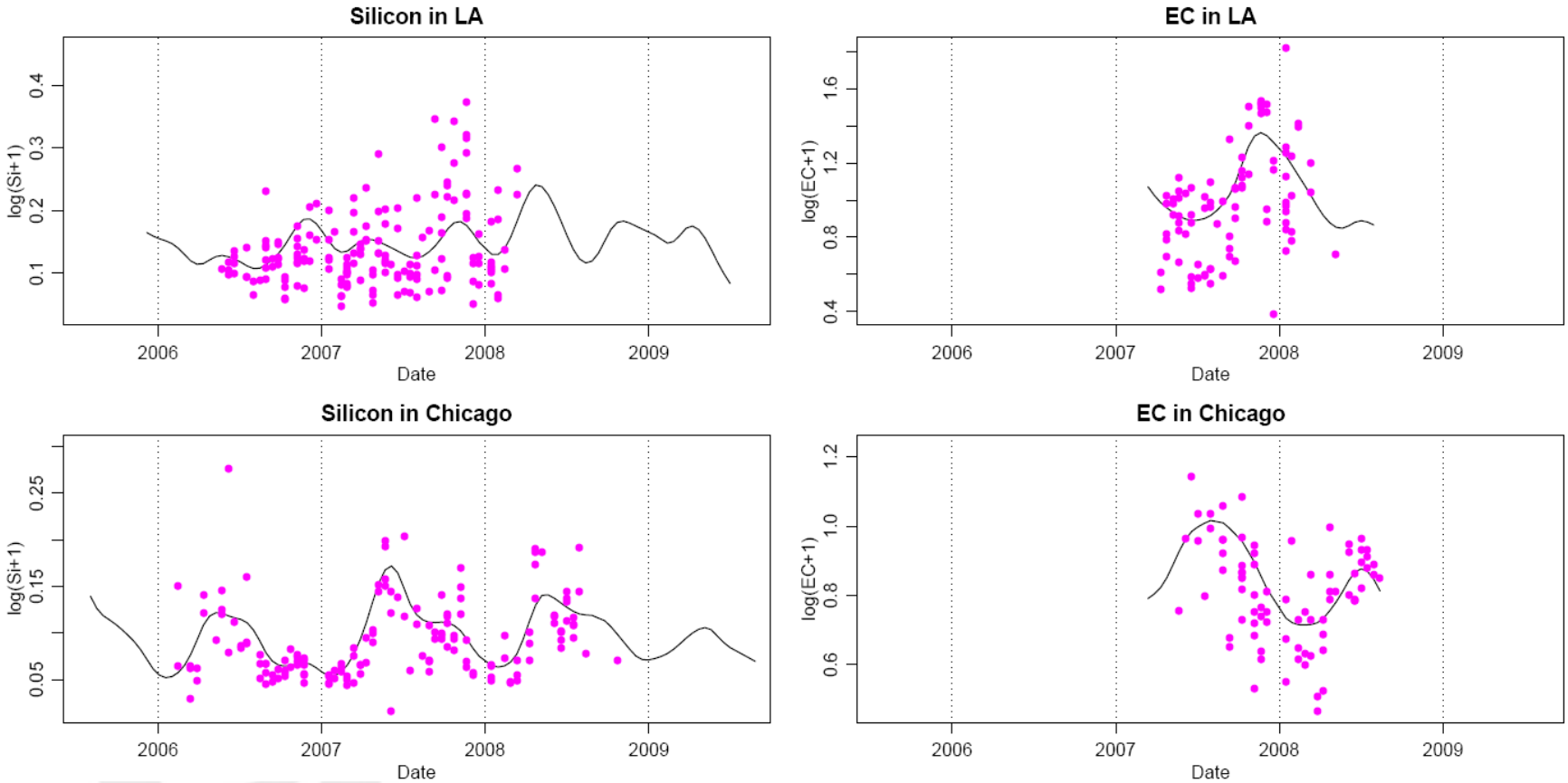


Figure 8.Time series of 2-week averages of silicon and EC across home-outdoor sites along with one temporal trend estimated using NPACT fixed sites in Los Angeles and Chicago

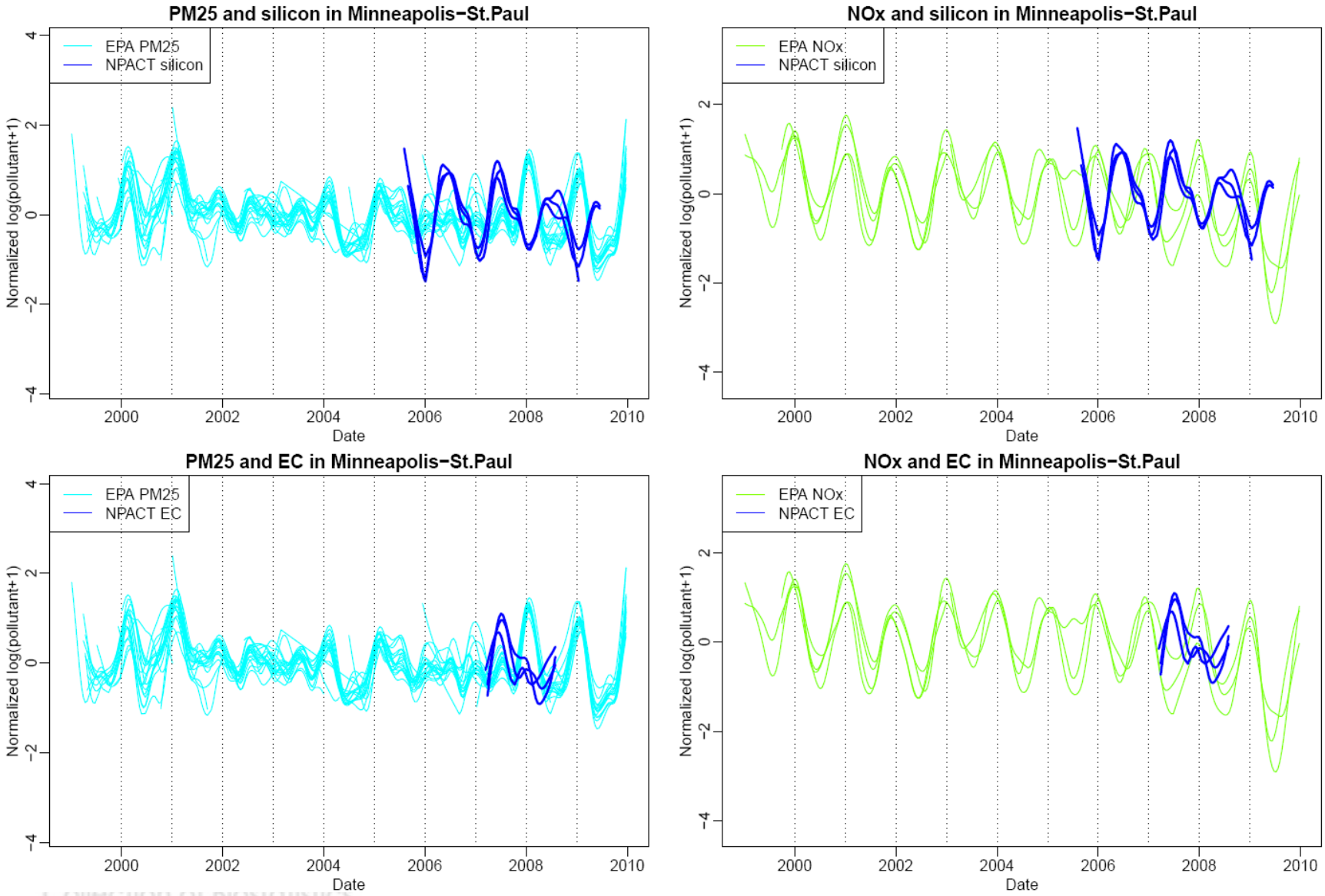


Figure 9. Temporal trends of 2-week averages of silicon (top) and EC (bottom) across NPACT fixed sites along with trends of $PM_{2.5}$ and NOx across EPA sites in the Minneapolis-St. Paul area

SUPPLEMENTAL MATERIALS

Issues Related to Combining Multiple Speciated $PM_{2.5}$ Data Sources in Spatio-Temporal Exposure Models for Epidemiology: The NPACT Case Study

Sun-Young Kim[1], Lianne Sheppard[1,2], Timothy V. Larson[3], Joel D. Kaufman[1,4,5], Sverre Vedal[1]

[1]Department of Environmental and Occupational Health Sciences, University of Washington, WA, USA

[2]Department of Biostatistics, University of Washington, WA, USA

[3]Department of Civil and Environmental Engineering, University of Washington, WA, USA

[4]Department of Medicine, University of Washington, WA, USA

[5]Department of Epidemiology, University of Washington, WA, USA

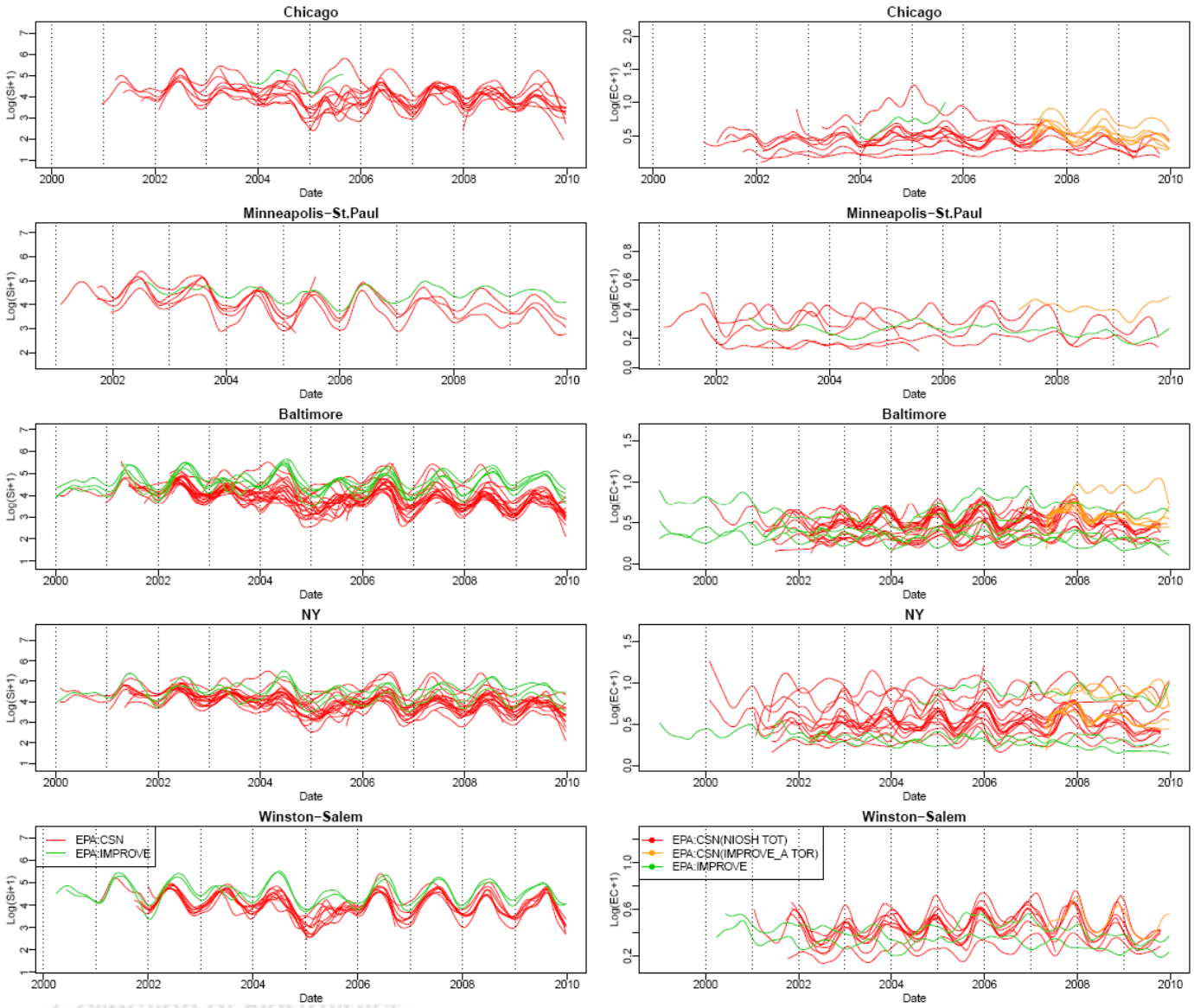
Contents

Supplemental Table 1. Number of sites and 2-week average observations for silicon, EC, and $PM_{2.5}$ used for spatio-temporal exposure prediction models by monitor type and city in EPA and NPACT monitoring networks from 1999 through 2009

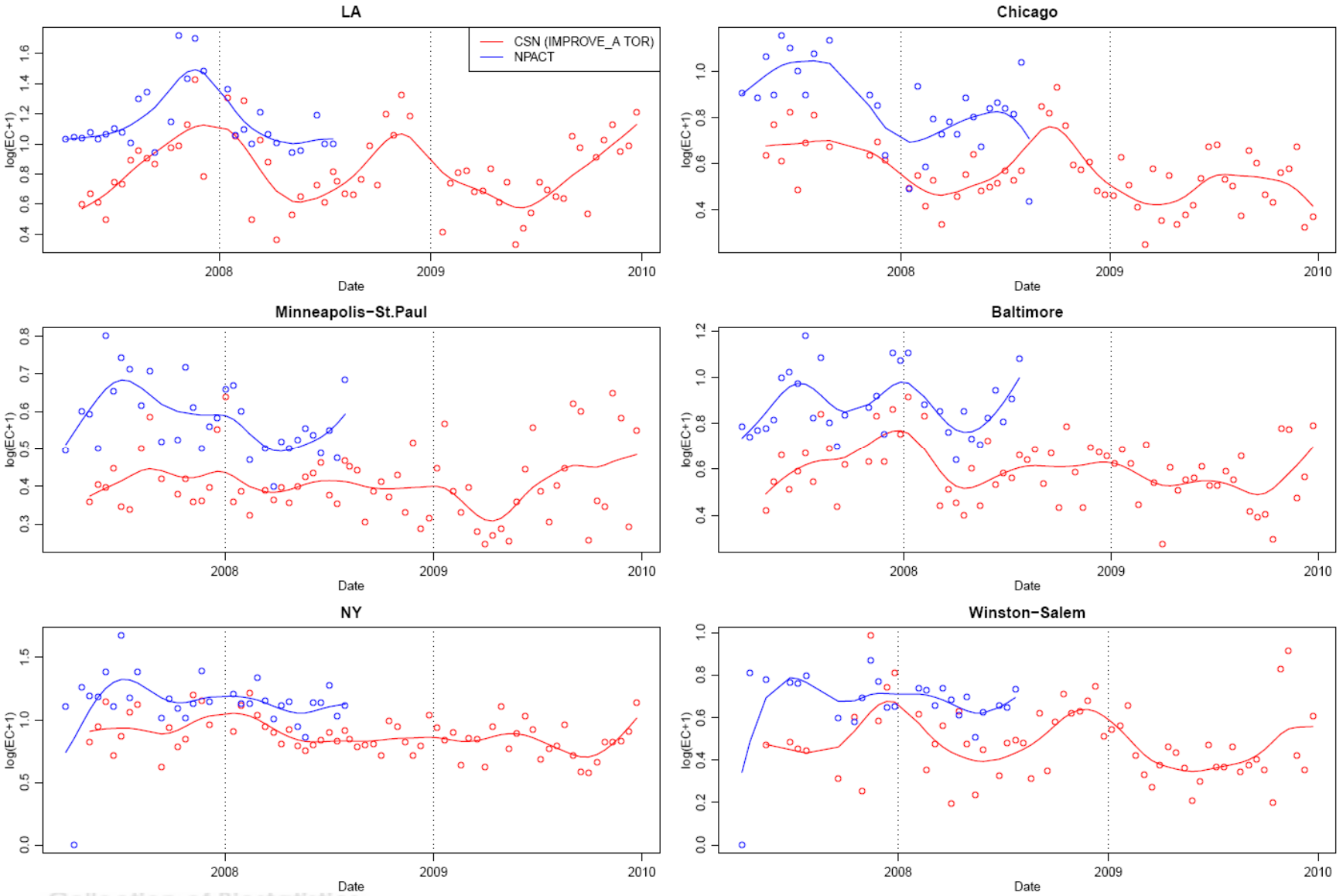
City	Type	Silicon			EC			$PM_{2.5}$*		
		# sites	Min # obs./sites	Max # obs./sites	# sites	Min # obs./sites	Max # obs./sites	# sites	Min # obs./sites	Max # obs./sites
Los Angeles	EPA	—	—		—	—		24	177	363
	Fixed+	7	73	81	7	74	84	7	19	28
	Home+	113	1	2	116	1	2	120	1	2
Chicago	EPA	—	—		—	—		44	133	364
	Fixed	7	6	87	7	89	99	7	7	34
	Home	99	1	3	99	1	3	113	1	2
Minneapolis-St. Paul	EPA	—	—		—	—		41	88	365
	Fixed	3	79	86	3	79	86	3	27	29
	Home	104	1	3	104	1	3	129	1	2
Baltimore	EPA	—	—		—	—		39	205	365
	Fixed	5	18	85	5	18	86	5	14	33
	Home	86	1	3	87	1	3	87	1	1
New York	EPA	—	—		—	—		45	111	365
	Fixed	3	49	83	3	53	87	3	31	32
	Home	107	1	3	107	1	3	119	1	2
Winston-Salem	EPA	—	—		—	—		29	116	365
	Fixed	4	79	92	4	82	92	4	18	35
	Home	92	1	3	92	1	3	117	1	2

* Obtained from Sampson et al 2011 which used the $PM_{2.5}$ data from 2000 through 2006

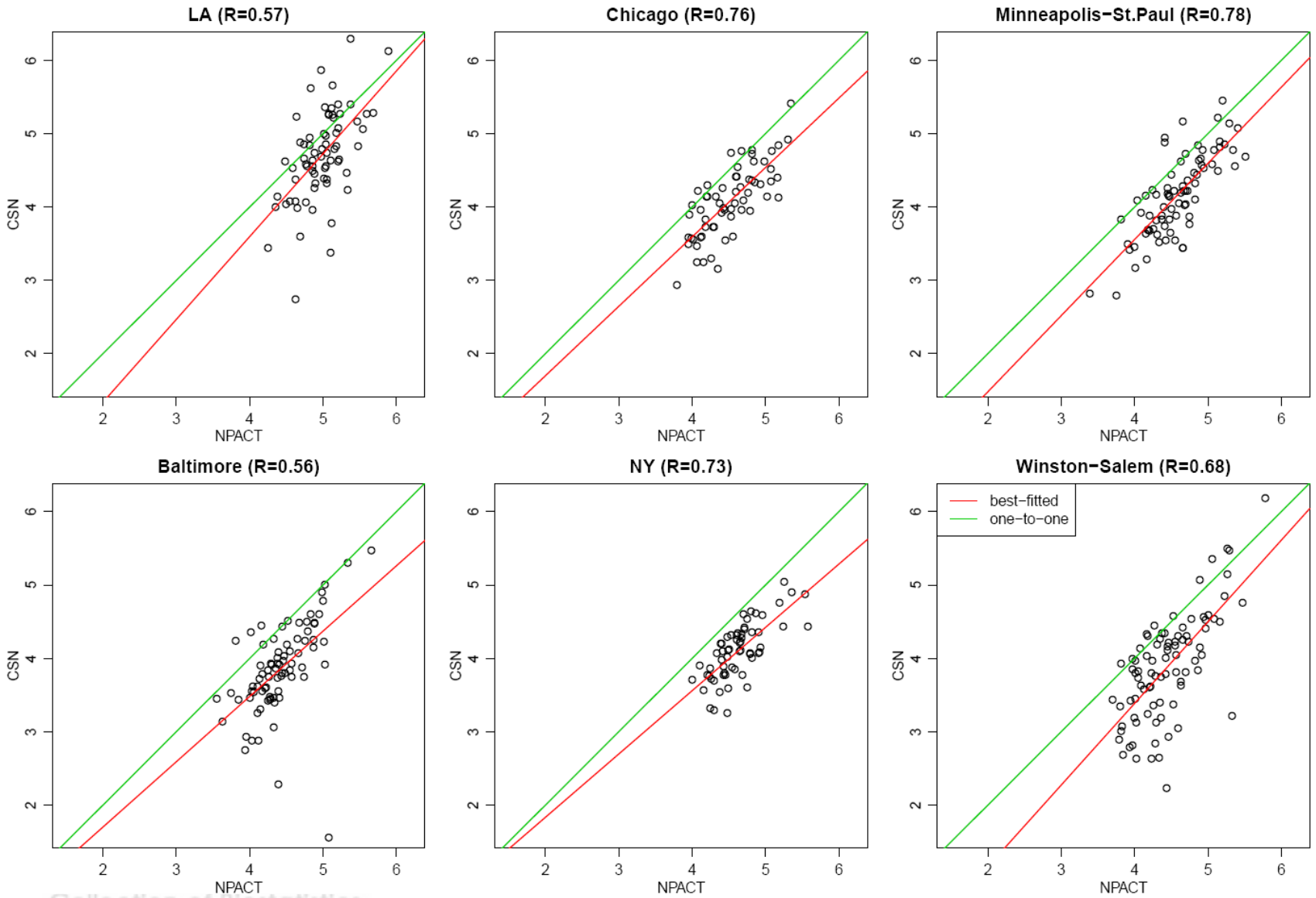
+ NPACT monitoring sites



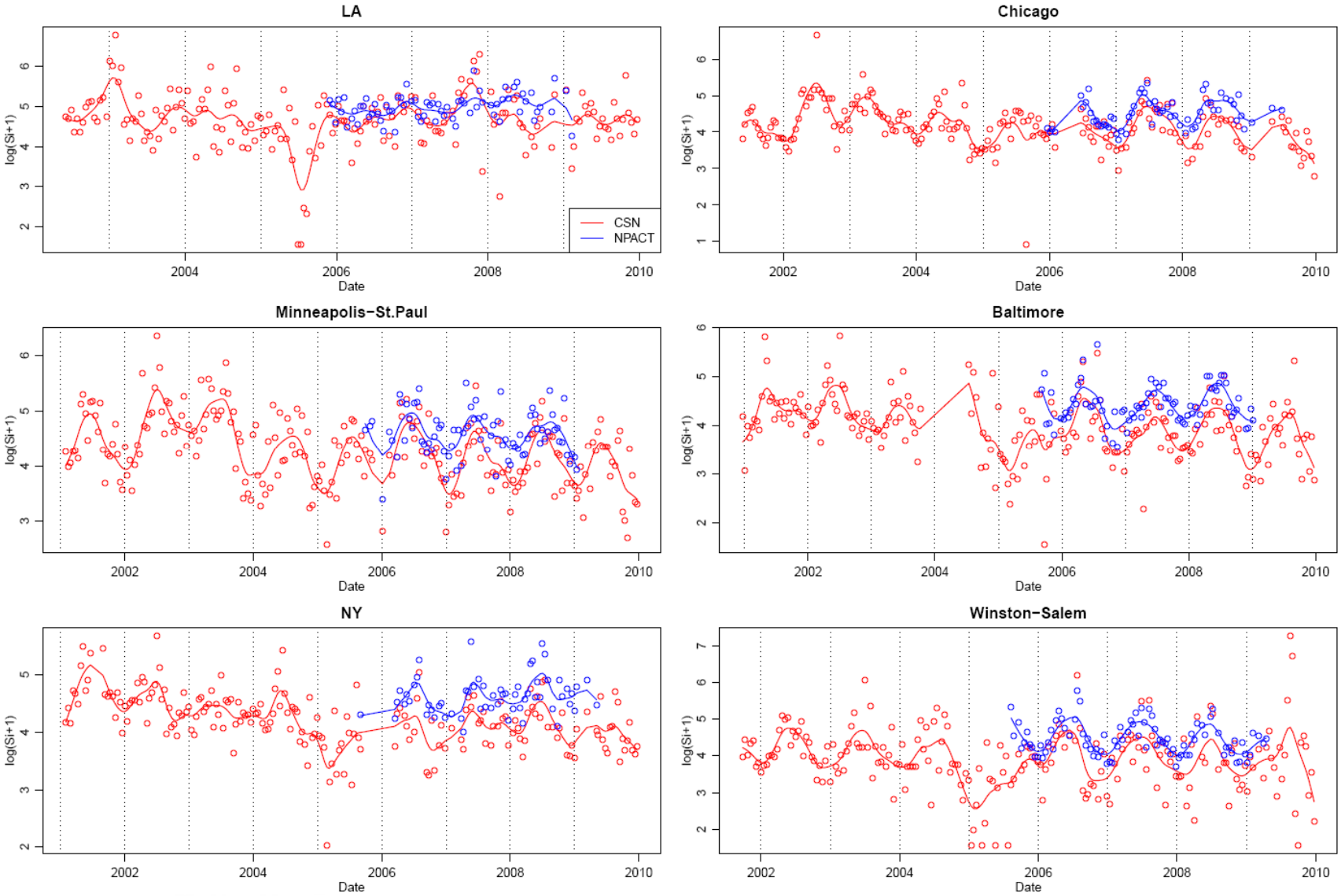
Supplemental Figure 1. Temporal trends of 2-week averages of silicon and EC measured by CSN and IMPROVE sites in Chicago, Minneapolis-St. Paul, Baltimore, New York, and Winston-Salem from 1999 through 2009



Supplemental Figure 2. Temporal trends of 2-week average of EC for the overlapping period from May 2007 through August 2008 between co-located CSN and NPACT fixed sites in each of six MESA city areas



Supplemental Figure 3. Scatter plots of 2-week averages of silicon for the overlapping period from August 2005 through August 2009 between co-located CSN and NPACT fixed sites in each of six MESA city areas



Supplemental Figure 4. Temporal trends of 2-week averages of silicon for the overlapping period from August 2005 through August 2009 between co-located CSN and NPACT fixed sites in each of six MESA city areas