SEASONAL VARIATION OF EMISSION FACTORS FOR ULTRAFINE PARTICLES FROM ON-HIGHWAY VEHICLES

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ABSTRACT
As urban populations increase, the need to estimate urban exposure to harmful aerosols increases. Simple Gaussian screening models can provide initial exposure estimates, but urban environments can create complex wind fields that exceed the capabilities of simple models. To fill this need, sophisticated models, such as the Quick Urban & Industrial Complex (QUIC) dispersion model, have been developed to model pollutant flow through urban environments. One pollutant of recent concern is ultrafine particles (UFP), particles whose diameter is less than 100 nm, and vehicle emission factors (EF) have been developed for UFP, in units of # of UFP/vehicle-km. The validity of urban exposure estimates depends on the predictive capacity of the EF estimate, and it is unclear if an EF estimate generated from average annual data can be used to adequately predict UFP concentrations at all times during a year. The goal of this research was to examine if an EF estimate that allowed average annual monitored data, collected in Ten Hills, MA, to fall within a factor-of-2 of QUIC model predictions could be used to adequately predict UFP concentrations in the summer and winter months (June through August and December through February, respectively). Monitored concentrations along Temple Road, a road orthogonal to highway I-93 (the main source of UFP in Ten Hills), were compared to QUIC predicted UFP concentrations. For data collected from hours where the prevailing wind direction was from the southwest (i.e., orthogonal to I-93), an EF of $5*10^{14}$ #/veh-km captured 92% of the annual average data within a factor-of-2 estimate, 68% of the summer data, and 52% of the winter data. When the prevailing wind direction was from the northwest (parallel to I-93), an EF estimate of $5*10^{14}$ #/veh-km captured 91% of the annual average data within a factor-of-2 estimate, 50% of the summer data, and 60% of the winter data. The impact of varying the EF based on the season was also investigated. Measured concentrations of UFPs were higher in the winter than in the summer, and varying the UFP EF with season greatly improved the amount of data captured by the factor-of-2 envelope from model estimates. The results of this research highlight the modeling capacity of QUIC, and show that season-sensitive EF estimates could generate more accurate UFP exposure estimates.

1 INTRODUCTION

1.1 Background

The adverse health effects associated with exposure to ultrafine particles (particulate matter less than 100 nm in diameter) in urban air have been the topic of recent publications (Weichenthal et al, 2011; Cho et al., 2009). Although researchers have been measuring ultrafine particles (UFP) in urban areas for more than 20 years (Seaton et al., 1995), the exact nature of the spatial distribution of UFP in urban areas is still being investigated (Zwack et al., 2011). To reduce health risks to urban populations, researchers must continue studying the sources and transport of UFP in urban areas.

The primary source of UFP in urban environments is vehicle exhaust (Shi et al., 2001; Wahlin et al., 2001). Recent studies have generated UFP-emission factor (EF) estimates for light duty vehicles, heavy duty vehicles, and a generalized fleet of vehicles (Zwack et al., 2011; Gidhagen et al., 2005; Kittelson et al., 2004; Gramotnev et al., 2003). EF estimates can be derived by calculations based solely on monitored UFP concentrations and traffic estimates (Kittelson et al., 2004), or by numerical models that use traffic levels and meteorological data to predict UFP concentrations. Regression and dispersion models can both be used to make EF estimations (Zwack et al., 2011), and once an EF has been determined, models can be used to give estimates for UFP concentrations across an entire site at a particular time. With models, researchers can make estimates of UFP levels over time in an urban area, and thus can make exposure estimates for urban populations.

1.2 Problem Statement

Conceivably, the need may arise for researchers to provide estimates of UFP exposure to urban populations (Cheng 2011, Mullen 2011, Knol 2009). However, the predictive value of the UFP exposure estimates depend heavily on the data on which the estimates are based. The goal of my research was to investigate if an EF estimate generated from averaged annual data can be used to adequately) estimate (i.e., the data is within a factor of 2 of the predictions) UFP concentrations in the seasonal extremes: winter (December through February) and summer (June through August).

1.3 Literature Review

Recent studies in Europe have shown that, in urban areas, background UFP concentrations vary seasonally, with background winter UFP concentrations being 2-3 times higher in the than background summer UFP concentrations (Virtanen et al, 2006, Pirjola et al, 2006). One possible reason for high UFP concentrations in the winter is the simultaneous occurrence of meteorological conditions that favor UFP formation; during the winter, the ambient temperature is lower and there is greater atmospheric stability (i.e., less atmospheric mixing). Kittelson (2000, 1999) showed that UFP formation during exhaust dilution and cooling is greater in lower ambient temperatures (exhaust dilution is defined as the cooling and expansion that occurs immediately after exhaust exists the exhaust pipe). Zhang et al. (2004) suggested a 2-dilution-stage model for UFP formation: traffic-generated turbulence-based dilution that occurs immediately at the exhaust exits the vehicle tailpipe (i.e., "tailpipe to road" dilution), and atmospheric dilution that occurs, due to atmospheric turbulence, in the minutes after exhaust is released (i.e., "road to ambient" dilution). In both dilution stages, lower atmospheric temperatures lead to higher UFP generation. In the first dilution stage ("tailpipe to road dilution), nucleation and condensation of UFP are driven by the steep temperature gradient that exists between the vehicle tailpipe and the ambient air immediately outside the tailpipe. When the temperature gradient is steepest (i.e., lowest ambient temperatures), the UFP formation processes in the first dilution stage (i.e., nucleation and condensation) increase in magnitude (see Zhang et al. 2004 for a more in-depth description of the nucleation and condensation processes that occur in the first dilution stage).

Robinson et al. (2009) also quantified the specifics of the generation of fine particulate matter from vehicle engines. The study found that gas-particle partitioning (i.e., the end destinations for particle matter produced by combustion reactions) varies with temperature and concentration, and both factors vary greatly as the gas moves from in the exhaust pipe (hot and highly concentrated) to the ambient air (cold and less concentrated). Fine particulates can also form as products in photochemical reactions once the hot exhaust gas is exposed to sunlight and oxidants in the ambient air. Based on the literature, the worst case scenario for UFP formation would be when low atmospheric temperatures are combined with high traffic levels and high atmospheric stability (a confluence of events that occurs during winter morning rush hours in urban centers in the northern hemisphere).

In an effort to quantify the contribution of traffic-generated UFP to measured UFP concentrations, researchers have calculated vehicle UFP EF. The EF calculated has units of particle number count (PNC or #) per vehicle per kilometer (i.e., #/veh-km). A range of EF have been found (Gidhagen 2005, Kittelson 2004, Gramotnev 2003, Zwack 2011), with most estimates falling in between 1.9 and 9 * 10^{14} #/veh-km for a gasoline-dominated vehicle fleet (containing a mix of heavy and light duty vehicles). The EF estimates vary from 0.41 to 6.5 * 10^{14} #/veh-km for light duty vehicles (passenger cars) and 25 to 113 * 10^{14} #/veh-km for heavy duty vehicles (commercial diesel vehicles). The time periods over which data was collected to produce these EF estimates varies from a few days (Kittelson 2004), to a few weeks (Zwack 2011), to a few months (Gramotnev 2003).

1.4 Motivation

Based on the current literature, I believe I have a unique opportunity to look at an unexplored area of UFP EF work. I have access to a large database of near-highway UFP concentration measurements, taken from a mobile monitoring vehicle in an urban area, from all seasons across 2 years, and a new dispersion-modeling program that may serve as an alternative to more simple line-source dispersion models.

1.4.1 Ten Hills, MA, Site Description

The data I used for my research was collected by the Tufts Air Pollution Lab (TAPL) in Somerville, Massachusetts (Ten Hills is included in the Somerville driving loop) (Padro-Martinez et al. 2012). This mobile monitoring lab is an RV owned by Tufts that is outfitted with rapid response equipment to measure UFP and other air pollutants. Over the course of 2009 and 2010, the TAPL equipment was used to measure UFP concentrations in Somerville, MA (~300 hours of data from all seasons). The driving loop in Somerville included Temple Road, a road that bisects Ten Hills and is perpendicular to highway I-93 (Figure 1, 2). The position of Temple Road with respect I-93, the major source of UFP in Ten Hills, makes it an ideal location for mobile monitoring and model calibration. The highway is elevated 5 m above ground level and there is a 3 m high sound barrier on the north side of I-93; Hagler et al (2012) showed how the presence of near highway sound barriers consistently reduced near highway UFP concentrations due to obstruction of wind flow off the roadway.

1.4.2 QUIC Model

Simple Gaussian dispersion models based on a line source are currently the EPA standard for estimating pollutant concentrations near busy roadways. However, line-source dispersion models may be too simplistic to capture the complexities of pollutant dispersion in urban environments. For my thesis, I used the recently-developed Quick Urban & Industrial Complex (QUIC) dispersion modeling system to model UFP transport through Ten Hills, Massachusetts (Nelson and Brown, 2010). QUIC is designed to account for pollutant flow around complex building geometries, and has been used previously to generate UFP EF estimates (Gowardhan et al. 2006, Bowker et. al. 2007, Zwack et al., 2011).

1.4.3 Mobile Monitoring Data

From the data collected in Somerville along Temple Road, I created UFP concentration profiles with respect to perpendicular distance from I-93. I then compared my QUIC-generated UFP concentration estimates along Temple Road to measured concentration profiles and calibrate the QUIC model by adjusting the EF of vehicles on I-93. The comparison of dispersion model output with monitored concentrations is specifically mentioned in Zwack et al. (2011) as a strategy that can be potentially used to capture the spatial variations of UFP in an urban area. I used data from many Somerville driving loops in an effort to capture the temporal variations of UFP. The depth of the Somerville data set allowed me to observe the effect of a wide range of meteorological conditions on the QUIC EF estimate.

1.5 Goals and Objectives

I hypothesize that the vehicle UFP EF based on averaged annual UFP concentrations will overestimate summer UFP concentrations along Temple Road and underestimate winter UFP concentrations along Temple Road. To test this hypothesis, I matched the QUIC output along Temple Road to mobile-monitoring data collected along Temple Road by adjusting the QUIC source emission factor (i.e., the vehicle emission factor in #/veh-km). I will use data from different monitored days to capture the widest range of mean hourly temperatures and wind directions.

2 METHODS

2.1 Mobile Monitoring Data Preparation

Mobile monitoring data was collected on Temple Road for almost 300 hours throughout 2009 and 2010. The data had already undergone an extensive quality assurance and quality control regimen (as described by Padro-Martinez et al. 2012). The monitored concentrations were averaged into bins by distance from I-93 (Appendix A). The first bin is at ~14m from the highway. I created a database of UFP measurements taken along Temple Road so that I could easily compare QUIC model results to the measured concentrations. Background UFP concentrations of 24,010 (winter), 13,730 (summer), and 17,808 (annual average) #/cm^3 were used (Padro-Martinez et al. 2012).

2.2 QUIC Model Setup and Implementation

The newest version of QUIC has the ability to import a GIS shapfile of buildings into its domain. Since there are more than 200 buildings in Ten Hills, I made use of this import function (Figure

3). The entire Ten Hills site north of highway I-93 can be contained in an 800 m (x) by 600 m (y) by 50 m (z, elevation) grid, which is within the limits of QUICs computational ability. I included an 800 m stretch of the I-93, elevated at 5 m above ground level, and included an approximation of the sound barrier that extends 3 m above the highway surface on the north side of the highway (~600 m long). Although there are some elevation changes in the Ten Hills site, I did not include them in the model setup (due to time constraints and the added model complexity). I also assumed that the 800 m of highway that I included in the domain adequately represents the main source of UFP to Ten Hills during most wind directions (i.e, W, SW, NW).

The first step of running QUIC is the wind field generator, QUIC-URB. QUIC-URB is a simplified computational fluid dynamics model (CFD), and I ran it in its simplified form (there are options to make QUIC-URB make CFD calculations, but it drastically increases the QUIC-URB runtime). Wind direction and wind speed are the only inputs needed to QUIC-URB, and it outputs wind field approximations at all designated elevations throughout the site (Figure 4). The wind speed and direction measurements were taken from the Hormel Stadium weather station, approximately 1.6 km northwest of Ten Hill. I used the geometric mean wind speeds in my model, but other studies have used harmonic mean wind speeds (Marshall 2005). Although the Hormel station is elevated 34m above ground level, QUIC-URB has a wind profile selection tool that I used to extrapolate the wind speed down to ground level. The study area of Zwack et al. (2011) was an urban neighborhood of Brooklyn, NY, and they used the "urban canopy" wind profile (other options include "power law" and "logarithmic"). For the Ten Hills domain, I chose to model the wind profile as an "urban canopy," due to the density and height of buildings in Ten

Hills. I also assumed that the wind conditions in Ten Hills could be approximated using wind conditions at the Hormel station.

The second and final step in the QUIC model process is the computation of the spread of an aerosol or gas through the study area, a process that is accomplish by QUIC-PLUME (a sub-program in QUIC). In my scenario, I modeled I-93 as an elevated rectangular source that was continuously emitting pollutant "particles" (mass/time). An argument could be made to model UFP as an inert gas, since its settling velocity is insignificant; however, since the goal of this research was to generate a particle number count emission factor for UFP, I chose to model UFP as a particle.

To convert from the standard unit of UFP EF, #/veh-km to mass/time (mg/s), I assumed a constant UFP density of 1 g/cm^3 and a median UFP diameter of 0.1 μm, and performed the following unit conversions:

$$\frac{\text{\# of UFP}}{\text{vehicle} * \text{km}} * \mathrm{L} * \ \mathrm{V} * \left(\frac{4}{3}\pi\left(\frac{\mathrm{D_{UFP}}}{2}\right)^3\right) * \rho_{\mathrm{UFP}} * \frac{1\ \text{hour}}{3600\ \text{sec}} = \frac{Mass}{Time}$$

Where: L = length of highway in domain (800m)

V = number of vehicles per hour (vehicles/hour)

$\mathrm{D_{UFP}}$ = assumed median diameter of UFP (0.1μm, the minimum allowed median aerosol diameter in QUIC)

ρ_{UFP} = assumed density of UFP (1g/cm^3)

The number of vehicles per hour were taken from the MASS Highway database (MassGIS); the averaged hourly number includes both northbound and southbound traffic on I-93.

One important variable to the QUIC-PLUME program is the description of the temperature and humidity profiles of the site. The temperature and relative humidity measurements were taken by the mobile monitoring lab. To attain a temperature and humidity profile, I assumed that humidity was constant throughout the study area (up to 50 m of elevation), and I assumed that the temperature profile followed the adiabatic lapse rate (in all simulations, the temperature from the surface to 50m changed by less than 0.2 K).

Another important input variable to QUIC-PLUME is the number of particles released in the simulation. This variable is important because of the method by which QUIC-PLUME calculates concentration estimates. The QUIC-PLUME source term for roadway UFP emissions is represented by a mass flow rate. The user then defines the number of particles to be released in the simulation, and the length of the simulation. From those two inputs, QUIC-PLUME assigns each released particle an emitted mass weighting. If the user chooses to release fewer particles, each particle will carry a greater percentage of the total mass emitted during the simulation. If the user chooses to release more particles, each particle is assigned a smaller mass weight. The mass flow rate from the source in either case is the same, as is the physical mass of each particle; the mass weighting is purely a method for calculating the concentration. Concentrations at certain locations in the study area are calculated by summing the amount of particles that pass through a cell in the user defined "collection grid." The "collection grid" is a series of 3-D cells through which emitted particles can pass unobstructed. I chose a collection grid along Temple

Road, made of 1 m x 1 m x 1 m cells at an elevation of z = 3 m, approximately the height that the RV collected measurements. QUIC-PLUME sums how many particles pass through each grid cell over the simulation time, and calculates the concentration estimate at a specific cell from the mass of particles that came through the cell, and the volume of air in the cell. Zwack et al. (2011) released 10^6 particles in their simulations. For my simulations, I set QUIC-PLUME to release $2*10^5$ particles so I could view the impact of EF adjustment more quickly. I ran the QUIC-PLUME simulations for 1 model hour = 3600 sec, and started collecting concentration measurements at 1200 sec (I assumed that the initial released particles had passed out of the domain by that time). I averaged the concentration estimates over the remaining 2400 seconds of the simulation.

QUIC-Plume has several methods by which to display the output data. Similar to the output from QUIC-URB, the output of concentration estimates across the entire study area at various elevations can be generated (Figure 5); however, for this project, I only used the model output specific to Temple Road.

2.3 Output Analysis

The mobile monitoring data was averaged into bins based on distance from I-93(Appendix A), and the QUIC model results were adjusted to match those bins. For example, if the data contained measurements at 5 m, 15 m, and 25 m from I-93 along Temple Road, then QUIC output from 0 m to 10 m away from I-93 would be averaged and compared against the monitored concentration at 5 m from I-93, and QUIC output from 10 m to 20 m would be averaged and compared against the monitored concentration taken at 15 m from I-93.

The mobile monitoring dataset includes data over the course of 2 years, from all seasons and wind directions. I separated the monitored data into 12 sections, by prevailing wind direction (i.e., NW, NE, SE, SW) and by season (i.e., annual average, summer, winter), and analyzed QUIC results specific to each dataset.

To better facilitate rapid comparison between QUIC model results and mobile monitoring measurements, I created an EXCEL macro that takes a QUIC output text file and displays, graphically and statistically, the relationship between the model output and the relevant subset of data (the GUI of the macro are included in Appendix B).

Several quantitative goodness-of-fit statistics were used to compare QUIC model results to mobile monitoring data. The most common goodness-of-fit statistic is the coefficient of determination, or R^2. R^2 is calculated by the following formula:

$$R^2 = \frac{\left[\sum_{i-1}^{N}(\mathrm{Modeled}_i - \mathrm{Mean}_{\mathrm{Modeled}})(\mathrm{Measured}_i - \mathrm{Mean}_{\mathrm{Measured}})\right]^2}{\sum_{i-1}^{N}(\mathrm{Measured}_i - \mathrm{Mean}_{\mathrm{Measured}})^2 \sum_{i=1}^{N}(\mathrm{Modeled}_i - \mathrm{Mean}_{\mathrm{Modeled}})^2}$$

However, R^2 can give misleading results if the model exhibits bias (Montgomery 1970); therefore, I included a calculation of normalized model bias in my goodness-of-fit statistics. Normalized model bias is calculated by the following formula:

$$\mathrm{Bias} = \frac{\mathrm{Mean}_{\mathrm{Modeled}} - \mathrm{Mean}_{\mathrm{Measured}}}{\mathrm{Mean}_{\mathrm{Measured}}} * 100\%$$

The Nash-Sutcliffe Efficiency Criterion (or E-value) was used in place of R^2 to compare the fit of the model to the data (Nash 1970). The E-value is calculated by the following formula:

$$E = 1 - \frac{\sigma^2_{\text{residuals}}}{\sigma^2_{\text{Measured}}} = 1 - \frac{\sum_{i=1}^{N}(\text{Measured}_i - \text{Modeled}_i)^2}{\sum_{i=1}^{N}(\text{Measured}_i - \text{Mean}_{\text{Measured}})^2}$$

The E-value can be calculated in real or log space (by substituting ln(Measured) in for Measured concentrations and ln(Modeled) in for Modeled concentrations in the formula).

A final statistic that was calculated was the coefficient of variation of model residuals (Coeff. of Residuals). The coefficient of residuals estimates the average percent error associated with model output, and is calculated by the following formula:

$$C_{\text{residuals}} = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left[(\text{Modeled}_i - \text{Measured}_i) - \frac{1}{N}\sum_{i=1}^{N}(\text{Modeled}_i - \text{Measured}_i)\right]^2}$$

3 RESULTS

3.1 Mobile Monitoring Data Summary

Investigation of UFP trends over the course of the year reveal that, with the exception of UFP data from hours with prevailing southeast winds, the highest measured UFP concentrations occur in the winter (Dec, Jan, Feb) and the lowest measured UFP concentrations occur in the summer (Jun, Jul, Aug), with the annual average UFP concentrations (i.e., concentrations averaged over all monitoring days from 2009 and 2010) falling in between the winter and summer UFP measurements (Figure 6). The winter data from hours with prevailing southeast winds breaks from the trend observed in the other datasets (Figure 6d); however, this dataset is derived from a small number of monitoring hours (2 hours), and thus averaged UFP concentrations from the winter southeast wind dataset could potentially not be representative of actual UFP concentrations on winter days with prevailing southeast winds.

In monitored data from hours with prevailing southwest winds and hours with prevailing northwest winds, a slight 'spike' in measured UFP concentrations occurs at approximately 80-90 m down Temple Road (Figure 6). For days with prevailing northwest and southwest winds, QUIC models this spike in monitored data (Figure 7 & 8). A possible explanation for this spike in the data is configuration of the Grimmons housing complex on the north side of Temple road and Governor Winthrop road (Figure 2).

3.2 Mobile Monitoring Data Analysis

Since the goal of this thesis is to investigate vehicle emission factors, it follows that model comparison to data should be conducted on data in which I-93 is the main source of UFP to Ten Hills. This eliminates from investigation monitoring hours where the prevailing wind was from the southeast or northeast. Additionally, the limited winter (11 hours of winter northeast-wind UFP measurements, 2 hours of winter southeast-wind UFP measurements) and summer (6 hours of summer northeast-wind UFP measurements, 11 hours of summer southeast-wind UFP measurements) datasets from hours with prevailing southeast and northeast winds makes hours with prevailing northwest and southwest winds the better choice for analysis and model comparison. The measurements taken during the winter with prevailing southwest winds were presumed to be representative of average UFP concentrations under those conditions, even though the averaged values are based on only 4 hours of data (the implications of this assumption will be discussed).

3.3 Emission Factor Fitting

A vehicle EF of $5*10^{14}$ #/veh-km was shown to have the best relationship with data when applied, over the annual average of data, to hours with prevailing northwest winds and hours with prevailing southwest winds. An EF of $5*10^{14}$ #/veh-km is well within the literature supported range of UFP EF (1.9 - 9.0 #/veh-km as per Kittelson 2004).

3.3.1 QUIC Results vs. Data with Prevailing Northwest Winds

QUIC results moderately predicted monitored data from hours with prevailing northwest winds (Figure 7). The EF of $5*10^{14}$ #/veh-km was found to give the closest model fit to the data for monitored data over the whole year. The closest relationship between QUIC results and data was average annual data: 91% of monitored data was within a factor of 2 (i.e., data<model*2 and data>model/2) of QUIC model results (Table 1).However, the shapes of the profile of modeled concentrations and profile of data are not similar (Figure 7a), and the model misses the characteristic spike in the data at 80-90 m. For summer data with prevailing northwest winds, the model captured monitored UFP concentrations at a distances greater than 75 meters from I-93 within a factor of 2, but overestimated the near-highway (distance from I-93 < 75m) UFP concentrations by a factor of 3 (Figure 7b). The shape of the profile of modeled winter concentrations is similar to the profile of measured winter data, however the model underestimates winter concentrations by a factor of 1.5 (Figure 7c). The model results from all three datasets do not extend to the edge of the QUIC domain as a result of the wind direction and size of the QUIC domain. In the northwest wind datasets, the wind is blowing almost straight down the highway (I-93 in the QUIC domain is situated at exactly 315^o; the average wind directions from the annual average, summer, and winter datasets are 310^o, 299^o, and 314^o respectively). Thus, in the QUIC simulation, emitted particles are blown out of the QUIC domain before they can travel down the length of Temple Road.

3.3.2 QUIC Results vs. Data with Prevailing Southwest Winds

Using the same EF as was used to investigate days with prevailing northwest winds (EF = $5*10^{14}$ #/veh-km), QUIC-predicted UFP concentrations had moderate agreement with monitored data from days with prevailing southwest winds averaged over both years of data (Figure 8a). For the annual average of data (the average UFP concentrations on Temple Road on all monitoring hours in 2009 and 2010 that had prevailing southwest winds), 92% of data fell within a factor of 2 of QUIC model results. However, the model underestimated the UFP concentrations nearest to I-93 (<100 m). A moderate relationship existed between model results and data for summer days with prevailing southwest winds: 68% of data fell within a factor of 2 of QUIC results. A slightly poorer relationship existed between model and data for winter days with prevailing southwest winds: only 52% of data fell within a factor of 2 of QUIC predications.

The dataset derived from winter days with prevailing southwest winds is an average of only 4 monitoring hours (Figure 6c): therefore, similar to averaged UFP concentration from winter hours with prevailing southeast winds, data from winter days with prevailing southeast winds is likely not representative of actual average UFP conditions on those days.

4 DISCUSSION

4.1 Use of R^2, Nash-Sutcliffe, % Bias, Correlation Coeff., and "Factor of 2 Envelope"

In cases where there exists significant model % bias, R^2 terms may be misleadingly high and should be not be considered for model-fit analysis. For example, for QUIC predictions of UFP concentrations during summer hours with prevailing northwest winds using an EF of $5*10^{14}$ #/veh-km, the QUIC model exhibits high model bias (%Bias = 116%), but the model vs. data R^2 was high (R^2 = 0.824). Visual inspection of the corresponding plot (Figure 7b) confirms this fallibility of R^2, as the model over-predicts the first 100 m of monitored data by a factor of 3 or greater. The Nash-Sutcliffe E-value (both in real and log space) was used to compare QUIC results to data in place of the unadjusted R^2 value (Table 1).

As an alternative to the Nash-Sutcliffe E-value and the R^2 value, a third method for comparing the model to the data was used: the "Factor of 2 Envelope." If a data point is less than the model prediction*2 and greater than half the model prediction, than in falls within the "Factor of 2 Envelope." The "Factor of 2 Envelope" approach has been used in previous studies with dispersion models (Yura 2007), and I used it as the main analytical tool in this investigation.

4.2 Model Choice Evaluation

Simpler dispersion models than QUIC exist that could have been used for this research. One such model is CALINE4 (Benson 1984), a simple line dispersion model that doesn't apply complex site topography (such as a sound barrier) to UFP concentration calculations. CALINE4 is currently used by the EPA in the permitting process. Although CALINE4 could have been used to perform my analyses, one of the purposes of performing this research was to evaluate QUIC as a tool for future modeling efforts. Since QUIC has the capacity to incorporate complex urban terrain into model calculations, it has potential applications in public health studies in urban areas (e.g., CAFEH). The results of my research show that QUIC can be used to predict UFP concentrations in certain situations.

4.3 Model Performance Evaluation

Overall, QUIC predictions of UFP concentrations were closer to measured UFP concentrations for hours where the prevailing wind was from the southwest as opposed to hours where the prevailing wind was from the northwest (Table 1). QUIC model predictions captured (for the annual average, summer, and winter datasets respectively) 91%, 50%, and 60% of data in the factor of 2 envelope for hours with prevailing northwest winds, as opposed to 92%, 68%, and 52% of the data from hours with prevailing southwest winds. These results indicate that an EF calculated from average annual data can provide good model fit to annual data, but will likely overestimate summer UFP concentrations and underestimate winter UFP concentrations.

One possible reason for the moderate model fit with data from hours with northwest winds is the known shortcoming of dispersion models on days with high wind speeds (m/s > 4.0) where the wind direction is parallel to the line source in the site (Benson 1992). For each dataset analyzed (annual average, summer hours, winter hours) the average wind speed from the northwest was >15% higher than the average wind speed from the same dataset with a prevailing wind from the southwest (average northwest wind speeds are 14%, 25%, and 20% higher than average southwest wind speed for annual average, summer, and winter hours of data, respectively). However, each northwest wind dataset is averaged from more than 20 different hours of data, so it is unlikely that a single outlier is skewing the averaged UFP concentrations. Although this shortcoming of dispersion models is well documented in the literature, it may be more likely that a model assumption (e.g., the complex wind patterns around the sound barrier, near-highway UFP formation processes) led to the inability of QUIC to adequately predict data on days with prevailing southwest winds.

4.4 Validity/Impact of Assumptions

Throughout the investigation, major assumptions were made concerning the definition of the source term (highway I-93), the path of wind through the study area (Figure 4), and the physical properties of UFP in the QUIC domain (Appendix C). These assumptions were classified as "major" assumptions based on the level of influence each factor had on model UFP predictions. Along with the major assumptions that were made, several "minor" assumptions were made including the representativeness of the measured data for the specific season the measurement was taken in, the composition of the vehicle fleet, and the background concentration of UFP. The effect of each these assumptions was analyzed.

4.4.1 Source Term Definition

A major assumption made in the QUIC model was the assumption that the part of I-93 contributing the most of the UFP year-round was the 800 m stretch of highway that bounds Ten Hills on the southwest side (Figure 2). Especially on days with prevailing northwest winds, in combination with the physics of the QUIC model, this assumption may have caused QUIC to underestimate UFP concentrations on Temple Road, especially on days where the section of I-93 not included in the QUIC domain could potentially contribute more UFP to the study area than UFP contributed by the section of I-93 included in the present domain (i.e., hours in which the prevailing wind direction is parallel to I-93, either from the northwest or southeast). An analysis of the effects of including a greater length of highway in the QUIC domain is recommended for future studies in this study area.

4.4.2 Highway Elevation and Sound Barrier

The definition of the source term in Ten Hills was made more difficult by the elevation of the highway (5 m above ground level), and by the presence of the 3 m high sound barrier on the north side of the highway. QUIC can model an elevated line source, however, estimating the passage of air and UFP under the highway adds additional complexity to the QUIC model.

The presence of the sound barrier also increases the complexity of the physics in the QUIC domain, and may be a source of error between QUIC model results and monitored data, especially for near-highway UFP measurements. Previous studies have suggested that line sources, such as I-93, only contribute significantly to near-highway (<100m) UFP concentrations (Zwack et al. 2011), and the Ten Hills near-highway geometry is complex (there is a row of elevated trees behind the sound barrier and Temple Road slopes up approximately 25m north of I-93), further increasing the difficulty of assessing the influence of I-93 vehicle emissions on concentrations of UFP on Temple Road. Especially in reference to the estimation of the wind streamlines through the study area, the complex near-highway geometry in Ten Hills could be a source of discrepancy between QUIC model results and the data. Comparison of model results to data suggests that the near-highway zone is more well mixed than the model is predicting, at least for hours with prevailing southwest winds (Figure 8).

4.4.3 Hormel Stadium Wind Data

The first step of QUIC's modeling approach is the approximation the wind field through the study area (a simplified CFD model called QUIC-URB). The wind data used by the TAPL dataset is taken from the Hormel Stadium weather station (station elevation = 34 meters above ground level), approximately 1.6km northwest of the Ten Hills site. Inevitable disparities between the measured and actual wind direction and speed would arise if the weather station were located in the study area, and these disparities could potentially be magnified since the weather station is elevated, 1.6 km northwest of the study area, and located above a stadium, as opposed to an urban canopy (i.e., Ten Hills). QUIC has methods for approximating a wind speed profile based on reference height, wind speed, and terrain (e.g., "urban canopy" is a wind speed profile option, other options include "power law" and "logarithmic"), and it is recommended that the effects of using different wind profiles being investigated in future studies. Additionally, using a weather station that is closer in elevation and proximity to the study area would likely improve the accuracy of the wind field approximation.

4.4.4 Source Term Calculation

In the calculation of the QUIC-PLUME UFP source term, a median particle diameter of 0.1μm and a particle density of 1 g/cm^3 were assumed (Appendix C). The median particle diameter of 0.1 μm is the minimum median particle diameter allowed by the QUIC code. These same assumptions were made by Zwack (2011a). For future studies, it is recommended that a particle sensitivity analysis be conducted, involving examining the effects of smaller median particle diameters (e.g., 0.05μm, 0.02μm) and various particle densities (e.g., 2 g/cm^3, 0.5 g/cm^3, 0.2 g/cm^3), to examine the effects of varying UFP characteristics.

4.4.5 Topography of Ten Hills

A major topographical element of the Ten Hills domain was left out of the model creation: Ten Hills is located on a hill, whose apex is approximately 6m above the base. The effects of this elevation change should be investigated by future research, although the ability of QUIC to adequately model monitored data under certain conditions suggests that including this elevation change may not drastically improve model results.

4.4.6 Vehicle Fleet Composition

Although traffic composition in most monitoring hours were dominated by class 1 vehicles, non-commercial passenger cars, there were hours that contained class 4 and class 5 vehicles, heavy duty diesel vehicles (Appendix A). Kittelson (1999) suggests a variety of emission factors for a gasoline dominated fleet ($1.9 - 9 * 10^{14}$ #/veh-km), but a better estimate of the source term in this QUIC domain could include a composite EF, derived from the combination of the gasoline and diesel EF weighted by their respective contributions to the total UFP generation.

4.4.7 Background UFP Concentration

Background UFP concentrations of 24,010 (winter), 13,730 (summer), and 17,808 (annual average) #/cm^3 were used in model calculations (Padro-Martinez et al. 2012). However, the background location was ~3 km away from Ten Hills. A better background estimate for Ten Hills could likely be attained by measuring the near-highway UFP concentration profile directly upwind of the highway (<400 m) for any given day.

4.4.8 Representativeness of Winter Southwest Data

It was assumed that measured UFP concentration data taken during winter hours with prevailing southwest winds was representative of average UFP concentrations under those conditions, despite being averaged from only 4 monitoring hours. It is recommended that futures studies increase the monitored concentrations on winter days with prevailing southwest winds so that more accurate averaged UFP concentrations can be used in modeling efforts.

4.4.9 Temperature and Relative Humidity

I assumed adiabatic lapse rate for temperature profile (-9.8 K/1000 m), which under certain meteorology conditions may not be accurate (e.g, morning temperature inversions). However, for both temperature and relative humidity, I except that neither would not change significantly over the 50m of elevation in the QUIC domain.

4.5 Impact of Varying the EF

The effect of decreasing the EF estimate for model predictions during the summer and increasing the EF estimate for model predictions during the winter was investigated (Figure 9 & 10). For the summer data from hours with prevailing northwest winds and hours with prevailing southwest winds, decreasing the EF by approximately half greatly increased the percent of modeled data that fell within the "factor of 2 envelope" of the model (50% increase for northwest wind days, 22% increase for southwest wind days, with both adjust models capturing 100% of monitored data within a factor-of-2) and greatly increased the Nash-Sutcliffe E-value (both real and log space) of each model (Table 1). This result suggests that during the summer I-93 is the main source of UFP to Ten Hills.

Increasing the EF for winter hours (both wind directions) also greatly increased the amount of data that was captured by the factor of 2 envelope (60% to 100% for hours with northwest winds, 52% to 88% for southwest winds). However, visual inspection of QUIC results vs. data for both wind directions reveals that even with the increased winter vehicle EF, the model still underestimates the measured concentrations (Figure 9, 10). This result suggests that during the winter, there is potentially another source (other than I-93) of UFP to Ten Hills that is not captured in the current rendition of the QUIC model. Other possible sources of UFP include: additional atmospheric (1st or 2nd dilution stage) formation of UFP in the domain, or another upwind source of UFP that becomes relevant during the winter months. I recommend a future study investigate the additional sources of UFP during winter months.

5 CONCLUSIONS

Overall, the results of this study indicate that although it may be valid to use a single EF estimate across multiple seasons, better season specific UFP exposure estimates will come from better characterization of seasonal variation of EFs. An EF calculated from average annual data can provide good model fit to annual data, but will likely overestimate summer UFP concentrations and underestimate winter UFP concentrations. As a rule of thumb, EF should be used to predict data under similar conditions that the EF was measured/calculated.

Due to the nature of this data, it is impossible to characterize which dilution stage plays a bigger role ("tailpipe to road" or "road to ambient") in the increased measured UFP concentrations in the winter. It is likely that a combination of increased coagulation, nucleation and condensation, along with higher winter EF are the source for higher winter UFP concentrations. Future studies should work to characterize the relative contributions of the dilution zones to near-highway UFP concentrations. In addition, future work on EF estimates using QUIC should investigate more closely the near-highway modeling (<100 m from I-93) capabilities of QUIC, and should change QUIC model parameters to increase the precision of model results (i.e., run QUIC with 10^6 instead of $2*10^5$ particles).

6 ACKNOWLEDGEMENTS

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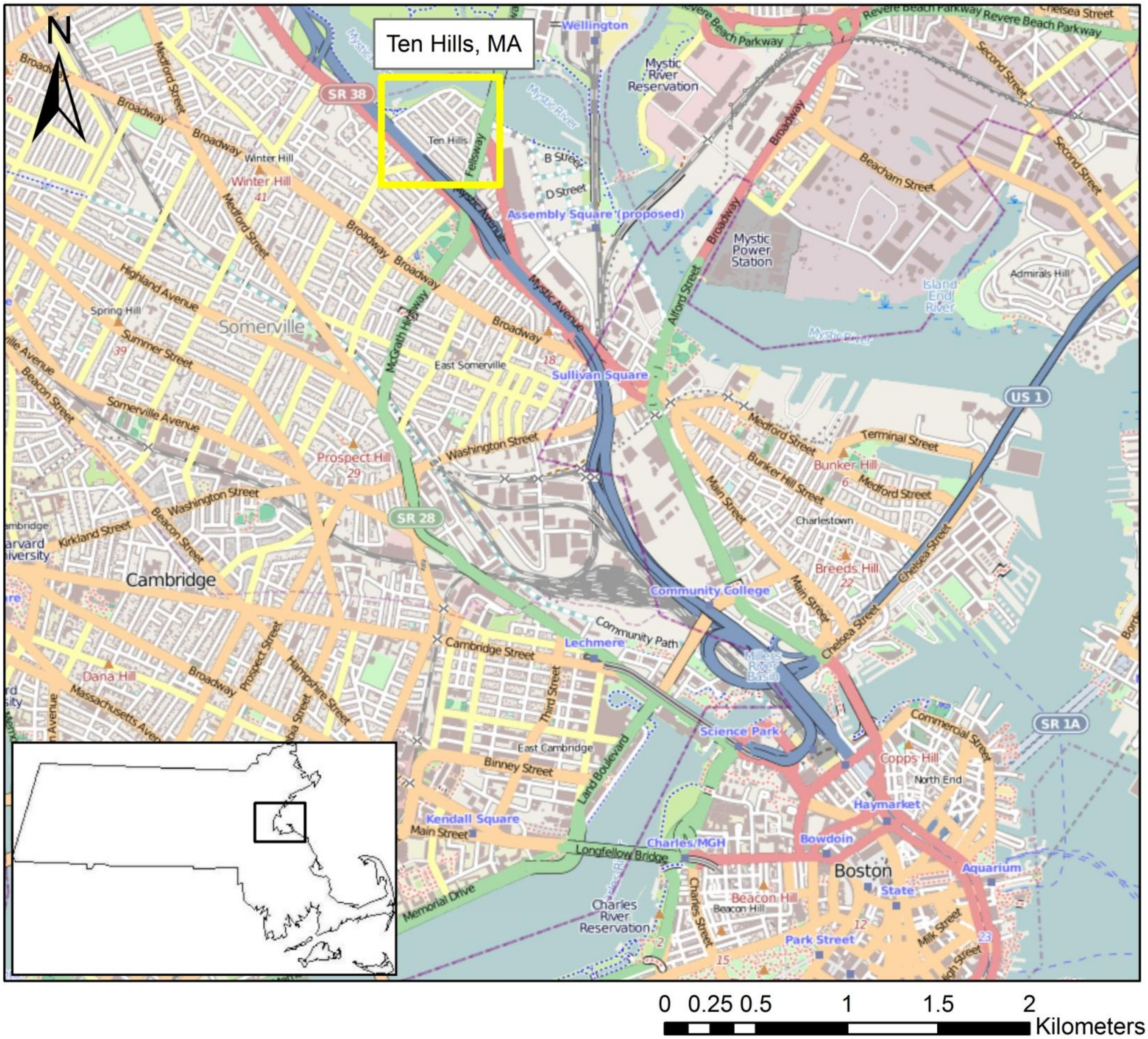


Figure 1. Location of Ten Hills in the Greater Boston Area.

Figure 2. Site map of Ten Hills, Massachusetts. Only the buildings north of I-93 will be used in my analysis.

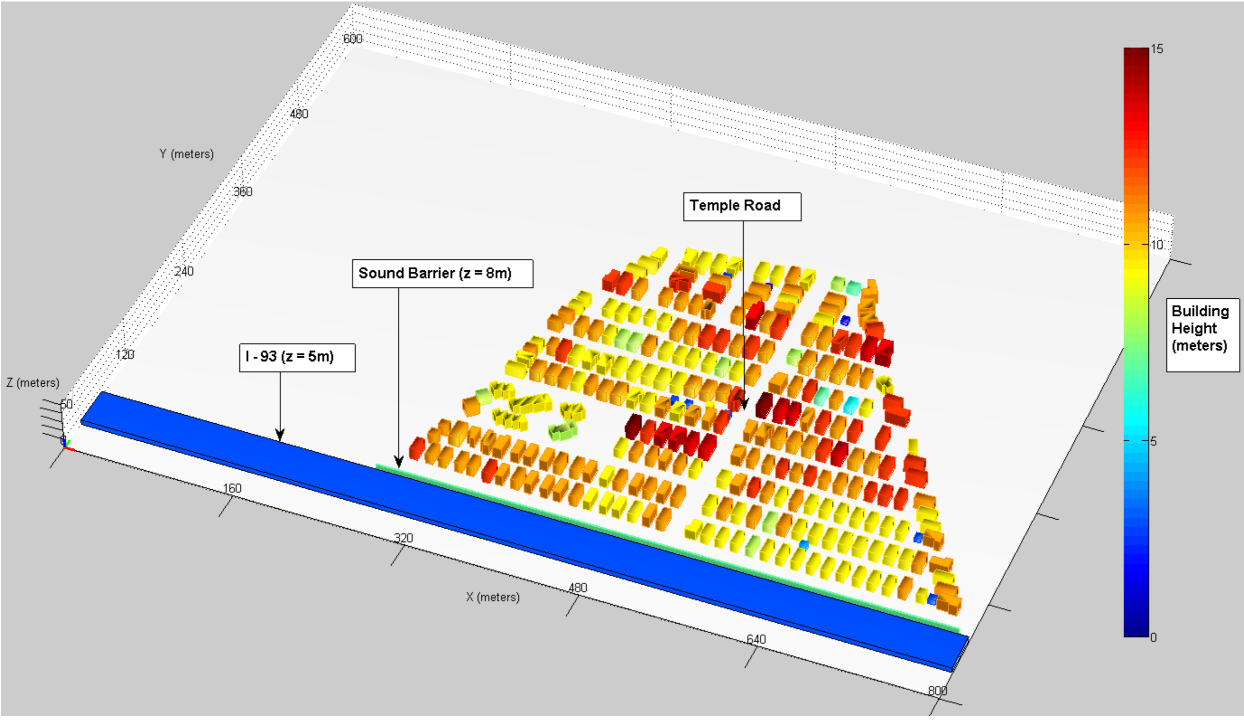


Figure 3. MATLAB-generated image of the Ten Hills site.

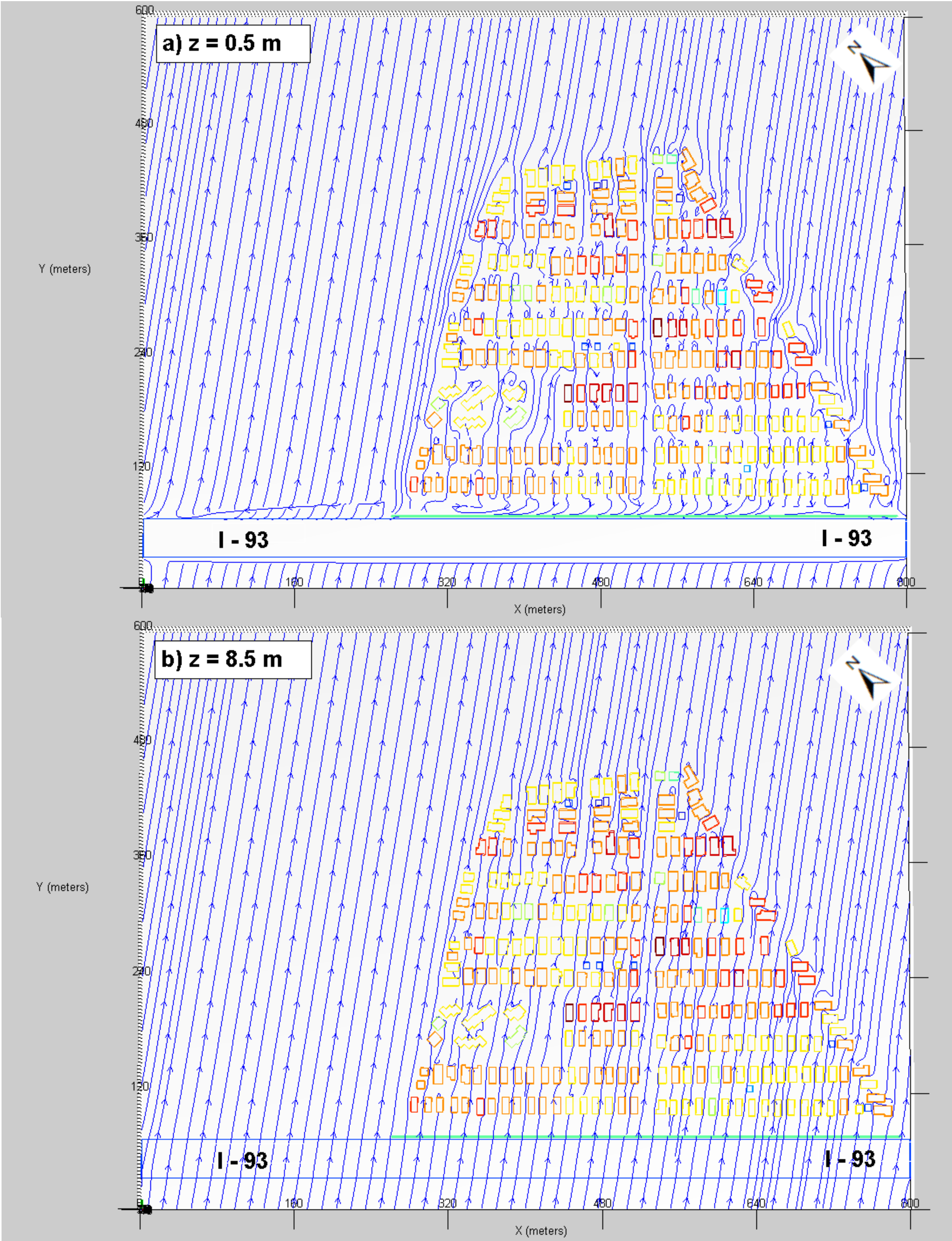


Figure 4. The wind profile generated by QUIC-URB at a height of (a) 0.5 meters and (b) 8.5 meters. On this day, the wind direction is 234^o, blowing at 4.6 m/s at a reference height of 34 meters. QUIC has algorithms to adjust the wind speed to a height of 3m.

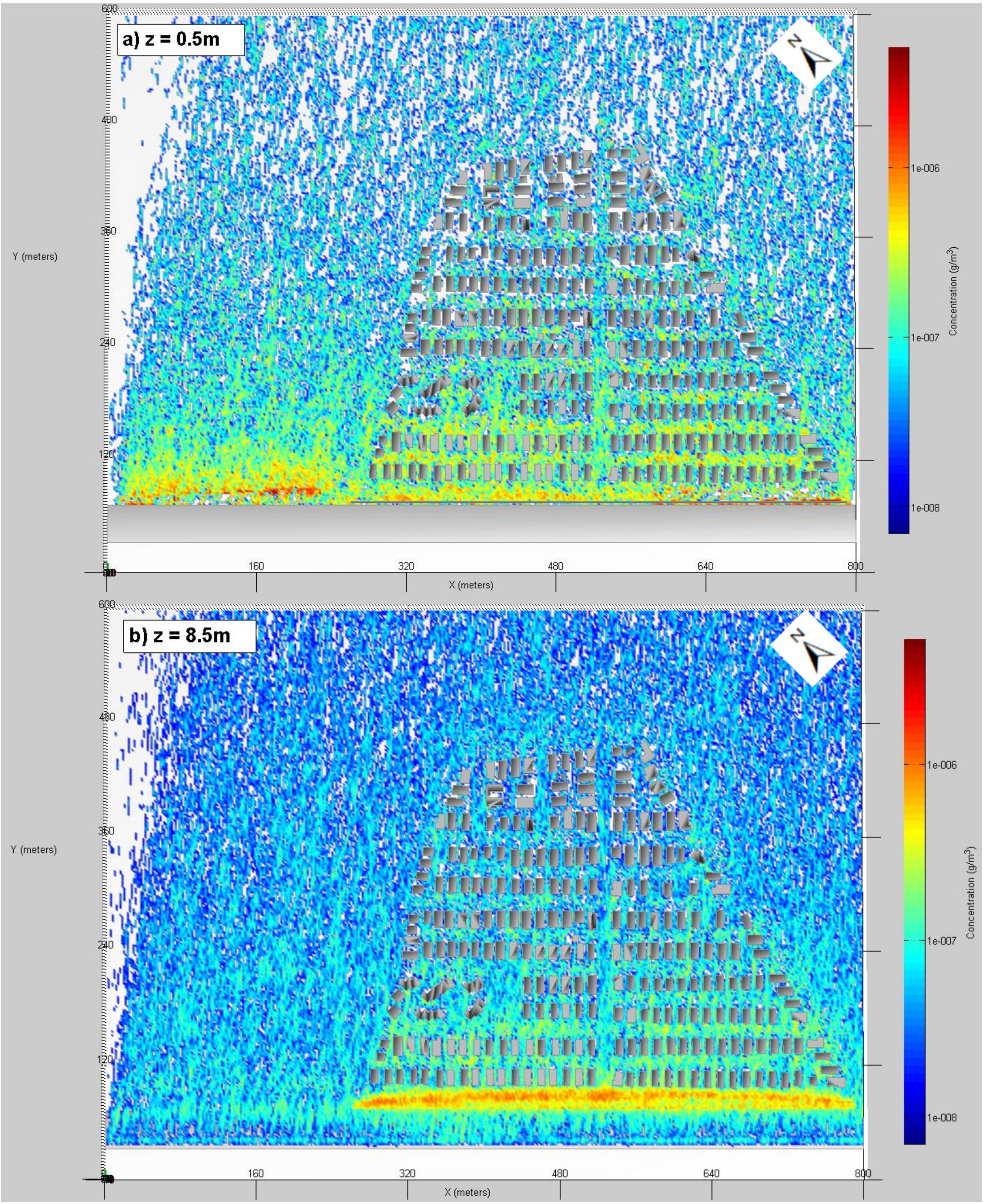


Figure 5. The concentration profile of an inert generic bulk aerosol from QUIC-PLUME at a height of (a) 0.5 meters and (b) z = 8.5 meters. In this simulation, the wind direction is 234^o, blowing at 4.6 m/s at a reference height of 34 meters. QUIC has algorithms to adjust the wind speed to a height of 3m.

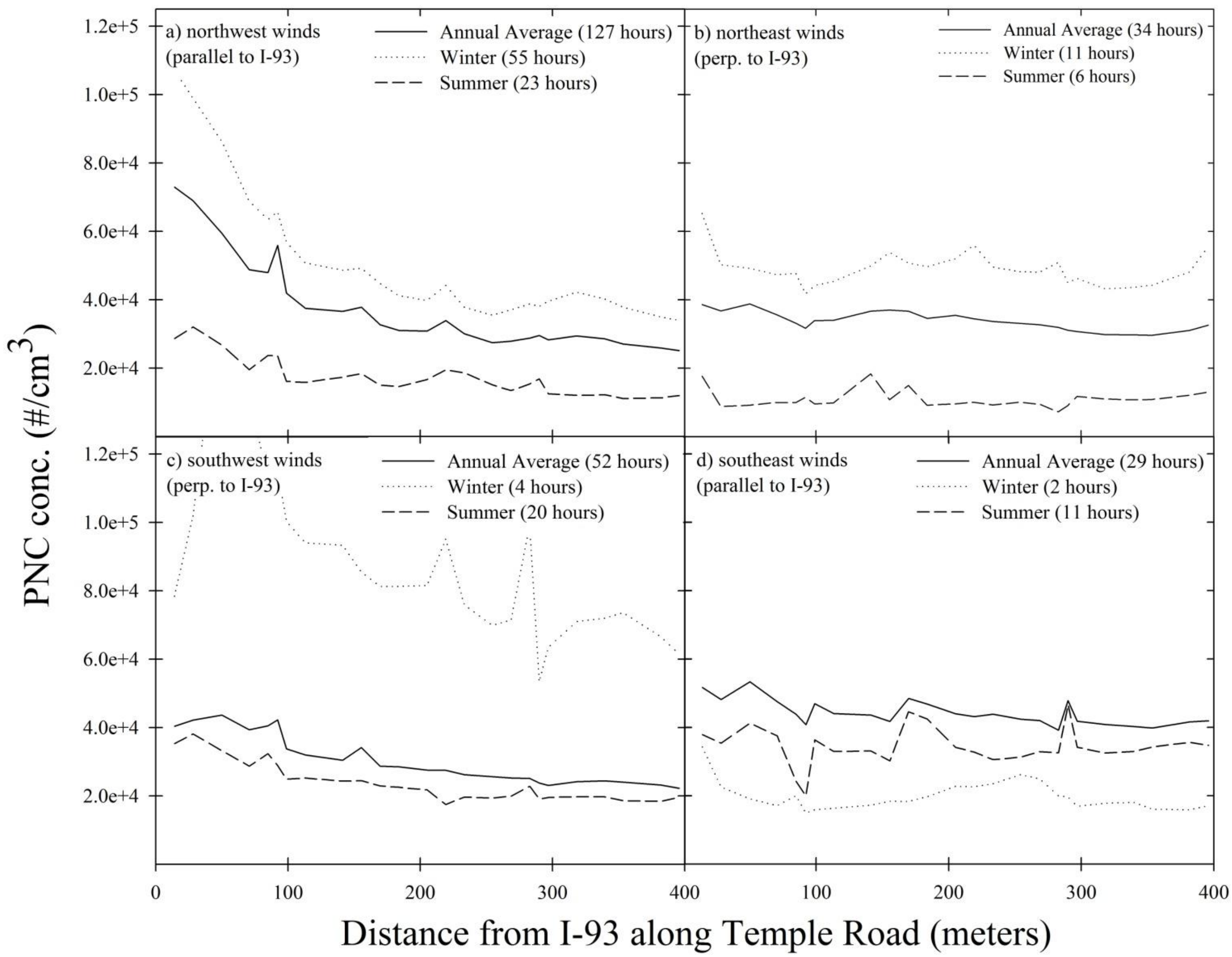


Figure 6. Measured UFP concentrations from hours with prevailing winds from the a) northwest (270^o - 0^o), b) northeast (0^o - 90^o), c) southwest (180^o - 270^o), and d) southeast (90^o - 180^o).

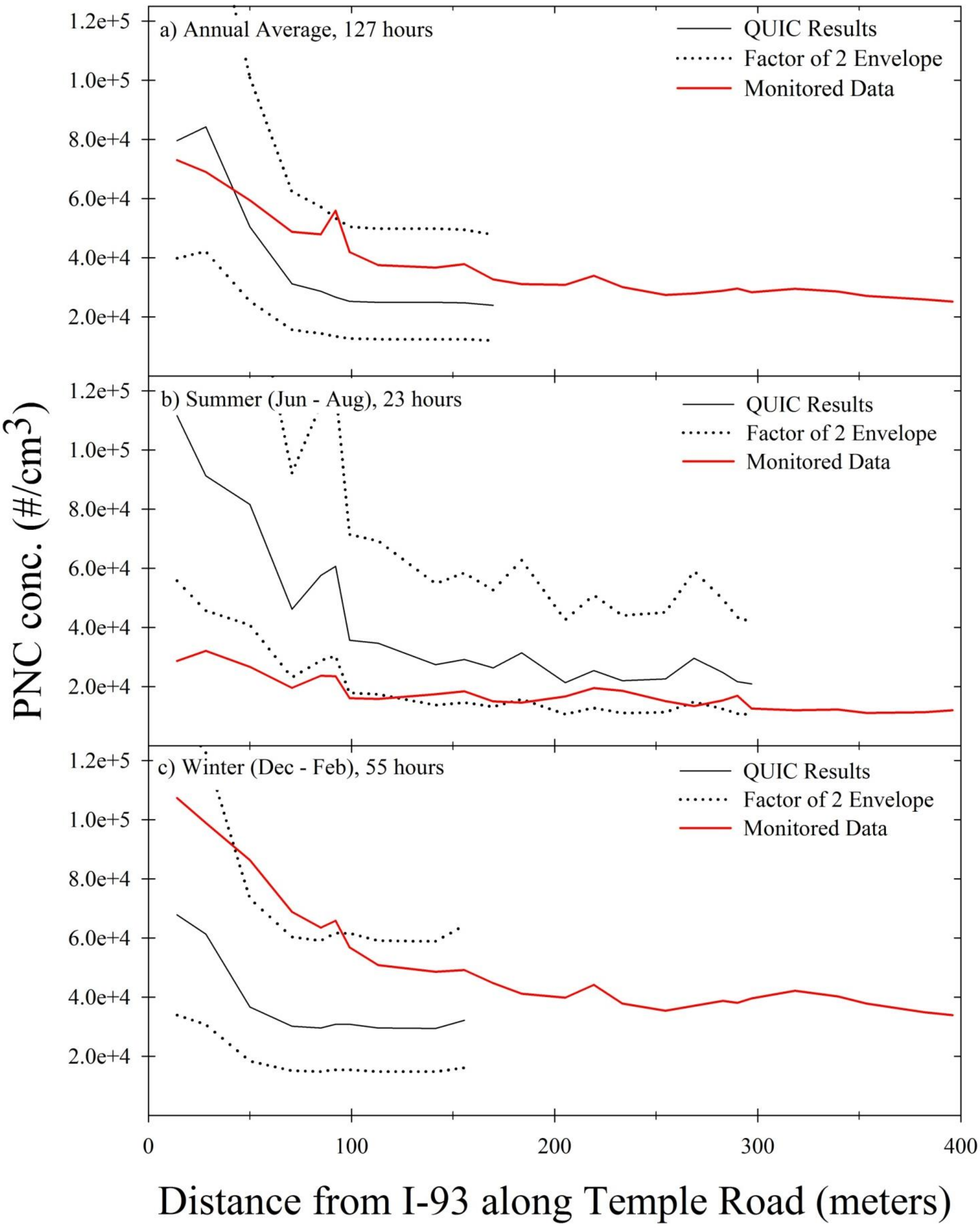


Figure 7. QUIC model results vs. measured data on days with prevailing northwest winds, with an EF of $5*10^{14}$ #/veh-km.

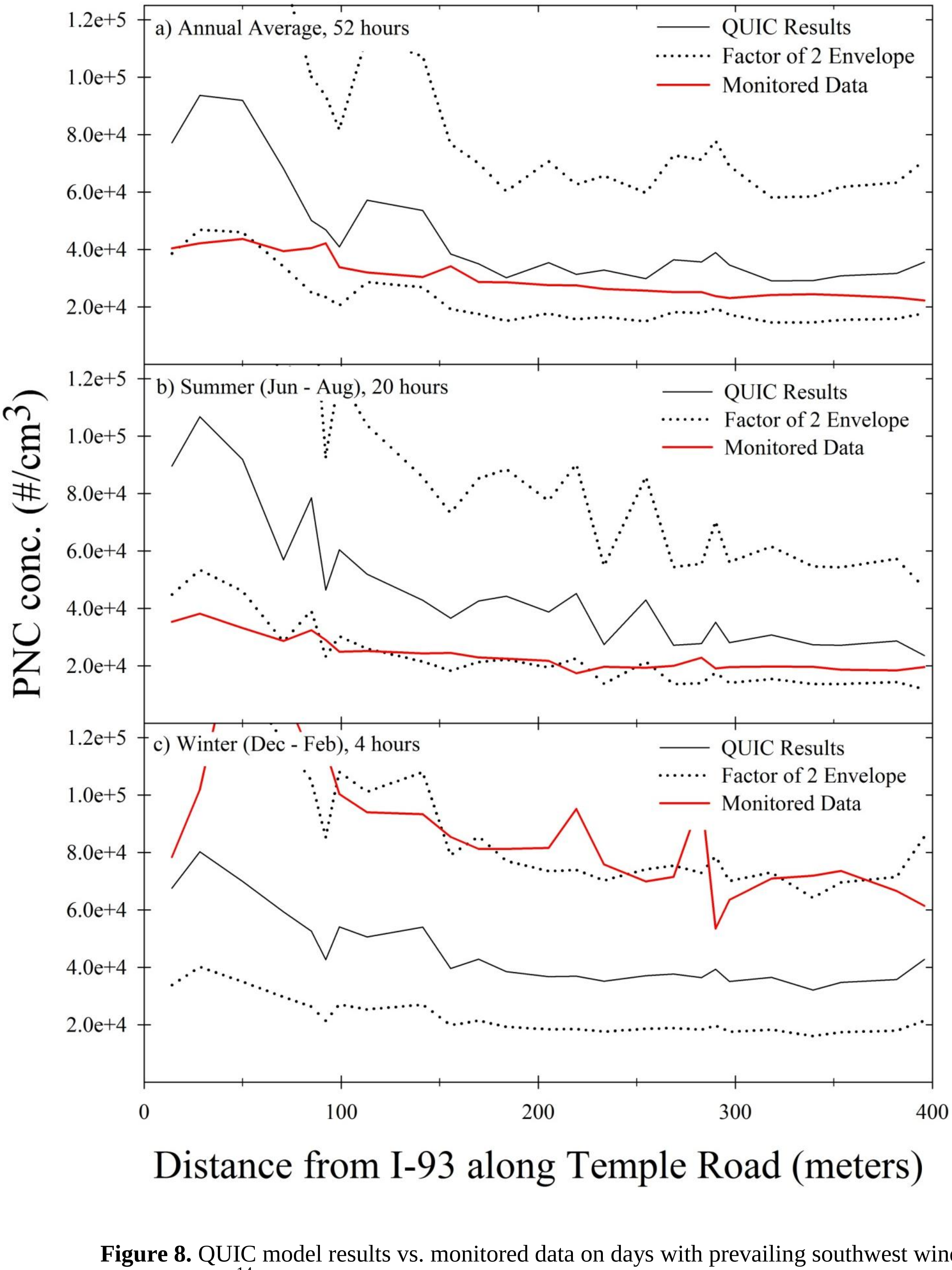


Figure 8. QUIC model results vs. monitored data on days with prevailing southwest winds, with an EF of $5*10^{14}$ #/veh-km.

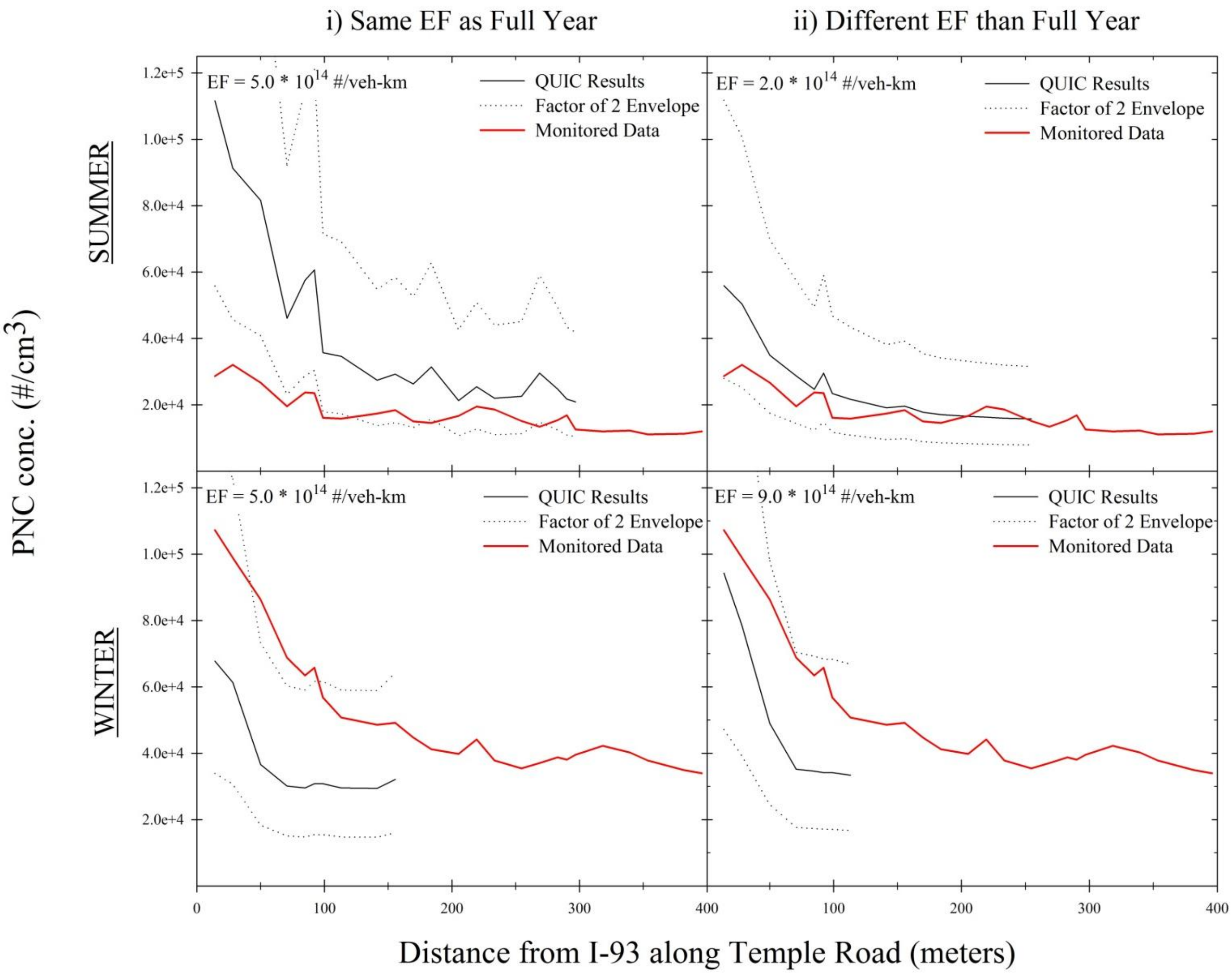


Figure 9. QUIC model results vs. monitored data on days with prevailing northwest winds, with i) the same EF as was used to estimate annual average data ($5*10^{14}$ #/veh-km) or ii) a different EF as was used for annual average data ($2*10^{14}$ #/veh-km and $9*10^{14}$ #/veh-km for summer and winter hours respectively).

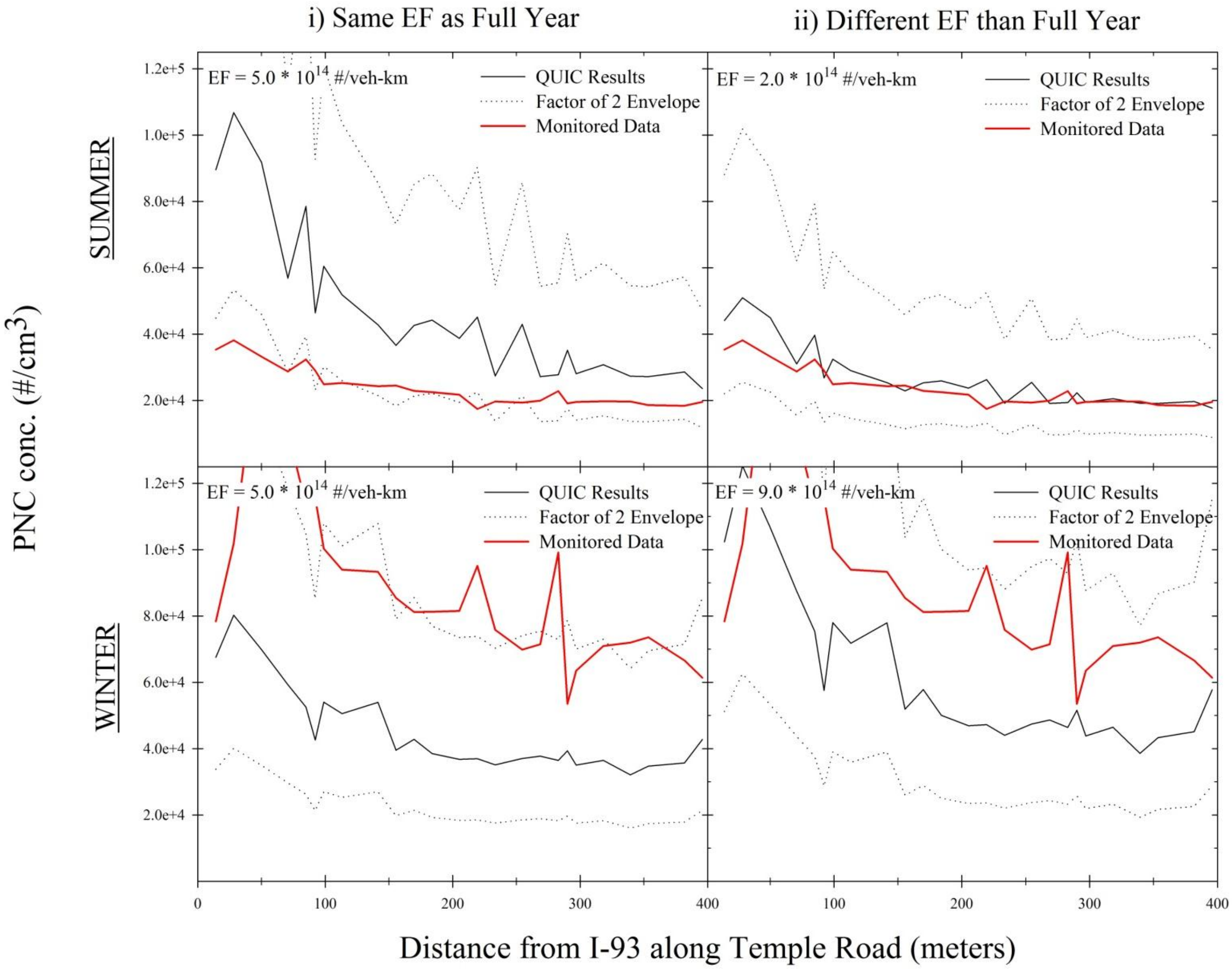


Figure 10. QUIC model results vs. monitored data on days with prevailing southwest winds, with i) the same EF as was used to estimate annual average data ($5*10^{14}$ #/veh-km) or ii) a different EF as was used for annual average data ($2.5*10^{14}$ #/veh-km and $9*10^{14}$ #/veh-km for summer and winter hours respectively).

Table 1. QUIC Model Results

Wind Direction	NORTHWEST (270^o - 360^o)[a]					SOUTHWEST (180^o - 270^o)[a]				
Season[b]	Annual Average	Summer (Jun - Aug)		Winter (Dec - Feb)		Annual Average	Summer (Jun - Aug)		Winter (Dec - Feb)	
Hours	127	23		55		52	20		4	
PNC Bkgrnd (#/cm^3)	17,808	13,730		24,010		17,808	13,730		24,010	
Quic Inputs										
avg. wind dir. (deg)	310	299		314		246	237		254	
avg. wind speed (m/s)	4.2	4.5		4.9		3.7	3.6		4.1	
avg. Temp. (K)	280.1	295.3		271.7		289.3	296.6		266.9	
avg. Humidity (%)	59	60		61		69	69		53	
Tot. # of veh (% of Tot.)	1,071,281	203,142 (19%)		476,979 (45%)		428,641	167,941 (39%)		28,169 (7%)	
EF (#/veh-km)	5E+14	5E+14	2E+14	5E+14	9E+14	5E+14	5E+14	2E+14	5E+14	9E+14
Source Term (mg/s)	490.7	513.8	205.5	504.5	908.2	479.6	488.5	195.4	409.7	737.5
QUIC vs Data										
Nash-Sutcliffe (real)	-0.460	-33.786	-2.285	-1.783	-0.973	-6.942	-24.107	0.143	-2.617	-0.795
Nash-Sutcliffe (log)	-1.648	-7.552	-0.419	-4.532	-3.165	-2.323	-7.679	0.389	-6.591	-2.108
Correl. Coeff. (R)	0.893	0.908	0.888	0.891	0.938	0.824	0.919	0.919	0.623	0.623
Coeff. of Residuals	0.248	1.135	0.393	0.151	0.114	0.455	0.740	0.186	0.228	0.239
% Bias	-21.490	116.622	26.806	-45.676	-34.244	46.970	94.211	12.228	-48.834	-29.690
R^2	0.797	0.824	0.789	0.794	0.879	0.680	0.845	0.845	0.389	0.389
%Inside Envelope	91%	50%	100%	60%	100%	92%	68%	100%	52%	88%

NOTES:

[a] due north is 360^o, due south is 180^o

[b] data was collected in 2009 and 2010

APPENDIX A - TAPL Dataset Sample

sgmt_ID	pnc_geomean	disti93	Hwy_ Direction	Hwy_ Side	year	day	hour	wsmph	wsmps	wdir	TempF	Humidity
16067	17175.86	233.4523	225	East	2009	261	5	3	1.4	269	49.5	89
16067	24564.82	233.4523	225	East	2009	261	6	7	3.1	262	50.1	89
16067	23602.89	233.4523	225	East	2009	261	7	10	4.4	262	51.9	88
16067	15366.66	233.4523	225	East	2009	261	8	15	6.5	280	55.9	82
16067	30356.55	233.4523	225	East	2009	261	9	13	6	270	59.9	76
16067	9551.757	233.4523	225	East	2009	261	10	15	6.5	287	63.6	67
16067	20279.99	233.4523	225	East	2009	263	7	1	0.6	291	46.1	83
16067	16731.54	233.4523	225	East	2009	263	9	1	0.5	276	56.3	65
16067	11705.46	233.4523	225	East	2009	263	11	7	3	257	63.6	49
16067	19236.06	233.4523	225	East	2009	265	14	14	6.2	247	74.2	68
16067	14634.81	233.4523	225	East	2009	265	15	11	4.9	256	73.8	68
16067	17679.63	233.4523	225	East	2009	265	17	10	4.7	240	72.6	71
16067	19049.12	233.4523	225	East	2009	265	19	10	4.6	234	70.5	75
16067	11926.4	233.4523	225	East	2009	281	14	13	6	331	60	52
16067	5941.252	233.4523	225	East	2009	281	16	13	6	312	60.8	51
16067	5815.307	233.4523	225	East	2009	281	17	6	2.9	322	60.1	54
16067	20360.65	233.4523	225	East	2009	281	18	3	1.6	308	58.7	60
16067	24135.03	233.4523	225	East	2009	281	19	5	2	331	57.1	65
16067	6880.839	233.4523	225	East	2009	290	14	12	5.4	39	46.4	68

APPENDIX B - Macro GUI

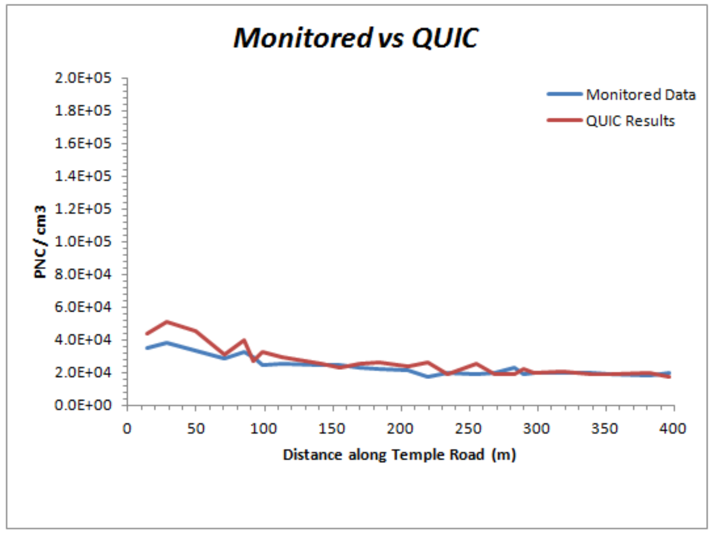
GUI

Import All Data | Clear All Data | Run Macro | Compare to QUIC results? YES | Custom Graph Limits? YES | Copy Results

Input File: summer_04

Background: 13,730 PNC/cm^3

Years	Hours	Days
2009	5	1-Jun
2010	6	8-Jun
	7	15-Jun
	8	23-Jun
	9	29-Jun
	10	13-Jul
	11	20-Jul
	12	28-Jul
	13	30-Jul
	14	2-Aug
	15	12-Aug
	16	25-Aug
	17	
	18	
	19	
	20	



Statistics

Nash-Sutcliffe (real)	0.143
Nash-Sutcliffe (log)	0.389
Correl. Coeff. (R)	0.919
Coeff. of Residuals	0.186
% Bias	12.228
R^2	0.845
%Inside	100.0%

Calculations

Emission Factor	2.0E+14	#/veh-km
particle diameter	0.0000001	meters
particle density	1000000	gram/m^3
gram/1 particle	5.23599E-16	gram/#
source length	800	m
Particles released	201600	#
Number of vehicles	167941	vehicles
Time	72000	seconds

Source Term

195.4 mg/s

MET Parameters

wind dir	237	
wind spd	3.6	mps
Temp	296.6	K
Humidity	69	%
#veh	167941	
hours	20	
UpperT	296.5	

Custom Graph Limits

Upper	0
Lower	200,000

Wind Parameters

wind dir	225
+/-	45

APPENDIX C - Sample EF Unit Conversion (from a standard EF unit to mass/time)

$$\mathrm{EF} = 5 * 10^{14}\ \frac{\#}{\mathrm{vehicle} * \mathrm{km}}$$

$$\mathrm{L} = 800\ \mathrm{m} = 0.800\ \mathrm{km}$$

$$\mathrm{D} = 1\mu\mathrm{m} = \ 1 * 10^{-11}\mathrm{km}$$

$$\mathrm{V} = 167{,}941\ \mathrm{vehicles}$$

$$\rho_{\mathrm{UFP}} = 1\,\frac{\mathrm{gram}}{\mathrm{cm}^3} = \ 1 * 10^{15}\,\frac{\mathrm{gram}}{\mathrm{km}^3}$$

$$\mathrm{EF} * \mathrm{L} * \ \mathrm{V} * \left(\frac{4}{3}\pi\left(\frac{\mathrm{D}_{\mathrm{UFP}}}{2}\right)^3\right) * \rho_{\mathrm{UFP}} * \frac{1\ \mathrm{hour}}{3600\ \mathrm{sec}} = \frac{Mass}{Time}\ \left[\frac{gram}{sec}\right] = 0.195\,\frac{\mathrm{g}}{\mathrm{s}} = 195.4\,\frac{\mathrm{mg}}{\mathrm{s}}$$