Assessing the Suitability of Multiple Dispersion and Land Use Regression Models for Urban Traffic-Related Ultrafine Particles

Allison P. Patton,*,[†,‡] Chad Milando,[‡,§] John L. Durant,[‡] and Prashant Kumar[∥,⊥]

[†]Environmental and Occupational Health Sciences Institute, Rutgers University, Piscataway, New Jersey 08901, United States

[‡]Department of Civil and Environmental Engineering, Tufts University, Medford, Massachusetts 02155, United States

[§]Department of Environmental Health Sciences, School of Public Health, University of Michigan, Ann Arbor, Michigan 48109, United States

[∥]Department of Civil and Environmental Engineering, Faculty of Engineering and Physical Sciences (FEPS), University of Surrey, Guildford GU2 7XH, Surrey United Kingdom

[⊥]Environmental Flow (EnFlo) Research Centre, FEPS, University of Surrey, Guildford GU2 7XH, United Kingdom

S Supporting Information

ABSTRACT: Comparative evaluations are needed to assess the suitability of near-road air pollution models for traffic-related ultrafine particle number concentration (PNC). Our goal was to evaluate the ability of dispersion (CALINE4, AERMOD, R-LINE, and QUIC) and regression models to predict PNC in a residential neighborhood (Somerville) and an urban center (Chinatown) near highways in and near Boston, Massachusetts. PNC was measured in each area, and models were compared to each other and measurements for hot (>18 °C) and cold (<10 °C) hours with wind directions parallel to and perpendicular downwind from highways. In Somerville, correlation and error statistics were typically acceptable, and all models predicted concentration gradients extending ∼100 m from the highway. In contrast, in Chinatown, PNC trends differed among models,

and predictions were poorly correlated with measurements likely due to effects of street canyons and nonhighway particle sources. Our results demonstrate the importance of selecting PNC models that align with study area characteristics (e.g., dominant sources and building geometry). We applied widely available models to typical urban study areas; therefore, our results should be generalizable to models of hourly averaged PNC in similar urban areas.
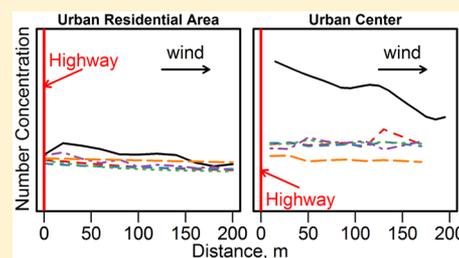
INTRODUCTION

People living within several hundred meters of highways experience increased risks of respiratory and cardiovascular disease,[1,2] and ultrafine particles (UFP; < 100 nm in diameter) emitted in motor vehicle exhaust may contribute to these risks.[3] UFPs have been shown to be associated with increased levels of inflammatory blood biomarkers in people living <500 m from major highways,[4,5] and UFP concentrations near highways can be twice as high as urban background concentrations.[6−9]

Potential exposures to UFP measured as particle number concentrations (PNC) have been quantified using mechanistic[10−12] and empirical[13,14] models across cities,[15−19] in urban street canyons,[20] and near roads.[22−25] Mechanistic models are based on physical theory and include dispersion models like the California Line Source Dispersion Model (CALINE4),[26] the American Meteorological Society/Environmental Protection Agency Regulatory Model (AERMOD),[41−44] the research line source model (R-LINE),[21] and the Quick Urban and Industrial Complex (QUIC) Modeling System.[28] Empirical models of PNC and other traffic-related air pollutants are often developed using land use regression (LUR), a technique that statistically relates pollutant measurements to road density, distance to roads and other variables.[13,22,23,25,29] While dispersion models

require detailed meteorological and traffic inputs and are broadly generalizable, regression models are based on monitoring data and are location-specific.[13,23,29]

Both dispersion and regression models contain uncertainties related to model structure assumptions (e.g., dispersion and chemical reactivity) and parameter value accuracy (e.g., meteorological data and emission factors).[12] For PNC, the structure of model treatment of dispersion is likely to be more important than inclusion of chemistry. While particle coagulation may reduce PNC by 25% over an entire city (∼1000 km),[15] dilution is expected to have a larger impact on PNC than coagulation or other reactions (e.g., evaporation, photooxidation) at the neighborhood scale.[12] At the same time, the particle emission rate is a key parametric uncertainty in PNC modeling because the emissions depend on the vehicle fleet, meteorology, and rapid transformation of emitted particles on and within a few meters of the road.[12,30] Development of locally suitable PNC emission factors could

Table 1. Models Compared in This Study

model	CALINE4	R-LINE	AERMOD	QUIC	regression
model type	Gaussian plume	Gaussian plume	Gaussian plume	Lagrangian plume	Multivariate regression
road geometry[a]	Line	Line	Area/Volume	line	line
atmospheric stability	Pasquil-Gifford	Monin-Obukhov	Monin-Obukhov	power-law	wind speed
spatial resolution[b]	0.5 m	0.5 m	0.5 m	10 m × 10 m × 10 m	20 m
approximate runtime[c]	200 ms	500 ms	500 ms	15 min	5 ms
Data Requirements					
pollutant emission rate	yes	yes	yes	yes	if desired[d]
surface meteorology	yes	yes	yes	yes	if desired[d]
upper air meteorology	no	yes	yes	no	if desired[d]
building footprints	no	no	no	yes	if desired[d]

[a]The AERMOD area geometry was used for this model comparison. The spatial-temporal regression models include road location but not source strength. [b]Spatial resolutions were defined by GIS input data resolution (CALINE4, R-LINE, and AERMOD),[52] user-defined grids (QUIC), and measurement resolution (regression). [c]Runtimes are the time to run Chinatown Scenario 1 on a Dell Precision T3600 with an Intel Xeon Processor E5−1620 (Four Core 3.6 GHz, 10M, Turbo) CPU, 2.0GB AMD FirePro V5900 GPU, and 16 GB of 1600 MHz DDR3 RAM running 64-bit Windows 7. [d]Regression models can use any available data correlated with air pollution. The models used in this paper incorporate traffic volumes (used in calculating emission rates) and surface meteorology, but not upper air meteorology or building locations and dimensions.

improve the performance of both regression and dispersion models.[12]

Comparative performance evaluations demonstrate the magnitude of uncertainties in estimated concentrations that are introduced by modeling decisions. In different European studies of NO_2, one component of traffic exhaust, performance (R^2 and standard error) of dispersion models was similar,[31−33] worse than[34,35] or better than[36] LUR performance relative to measurements. At different traffic sites, LUR and dispersion models either underestimated[31,32] or overestimated[37] air pollutant concentrations. Within most single studies, correlation coefficients (R^2) between NO_2 predicted by dispersion models and LUR, or two dispersion models, ranged from 0.55 to 0.90.[32,33,35,36,38] However, in one study comparing LUR to European regulatory dispersion models, the agreement between NO_2 models varied widely (R^2 range = 0.19−0.89) and was lowest for comparisons using the least spatially resolved (>500 m grids) dispersion models.[39] In the near-road environment, slight improvements were obtained by modeling plume meander and vehicle- and road-induced turbulence.[37,40] One near-road study using both a dispersion model (QUIC) and LUR to model PNC reported R^2 = 0.8 between QUIC and LUR, although model performance was not evaluated.[24] To our knowledge, there are no studies in the literature comparing performance of dispersion and regression models of PNC near roads. Therefore, we undertook the present study to evaluate how differences in model structure and inputs affect UFP concentrations predicted by near-road models.

The goals of this work were to evaluate the ability of line source dispersion and land use regression models to predict hourly PNC near busy roads in urban neighborhoods, and to provide insight about which kinds of models should be used for near-road PNC exposure assessment. We compared three Gaussian dispersion models (CALINE4, AERMOD and R-LINE), a Lagrangian dispersion model with empirical flow approximations (QUIC) and neighborhood-specific spatial-temporal regression models (Table 1). Our specific objectives were to (i) compare distance-decay gradients predicted under different wind direction and air temperature scenarios in two urban near-highway neighborhoods to determine which models generate reasonable PNC predictions, and (ii) evaluate and compare the performance of the models in predicting PNC relative to measurements and each other.

■ MATERIALS AND METHODS

Models. *CALINE4.* CALINE4 was developed in the 1970s by the California Department of Transportation to assess the impact of road vehicles on air quality and is an updated version of a United States Environmental Protection Agency (U.S. EPA) regulatory model.[26,41] The main advantages of CALINE4 are its ease of use and relatively few inputs. CALINE4 uses an analytical solution to a steady-state Gaussian plume model to predict pollutant concentrations. Vehicle-induced turbulence is modeled by a mixing zone 2 m wider than the road surface. Dispersion outside the mixing zone is modeled using Pasquil-Gifford stability curves.

AERMOD. AERMOD (v8.1.0, Lakes Environmental) is a regulatory model that was developed in the 2000s by the American Meteorological Society and the U.S. EPA to simulate industrial source air quality effects.[41−43] The main advantage of AERMOD over earlier models like CALINE4 is the improved parametrization of dispersion; however, AERMOD also requires more meteorological inputs (e.g., Monin-Obukhov length) to support the more complex dispersion algorithms. AERMOD is a steady-state plume model that incorporates Gaussian dispersion and the Plume Rise Model Enhancement (PRIME) algorithms.[42] Although AERMOD was developed for industrial point sources, it has previously been used to evaluate the effects of roads on local air quality.[37,45] AERMOD can treat line sources as a series of point or volume sources or as an area source. In this model comparison, interstate highways were modeled as area sources.

R-LINE. R-LINE (v1.2) was developed by the U.S. EPA in the 2010s for predicting mobile-source air quality impacts near roadways.[21,46] The main advantage of R-LINE is that it incorporates the advanced dispersion algorithms used in AERMOD into a line source model similar to CALINE. In R-LINE, roads are input as lines and simulated as a series of point sources. R-LINE can use analytical (used in this work) or numerical methods to predict hourly concentrations of inert traffic-related air pollutants.

QUIC. QUIC (v6.01) was developed by the Los Alamos National Laboratory and has been continuously updated since 1990 to model air pollutant releases in urban areas.[47−49] QUIC is the only model considered here that explicitly models individual obstructions, and therefore requires greater computational resources than the other models. The wind-field module
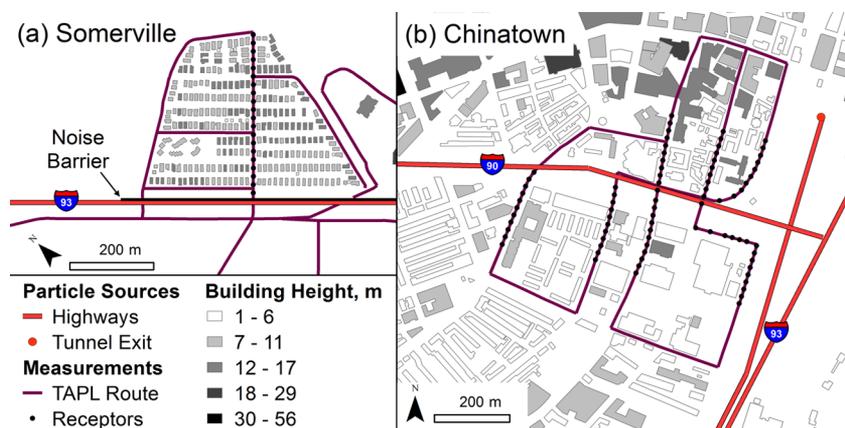
Figure 1. Somerville (a) and Chinatown (b) study areas with particle sources (highways and tunnel exit), Tufts Air Pollution Monitoring Laboratory (TAPL) routes and model receptors, and heights of buildings modeled in QUIC. Receptors represent the locations where the model predictions and measurements were compared. The building shapefile was obtained via the Tufts University GIS data server.[59]

Table 2. Meteorological and Traffic Input Conditions for Test Scenarios in Somerville and Chinatown

|	Somerville (2009−2010)[a]				Chinatown (2011−2012)[b]					
scenario	SV-1	SV-2	SV-3	SV-4	CT-1	CT-2	CT-3	CT-4	CT-5	CT-6
mobile monitoring data, min	13.5	11.3	7.8	15.8	31.1	40.6	30.5	31.7	29.4	15.5
Meteorology										
temperature, °C	−8.8	−5.6	28.1	30.6	7.8	9.4	22.8	19.4	−1.2	18.9
wind direction, °	239	334	249	301	75	202	202	87	352	342
relative to I-93[c]	⊥	‖	⊥	‖	⊥	‖	‖	⊥	‖	‖
relative to I-90[c]					‖	⊥	⊥	‖	⊥	⊥
wind speed, m/s	1.6	2.3	4.7	5.4	1.7	1.7	2.2	3.6	1.5	1.2
wind power law exponent p	0.15	0.15	0.07	0.15	0.30	0.30	0.20	0.15	0.30	0.15
stability class	D	D	B	D	F	E	C	B	E	B
mixing height, m	750	750	1500	750	1230	1750	3640	1100	610	1520
Traffic Emissions										
volume on I-93, veh h^{-1}	4551	9020	8701	9725	10315	10315	6737	9351	6568	10426
volume on I-90, veh h^{-1}					9634	9634	3320	5756	7698	6605
EF$_{PN}$, 10^{14} # veh^{-1} km^{-1}	2.3	2.3	0.54	0.54	1.48	1.42	0.99	1.09	1.88	1.10

[a]Scenarios for Somerville are SV-1 = Saturday December 19 (day 353), 2009 0700−0800; SV-2 = Saturday December 19 (day 353), 2009 1100−1200; SV-3 = Thursday June 24 (day 175), 2010 1000−1100; SV-4 = Thursday July 29 (day 210), 2010 1700−1800. [b]Scenarios for Chinatown are CT-1 = Wednesday April 12 (day 102), 2012 1800−1900; CT-2 = Tuesday January 24 (day 24), 2012 0500−0600; CT-3 = Tuesday June 12 (day 164), 2012 1200−1300; CT-4 = Friday September 2 (day 254), 2011 0900−1000; CT-5 = Wednesday January 11 (day 11), 2012 0600−0700; CT-6 = Friday September 9 (day 252), 2011 0700−0800. [c]Perpendicular downwind scenarios (⊥) were selected from those days with wind within 22 degrees of perpendicular from I-93 or I-90. Parallel wind scenarios (‖) were selected from those days with wind within 22 degrees of parallel from I-93 or I-90.

calculates three-dimensional flow fields around stationary obstacles including buildings, hills, and vegetation.[49] Subsequently, a Lagrangian random-walk dispersion model superimposes a pollution source on the wind field and tracks the dispersion of pollutants downwind of the source.[47] Although QUIC can simulate particle dynamics, particles were considered inert for this study.

Regression. Multivariate linear regression (land use regression) models of air pollution empirically relate measured pollutant concentrations to covariates including traffic volume, distance and direction to roads, and meteorology.[13,29] The hourly neighborhood-specific models of the natural logarithm of PNC evaluated in this study were developed for the Community Assessment of Freeway Exposure and Health (CAFEH), and are described elsewhere.[22,23] Briefly, the CAFEH models were spatial-temporal regressions developed using 1-s mobile monitoring measurements collected over the course of a year in each study area (see Model Inputs: Field Measurements). Land use, meteorological, and traffic variables

were added to the models if they had a plausible physical relationship to PNC and increased R^2 by >1%. Temporal variables in the final models included temperature, wind speed and wind direction. Spatial patterns were described by distance from I-93 and major intersections (but not distance from I-90) and by wind direction relative to I-93 and major nonroad sources (e.g., airport and train station; Supporting Information (SI) Table S1).[22,23] We did not have an independent data set to evaluate the models; however, no single hour of measurements substantially affected the regression models' performance in leave-one-out cross-validation.[22,23]

Study Area(s). The five models were compared in 0.2-km^2 areas <400 m from the edge of interstate highways in two contrasting neighborhoods in the Boston, Massachusetts, metropolitan area: the Ten Hills neighborhood in Somerville, and Chinatown in downtown Boston (Figure 1). In both areas, the fleet was 95−99% gasoline vehicles and 1−5% diesel vehicles depending on the day of the week and time of day.[50,51] The Ten Hills neighborhood is bordered by Interstate Highway
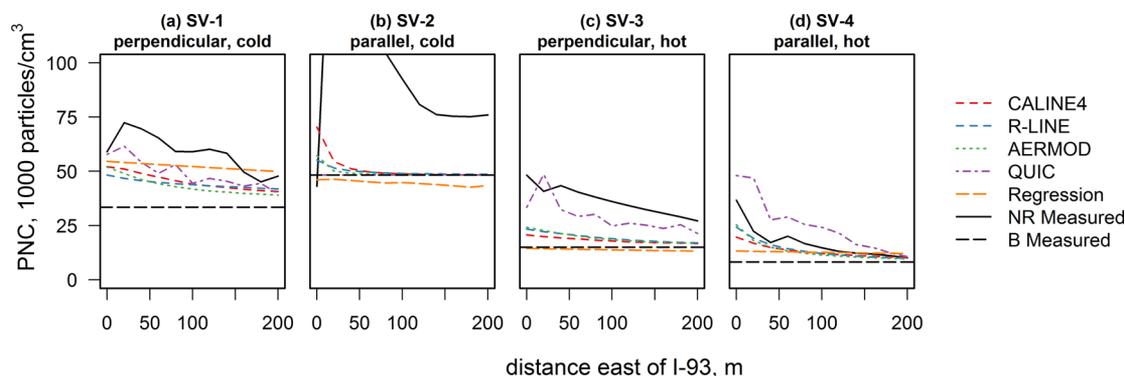
Figure 2. PNC-distance plots up to 200 m from the edge of I-93 in Somerville predicted by CALINE4, R-LINE, AERMOD, QUIC, and the CAFEH regression model. The four scenarios are for wind directions relative to I-93 and hot or cold air temperatures, as listed in the panels and described in Table 2. Smoothed near-road measurements (NR Measured) and background concentrations (B Measured) are shown for comparison. Values in (b) above the scale limit are 211, 135, 119, and 105×10^3 particles/cm³ at distances of 20, 40, 60, and 80 m, respectively, from the edge of I-93.

93 (I-93; 150 000 veh day^{-1}) and Massachusetts Route 38 to the southwest, Massachusetts Route 28 to the east, and the Mystic River to the north. Ten Hills is characterized by rectilinear blocks of 2 and 3 family homes (276 buildings with 10 m average height). Additionally, I-93 is elevated 5 m above grade with a 3-m-high noise barrier between I-93 and Ten Hills. The urban center of Boston Chinatown is bordered by I-93 to the east and bisected by I-90 (90 000 veh day^{-1}) from the east to west. Chinatown is characterized by street canyons lined with residential and commercial buildings up to 100 m tall (824 in total). I-93 emerges from the Central Artery Tunnel just north of the Chinatown study area; I-90 is mostly below-grade (≤5 m) as it passes through Chinatown.

Modeling Scenarios. *Highway Geometry and Emissions Inputs.* Highway locations were obtained from MassGIS.[52] In Somerville, I-93 was modeled as a line source 5 m above ground level. In QUIC, the elevated surface of I-93 was modeled as a 1-m thick block above 4 m of air, preventing particle transport downward through the road surface. The noise barrier was modeled as a 3-m-tall solid structure on the northeastern edge of the elevated highway surface. The highways in Chinatown were simulated as ground level sources. Highway traffic volumes and speeds were obtained from the Massachusetts Department of Transportation.[53] Dispersion models were run with unit emission factors and predictions were scaled by particle number emission factors (PN$_{EF}$; #-veh^{-1}km^{-1}) obtained from measurements on I-93 in the Central Artery Tunnel in Boston (see SI Section S1).[54] Each model treated particles as chemically inert and assumed that contributions of other PNC sources were negligible relative to the contributions of I-93 and I-90.

Meteorology Inputs. The models were tested for a range of meteorological conditions defined by wind direction and temperature (Table 2). The wind direction was parallel or perpendicular downwind relative to I-93 (both areas) and I-90 (Chinatown only). For each wind condition, one hot hour (>18 °C) and one cold hour (<10 °C) were selected. Surface meteorological measurements were obtained from Logan International Airport (KBOS).[55] Upper air data from balloon soundings (16 m−331 000 m) were obtained from Chatham, MA (CHH-74494). Pasquill−Gifford atmospheric stability class and mixing height were assigned using the Turner workbook and Monin−Obhukhov length was calculated with AERMET v12345.[56,57] Standard deviation of wind direction, σ_θ, was set to

20° following a sensitivity analysis showing that CALINE4 predictions are not strongly affected by σ_θ.[26]

Building Parameterization. In CALINE4, AERMOD, and R-LINE the aerodynamic roughness of the ground surface was assigned the CALINE4 default for each neighborhood: for Somerville, the roughness coefficient was 100 cm (suburban environment) and for Chinatown, the roughness coefficient was 400 cm (central business district).[58] For QUIC, building footprints and heights were obtained from a shapefile based on LIDAR measurements[59] and a building wall roughness, z_0, of 0.1 m was assumed.[20] The regression models did not consider building geometry.

Modeling Domain, Resolution, and Receptors. To avoid inaccurate dispersion model predictions due to changes in wind flow near model domain edges, the horizontal domains extended outside the study area by 50 m in Somerville and 500 m in Chinatown (i.e., 5 times the average building height).[20] The domain heights were 100 m in Somerville and 200 m in Chinatown. In QUIC, wind fields were resolved to 1 m in each horizontal direction, x and y; the vertical (z) resolution was 1 m at the surface and increased parabolically with elevation. Pollutant concentrations were calculated by QUIC on a 10 m × 10 m × 10 m three-dimensional grid. CALINE4, R-LINE, AERMOD, and the regression models were spatially continuous (i.e., nongrid) and therefore spatial resolution in the dispersion models was limited by accuracy in the GIS layer files and spatial resolution in the regression models was limited by the PNC measurements. Modeling results were exported to a 20-receptor transect in Somerville and an 80-receptor grid in Chinatown. Receptors were ~20 m apart at distances from 0 to 400 m from the edges of highways and 3 m above ground level.

Field Measurements. Mobile monitoring of PNC < 500 m (near-highway) and >1000 m (urban background) from the edge of I-93 was conducted with the Tufts Air Pollution Monitoring Laboratory (TAPL). Monitoring was conducted by driving the TAPL on fixed routes in Somerville on 43 days (September 2009 - August 2010) and in Chinatown on 47 days (August 2011 to July 2012).[7,8] PNC was measured each second with a condensation particle counter (CPC 3775, TSI, Shoreview, MN), assigned a location by matching the CPC time with that of a Garmin GPS V receiver, and quality controlled following standard CAFEH procedures.[7,8] To reduce noise in the measurements for comparative model performance evaluations and increase stability of spatial patterns, a loess
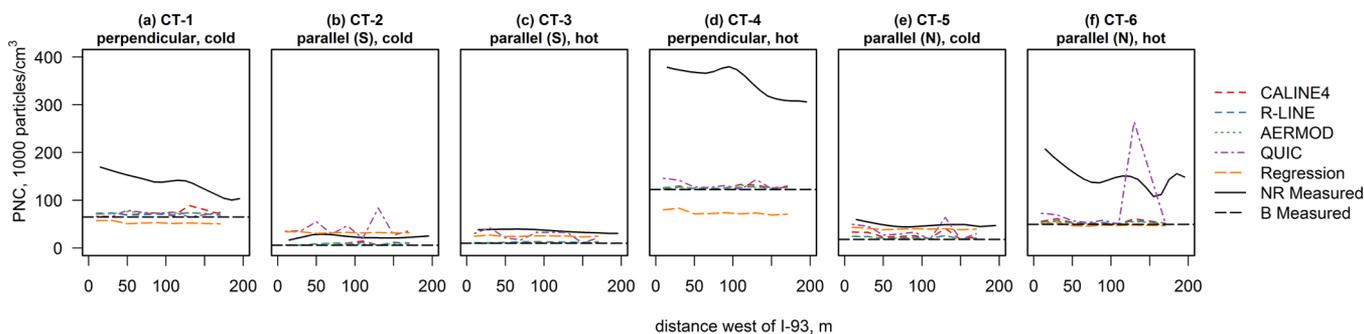
Figure 3. PNC-distance plots up to 200 m from the edge of I-93 in Chinatown predicted by CALINE4, R-LINE, AERMOD, QUIC, and the CAFEH regression model. The six scenarios are for wind directions relative to I-93 and hot or cold air temperatures, as listed in the panels and described in Table 2. For parallel wind directions, the descriptions include whether the wind was from the north (N) or south (S) across I-90. Smoothed near-road measurements (NR Measured) and background concentrations (B Measured) are shown for comparison. See SI Figure S2 for gradients from I-90.

smooth (span = 0.2 based on previous work[7]) of PNC as a function of distance to the highways was developed for each scenario to assign PNC measurements to receptors. Each near-highway (<400 m) loess smooth was the average of 3−12 near-highway transects. Background PNC in Somerville was calculated as the mean of ~10 min of measurements >1000 m from I-93 during the same hour as near-highway measurements.[7] Comparable background measurements were not available for Chinatown; therefore, background concentrations were estimated as the first percentile of PNC from each hour of monitoring. Sensitivity to this assumption was tested by repeating analyses for Chinatown using the 25th percentile of measurements as background.

Model Performance Metrics. Model predictions were evaluated for 4 h in Somerville (Scenarios SV-1 to SV-4) and 6 h in Chinatown (Scenarios CT-1 to CT-6), for a total of 10 test scenarios (Table 2). The metrics used for model evaluation relative to measurements were correlation coefficient (R^2), fraction of predictions within a factor of 1.5 (FAC1.5) and 2 (FAC2) of measurements, normalized mean square error (NMSE), and fractional bias (FB). These performance measures were calculated with a custom statistics function in R (see Section S2) and have been widely used to evaluate air pollution models.[20,60,61] Model performance relative to measurements was considered acceptable if $R^2 > 0.9$, NMSE ≤ 0.25, absolute value of FB ≤ 0.25, and FAC2 > 0.7.[60,61] In addition, the level of agreement among predictions from different models was assessed using Pearson correlations. All analyses were performed in R version 3.0.1.[62]

■ RESULTS AND DISCUSSION

Model Performance in Somerville. In Somerville, CALINE4, R-LINE, AERMOD, QUIC, and the regression model predicted near-highway PNC gradients that approached background concentrations at ~200 m from the edge of I-93 (Figure 2). In addition, arterial roads began to influence measurements at distances from I-93 greater than ~200 m. Therefore, all model evaluations and comparisons in both Somerville and Chinatown were made from 0 to 200 m from the edges of I-93 and I-90. The models reasonably predicted Somerville scenario measurements (SI Figure S1) for warm hours (Scenarios SV-3 and SV-4) and the cold hour when the wind was perpendicular to I-93 (Scenario SV-1; Table S2). QUIC most closely approximated the shape of the measured PNC distance-decay curves; however, CALINE4, AERMOD,

and R-LINE also performed reasonably well (and outperformed QUIC when the wind was parallel to the highway). Agreement of model predictions with measurements ranged from moderate to acceptable for individual scenarios ($R^2 = 0.43−0.96$, NMSE ≤ 0.22, |FB| = 0.12−0.90; SI Table S2). The Somerville regression model generally performed better for the four scenarios ($R^2 = 0.69, 0.28, 0.96,$ and 0.72) than it did for the full CAFEH data set ($R^2 = 0.42$).[22] An exception to the overall good performance was for the cold hour with wind parallel to I-93 (Scenario SV-2), when a wide zone of elevated PNC near I-93 was not predicted by the models (e.g., $R^2 \leq 0.52$ for all models). During the overcast midday of SV-2, PNC was unusually high both near I-93 (~75 000 particles/cm³) and in the urban background area (~50 000 particles/cm³), suggesting that decreased vertical mixing in the morning contributed to the buildup of PNC.

In Somerville, there was high agreement among models; Pearson's r among model predictions was >0.82 overall and >0.58 for individual scenarios (SI Table S3). QUIC generally predicted the highest PNC, except for Scenario SV-2 (cold air temperature, winds parallel to I-93) when QUIC was unable to predict a near-road gradient. The highest correlations were found for predictions from the three Gaussian dispersion models (CALINE4, AERMOD, and R-LINE), which had similar curves with $r > 0.98$ for all four scenarios. CALINE4 predicted slightly higher PNC than AERMOD and R-LINE near I-93 during cold hours (Scenarios SV-1 and SV-2) and slightly lower concentrations during hot hours (Scenarios SV-3 and SV-4). The regression model predicted lower PNC than the dispersion models except for when the wind was blowing perpendicular to I-93 from the west on a cold day (Scenario SV-1). Low correlations were found for QUIC with all other models for Scenario SV-3 ($r = 0.78−0.80$), and for the regression model with other models for Scenario SV-2 ($r = 0.63$ for CALINE4, 0.75 for R-LINE, and 0.58 for AERMOD). However, all the models had generally acceptable performance and could be applied to neighborhoods like Somerville.

Model Performance in Chinatown. On average, the models predicted weak near-highway concentration gradients extending 100−200 m downwind (west) of the edges of I-93 (Figure 3) and (north and south of) I-90 (SI Figure S2) in Chinatown. Gradients from the individual highways were more difficult to discern than those in Somerville. Complexities in spatial trends in Chinatown were not accurately captured due to the generally high background PNC and contributions from

multiple highways. All the models underestimated measured PNC in most Chinatown scenarios and underestimated the range in PNC upwind and downwind of I-93 and I-90 for warm and cold hours (Scenarios CT-1 to CT-6; Figure S3). R^2 was ≤0.45 and NMSE was ≤0.26 for all five models under all individual Chinatown test scenarios, and FAC2 (≤70% for all dispersion models) and FB (−1.21 to 0.68) were outside of generally accepted standards (SI Table S4). The performance of the Chinatown regression model for the six test scenarios (R^2 = 0.37, 0.00, 0.00, 0.26, 0.43, and 0.36) varied substantially compared to the model performance in the CAFEH data set as a whole (R^2 = 0.24).[23] The poor performance of the models in Chinatown reflected the inability to capture the more complex spatial patterns in PNC. In sensitivity analyses, changes to increase the loess smooth span (SI Figure S4, Figure S5) and the assumed background PNC (SI Figure S6) improved the fraction of predictions within a factor of 2 and 1.5 and the fractional bias (SI Table S5). Neither the R^2 nor the NMSE was affected by these adjustments because the changes did not substantially affect the spatial trends in PNC. Similar results were obtained when only those receptors downwind of I-90 were considered. While removing receptors upwind of I-90 improved FAC2 and FAC1.5 by ~12% on average (SI Table S6), correlations between measurements and PNC predictions at downwind receptors were not generally better than those using the full set of receptors (SI Figure S7).

PNC predictions from the five models had less agreement in Chinatown than in Somerville (SI Table S7). During cold hours correlations among predictions were as low as 0.57 between CALINE4 and AERMOD (Scenario CT-1: east wind perpendicular to I-93) and 0.77 between CALINE4 and R-LINE (Scenario CT-5: north wind perpendicular to I-90). Surprisingly, QUIC and the regression model sometimes predicted trends in the opposite direction from the other models. For example, during cold (Scenario CT-2) and hot (Scenario CT-3) hours with wind from the southwest, predictions from QUIC and the regression model were negatively correlated with predictions from CALINE4, AERMOD, and R-LINE (r = −0.75 to −0.11). These inconsistent results when the wind was from the southwest may be related to air recirculation in the street canyon formed by buildings north and south of I-90.[20] QUIC predicted upward air flow on the north edge of the street canyon and downward flow on the south side, leading to upward dispersion of particles north of I-90 (SI Figure S8). Because I-90 is actually below grade but was modeled at ground level in all five models, the wind flow deviations due to street canyon effects may have been larger than the deviations predicted by QUIC and therefore the models may have overestimated PNC relative to models of a below-grade highway.

Comparison to Other Evaluations of Near-Road Models. Our main findings that the five models tested were in generally good agreement with each other, but not necessarily with measurements, are consistent with the few available near-road PNC model evaluations. Our results are in line with studies that had qualitatively good agreement with measurements for both QUIC[28] and CALINE4,[63] and had R^2 of 0.2−0.5 between hourly or subhourly near-road PNC regression models and measurements.[17,23,25] In addition, our correlations between predictions from QUIC and regression models of PNC in Somerville (R^2 = 0.82) and Chinatown (R^2 = 0.79) were similar to those reported for New York City (R^2 = 0.80).[24]

Our results are also similar to other studies comparing models of traffic emissions. In previous studies of model performance, predictions of NO_2 and tracers (i.e., sulfur hexafluoride) have been within a factor of 2 of observations with reasonable agreement among models (e.g., CALINE4, AERMOD, R-LINE, QUIC, CAR, Urban, and regression).[31−39] We found that our tested models generally underestimated PNC relative to measurements, consistent with previous studies that reported underestimation of traffic-related air pollution by dispersion models for conditions of atmospheric instability, wind direction perpendicular to the highway, or low concentrations.[11,36,37,45] However, our results were different from studies that reported overestimation of traffic-related air pollution during stable or parallel wind conditions, and when concentrations were relatively high.[11,27,36,44,45,64] Differences between our study and those reporting overestimations of concentrations could be related to model characteristics (e.g., the importance of aerosol chemistry or other primary and secondary PNC sources) and uncertainty in the emission factor inputs. In addition, although dispersion models (e.g., AERMOD, CALINE4, and R-LINE) of near-road traffic-related air pollution have generally been reported to perform better for wind speeds >1 m/s,[11,21,37,38,45] we did not observe any consistent differences between high and low wind speeds in our study, possibly because we did not model any hours with low enough wind speeds to observe a difference.

Sources of Uncertainty. The main sources of uncertainty in model predictions of near-highway PNC include factors related to model inputs (e.g., emission factors and local street traffic) and structure (e.g., treatment of plume meander and particle dynamics). The uncertainty in particle number emission factors is about a factor of 10 because limited data are available on how particle number emission rates change as a function of fleet composition, vehicle speed, traffic congestion, and meteorological conditions.[54,65] To maximize the applicability of the EF_{PN} to this study, we used temperature-adjusted EF_{PN} from a study in the Boston Central Artery Tunnel.[54] Using these emission factors, we achieved reasonably good fits to measurements in Somerville but underestimated PNC in Chinatown by about a factor of 3. These results suggest that an emission factor closer to that reported for the Williamsburg Bridge in Brooklyn, NY (5.7×10^{14} # veh^{-1} km^{-1}, ~ 2.5 times higher than our emission factors) might be more appropriate for neighborhoods like Chinatown than the tunnel-derived values.[24]

Similarly, changes in local-scale meteorology and emissions from highway and nonhighway sources could impact PNC gradients near highways.[9,66,67] The monitoring data were not adjusted using fixed sites because the models were built to reflect the traffic and meteorological conditions when the measurements for the test scenarios were performed, and subhourly measurements were not available from any fixed site. In Chinatown, emissions from local traffic, diesel trains at South Station, and airplanes at Logan Airport east of I-93 may have contributed to the differences between the PNC models and measurements. In some applications, models of PNC from highway traffic might be appropriate even if the models do not agree with measurements of total PNC. However, researchers modeling total PNC would be well served to invest in modeling all nearby sources of ultrafine particles, especially in more complex areas like Chinatown.

Different treatments of plume meander and exclusion of aerosol particle dynamics contribute to structural uncertainties

in near-road models of PNC. Treatment of plume meander is one of the major structural differences among the models considered here; AERMOD and R-LINE assume radial dispersion at low wind speeds, CALINE4 has a parameter for the standard deviation of wind direction, and QUIC does not account for deviations from the mean wind direction unless a physical obstacle is present.[21,45,49,64] However, all of the tested dispersion models had similar PNC predictions at the low (<2 m/s) wind speeds (e.g., Scenarios SV-1, CT-2 and CT-5) in which plume meander is applied in CALINE4 and R-LINE. Similarly, while particle formation and removal can be important during episodes of very high PNC and over large distances,[12,15,30] these processes are not likely to be important for the near-road environments considered in this paper. Time-scales of ~200 s for advection, ~1000 s are expected for deposition, and ~10 000 s for coagulation for near-road environments with typical PNC (10^4–10^5 particles/cm^3).[12,68] Therefore, particles are likely to be advected out of the near-road environment (0–200 m) before evolving enough to substantially change the number concentrations.

Implications. This is one of the first studies, and the most comprehensive to date, comparing multiple near-road models of PNC on an hourly time-scale. We showed that near-road air pollution models agree in some, but not all, likely meteorological and building scenarios. This result is important because exposure assessment[4,69,70] and epidemiology[5,71] of traffic-related air pollution are increasingly incorporating participant time-activity patterns. Therefore, understanding the errors in different air pollution models over short periods is valuable for predicting potential biases in exposure assessment for those models.

Based on our results, we recommend that researchers carefully consider the impacts of choice of dispersion or regression model on their near-road PNC predictions. Differences among models may be most important in areas with complex roadway geometries and wind patterns like our urban center neighborhood (i.e., Chinatown) or <50 m from the highway edge, where all five models tested under-predicted the measurements by up to a factor of 3. Overall, the most important parameters affecting the model predictions were the locations of particle sources and buildings relative to wind direction. Depending on area geometry, modelers may choose to use a hybrid approach with one model (e.g., QUIC) for wind directions where street canyon effects dominate and another model for other wind directions where the benefits of including wind flow around individual buildings might not be realized.

The models and study areas used in this investigation were selected to maximize generalizability for other near-road PNC modeling efforts. The four dispersion models we tested are freely available to the air pollution modeling community and have been used in research and regulatory applications;[21,24,28,41,44,45] the regression models are similar to those being developed and used in epidemiological studies.[13,17,29,33,36] While the Somerville study area presented some modeling challenges (i.e., I-93 was elevated, had a noise barrier, and was parallel to a state highway), it is typical of the complexity of many urban neighborhoods near highways. In contrast, Chinatown was typical of an urban core area where model performance can be degraded by complex roadway and building geometries. While we expect our methods and results to be broadly generalizable to hourly models of PNC in similar urban areas near highways in the United States, future work is needed to assess the generalizability of the model comparisons to other locations and over longer time periods.

■ ASSOCIATED CONTENT

⑤ Supporting Information

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Additional information as described in the text (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: +1 848 445-3194; fax: +1 732 445-0131; e-mail: allison.patton@alumni.tufts.edu.

ORCID Ⓘ

Allison P. Patton: 0000-0002-8334-3580

Prashant Kumar: 0000-0002-2462-4411

Notes

The authors declare no competing financial interest.

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