Factors Controlling Spatial Variations of Combustion Related Pollutants in Urban Areas

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A number of epidemiological studies have shown that exposure to elevated levels of fresh vehicular emissions causes a wide range of adverse human health effects. Fresh vehicular emissions contain a wide range of particle- and gas-phase species. Because such emissions are emitted and diluted together, their individual impacts are difficult to separate. Ultrafine particles (UFP), a focus of this work, may contribute to the degradation of health associated with exposure to elevated levels of fresh vehicular emissions. Further, because ultrafine particles are short lived, they have a low urban background and thus are also an excellent tracer for fresh vehicle emissions.

An earlier mobile measurement study by our group demonstrated a large pollutant impact zone, extending beyond 2.5 km downwind of a freeway in Santa Monica, California during the hours immediately before and after sunrise [1], in sharp contrast to daytime, when plumes dissipate within a few hundred meters. The current study explores the variability of these extended freeway plumes at several locations (and geographies) in Southern California. A mobile measurement platform (MMP) was employed to measure vehicle-related pollutant concentrations on transects running perpendicular to four freeway segments located on the coastal plain, in downtown Los Angeles near several sets of foothills, and in an inland valley during the early mornings. Two transects passed under the freeway ("overpass" freeways) and two transects passed over the freeway ("underpass" freeways). In all cases, plume lengths on the downwind sides were measured to be ~2 km or more ([2], Figure 1). Dilution rate coefficients were consistently about a factor of ten lower than commonly observed for daytime.

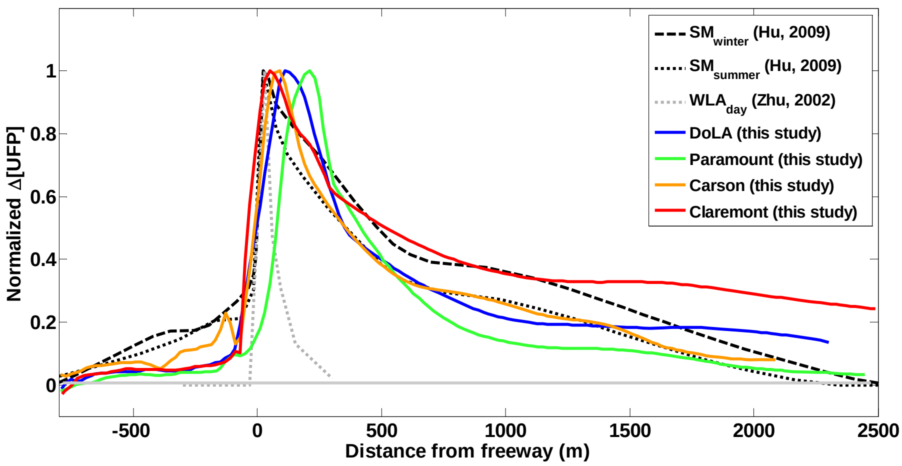

Figure 1. Variations in background-subtracted UFP concentrations with respect to distance from the freeway. Values are normalized to peak concentrations. Over-pass freeway transects: Blue, Downtown LA (DoLA) & green: Paramount; underpass freeway transects: orange: Carson & red: Claremont (this study). Black dashed and dotted lines Santa Monica (SM) [1]. Grey dotted line, daytime in West LA (WLA, Zhu et al. [3]). Horizontal line: upwind background. Positive distance indicates indicate the downwind side.

Several factors were found to control pollutant plume length and the shape of the decay curve downwind of freeways under stable conditions. In most cases, the geometry of the freeway - transect intersection (underpass or overpass) divided the dependence of decay characteristics on meteorological factors into two distinct groups (Choi et al., *in prep*.). As expected, plumes were transported further when winds were perpendicular to the freeway rather than parallel. Relationships with wind speed were complex; dispersion increased with increasing scalar windspeed, but as resultant wind both increases transport and is correlated with increased mixing, relationships are less clear. Variation in vertical stability played a minor role, given that in all cases boundary layer ranged from nearly neutral to strongly stable. Background-subtracted peak concentration (which is a function of traffic flows and temperature) was a major determinant in the plume shapes. While particle concentrations obviously do not influence mixing and dispersion, higher pollutant concentrations decay more rapidly compared to lower concentrations for a given amount of dilution with background air at fixed concentration.

Within the few hours after sunrise, the freeway plume begins to dissipate, and its extent retreats back to a few hundred meters, as has been observed in many daytime studies, although higher pollutant concentrations persist throughout the morning hours. The absolute concentrations were more than double in the winter measurements than summer, due to the interaction between the sunrise time and the timing of morning rush hour; during the summer the sun rises earlier and begins to initiate mixing before the morning rush hour has developed much [1]. The results are also provide evidence that in-use gasoline vehicle emissions of ultrafine particles in general have declined [4].

The mobile monitoring platform also was used to measure real-time air pollutant concentrations in a variety of built environments. These included the community of Boyle Heights (BH, a low income community surrounded and traversed by several freeways); Downtown Los Angeles (DTLA, adjacent to BH with taller buildings and surrounded by several freeways); and West Los Angeles (WLA, an affluent community traversed by two freeways), in summer afternoons of 2008 (all locations) and 2011 (WLA only). Significant inter-community and less significant but observable intra-community differences in traffic-related pollutant concentrations were observed both in the residential neighborhoods studied and on their arterial roadways between BH, DTLA, and WLA, particularly ultrafine particles (UFP). High emissions vehicles (HEV), defined as vehicles creating plumes with concentrations more than three standard deviations from the adjusted local baseline, were encountered during 6-13% of sampling time, during which they accounted for 17–55% of total UFP concentrations both on arterial roadways and in residential neighborhoods. If instead a single threshold value is used to define HEVs in all areas, HEV's were calculated to make larger contributions to UFP concentrations in BH than other communities by factors of 2-10 or more.

Santa Monica Airport, an airport dominated by small gasoline powered airplanes but also including ~10% small to intermediate sized jets located in WLA is a significant source for elevated UFP concentrations in nearby residential neighborhoods 80–400m downwind (see also [5]).

In the WLA area, we also showed, on a neighborhood scale, striking and immediate reductions in particulate pollution (~70% reductions in both UFP and, somewhat surprisingly, $PM_{2.5}$), corresponding to dramatic decreases in traffic densities (20 – 85%) during a major freeway closure event ("Carmageddon") compared to non-closure Saturday levels. Although pollution reduction due to decreased traffic is expected, this dramatic improvement in particulate pollution provides clear evidence air quality can be improved through strategies such as heavy-duty-diesel vehicle retrofits, earlier retirement of HEV, and transition to electric vehicles and alternative fuels, with corresponding benefits for public health.

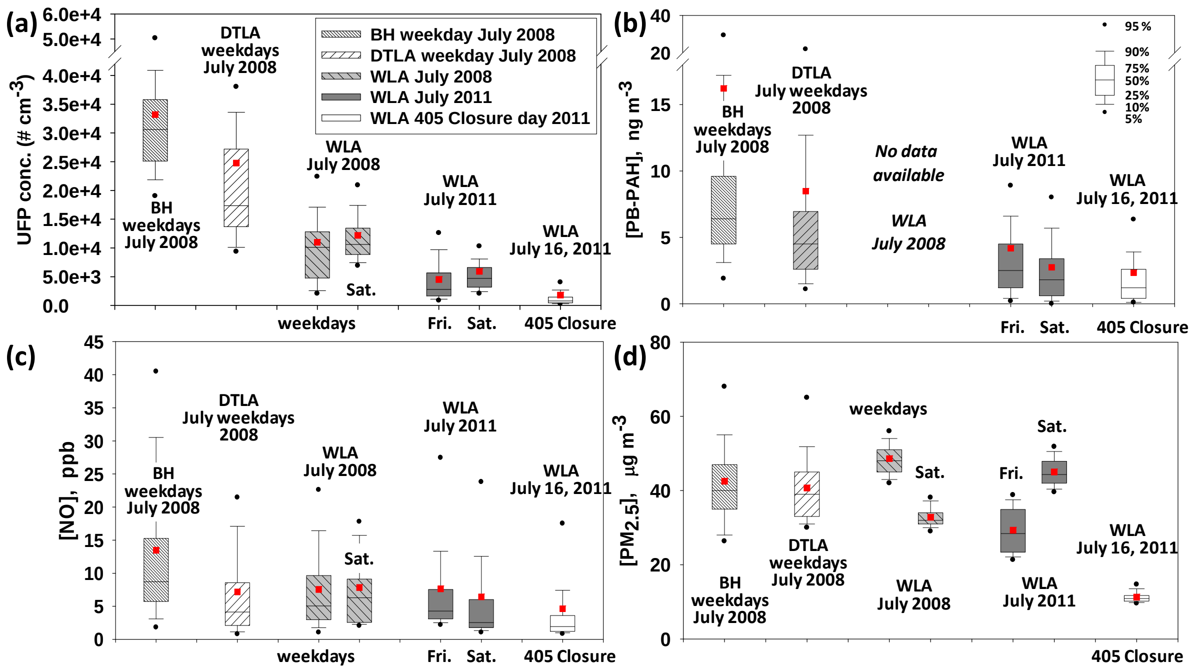

Figure 2. Box plots of pollutant concentrations measured on arterial roadways in BH in 2008 (fine slant lines in white boxes), DTLA in 2008 (coarse slant lines in white boxes), WLA in 2008 (coarse slant lines in grey boxes), WLA in 2011 (simple white boxes), and WLA adjacent to SMA in 2011 (simple dark grey boxes), and during "Carmageddon" (405 Closure): (a) UFP, (b) PB-PAH, (c) NO, and (d) $PM_{2.5}$. Red squares represent the mean values.

1. Hu, S.S., S. Fruin, K. Kozawa, S. Mara, S.E. Paulson, and A.M. Winer, *A wide area of air pollutant impact downwind of a freeway during pre-sunrise hours.* Atmospheric Environment, 2009. **43**(16): p. 2541-2549.
2. Choi, W.S., M. He, V. Barbesant, K. Kozawa, S. Mara, A.M. Winer, and S.E. Paulson, *Prevalence of wide areas of air pollutant impact downwind of freeway during pre-sunrise at several locations in Southern California.* Atmos. Environ., 2012. **62** p. 318-327.
3. Zhu, Y.F., W.C. Hinds, S. Kim, and C. Sioutas, *Concentration and size distribution of ultrafine particles near a major highway.* Journal Of The Air & Waste Management Association, 2002a. **52**(9): p. 1032-1042.
4. Choi, W.S., M. He, V. Barbesant, K. Kozawa, S. Mara, A.M. Winer, and S.E. Paulson, *Neighborhoods, roadways, and airports: Air quality benefits of emissions reductions from mobile sources.* Atmos. Environ., 2013: Accepted.
5. Hu, S., S. Fruin, K. Kozawa, S. Mara, A.M. Winer, and S.E. Paulson, *Characterization of aircraft emission impacts in a neighborhood adjacent to a general aviation airport in Southern California.* Env. Sci. Technol., 2009. **43**: p. 8039-8045.

Local Scale Spatial and Temporal Air Pollution Gradients in the Los Angeles Area

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Measurements

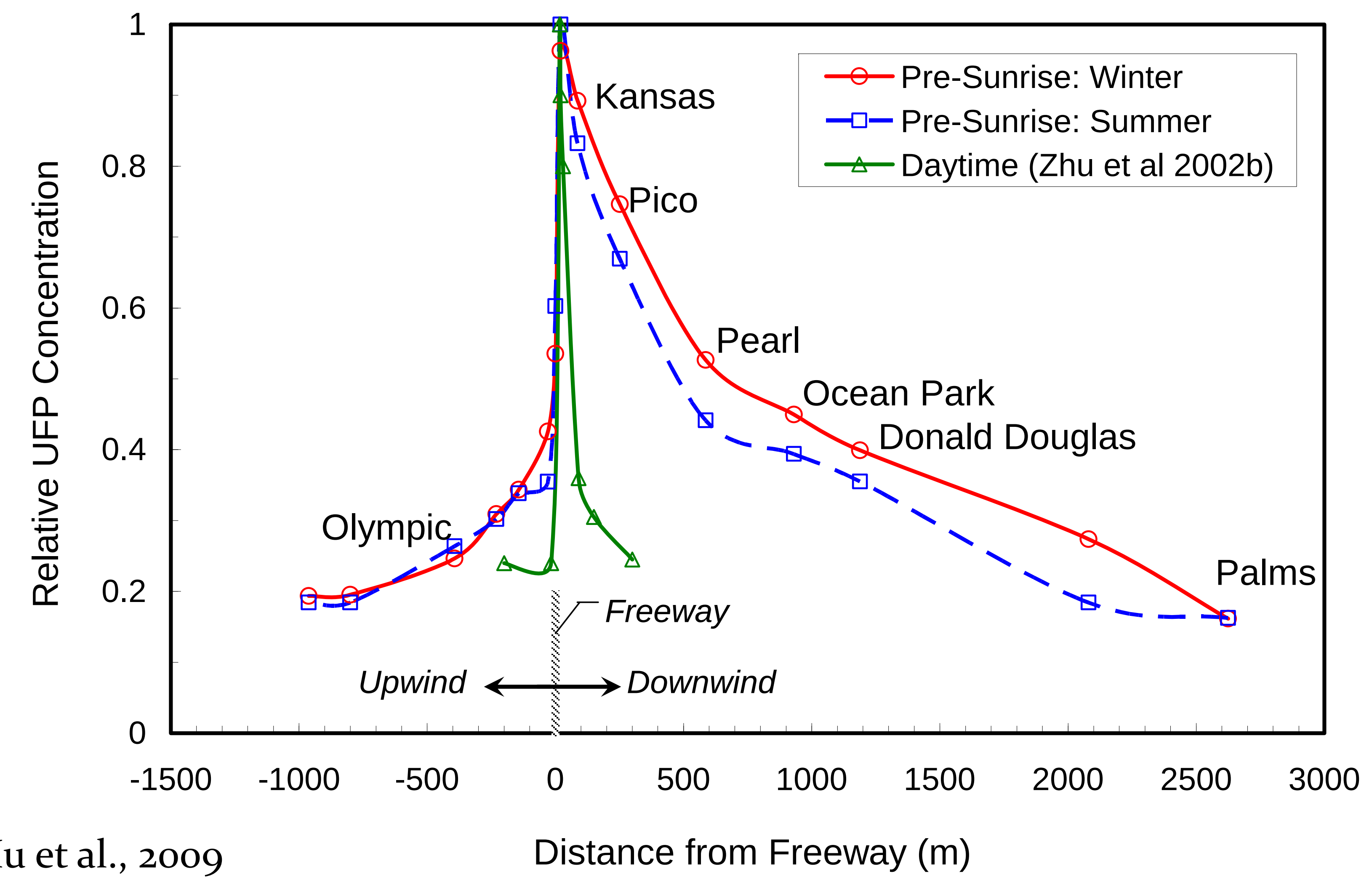
Instrument	Measurement Parameter
CPC (TSI, Model 3007)	UFP number concentration (10 nm ~ 1μm)
FMPS (TSI, Model 3091)	Particle size distribution (5.6~560 nm)
DustTrak (TSI, Model 8520)	$PM_{2.5}$ and PM_{10} mass
EcoChem PAS 2000	Particle bound PAHs
LI-COR, Model LI-820	CO_2
Teledyne API Model 300E	CO
Teledyne-API Model 200E	NO
Sonic Anemometer (Vaisala)	Temperature, Relative humidity, Wind speed/direction
Garmin GPSMAP 76CS	GPS
SmartTether™	Vertical profiles of temperature, *RH*, wind speed/direction
KciVacs video	Video record

ARB's Toyota RAV4 electric vehicle

SmartTether™

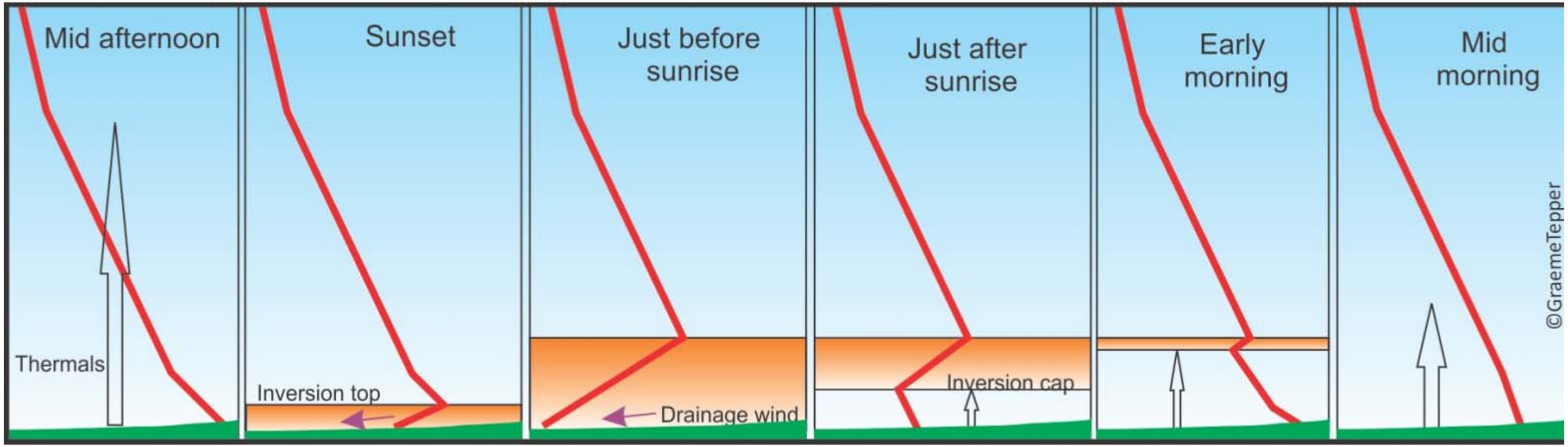
Freeway plumes in the early morning

The Freeway Imprint is Many Times Larger Before and Just After Sunrise (normalized data)



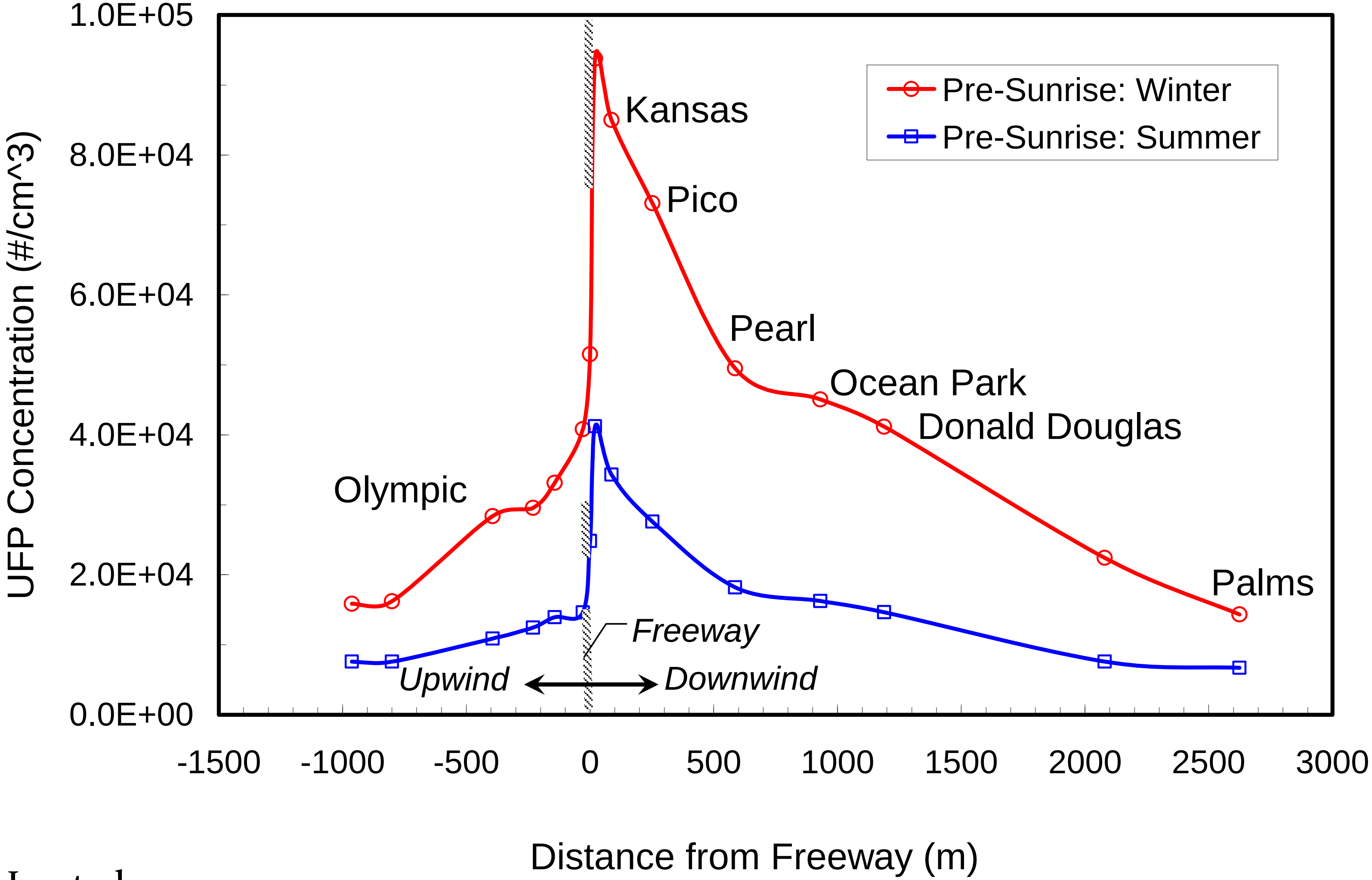
Hu et al., 2009

The Atmosphere Strongly Traps Pollution Near the Surface in the Early Morning



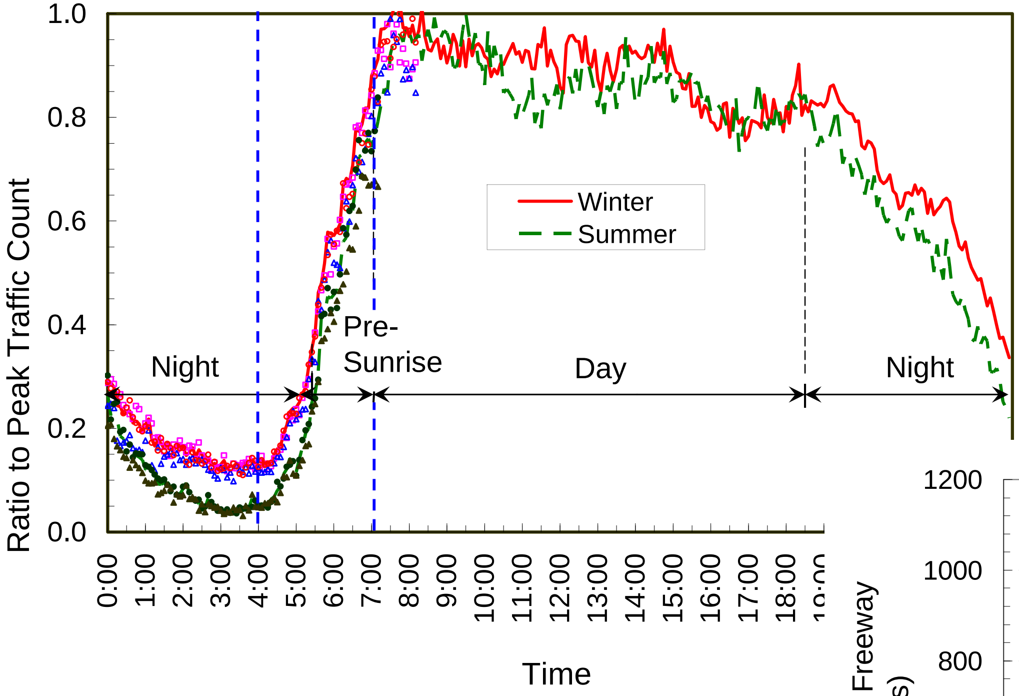
Red line indicates temperature profile

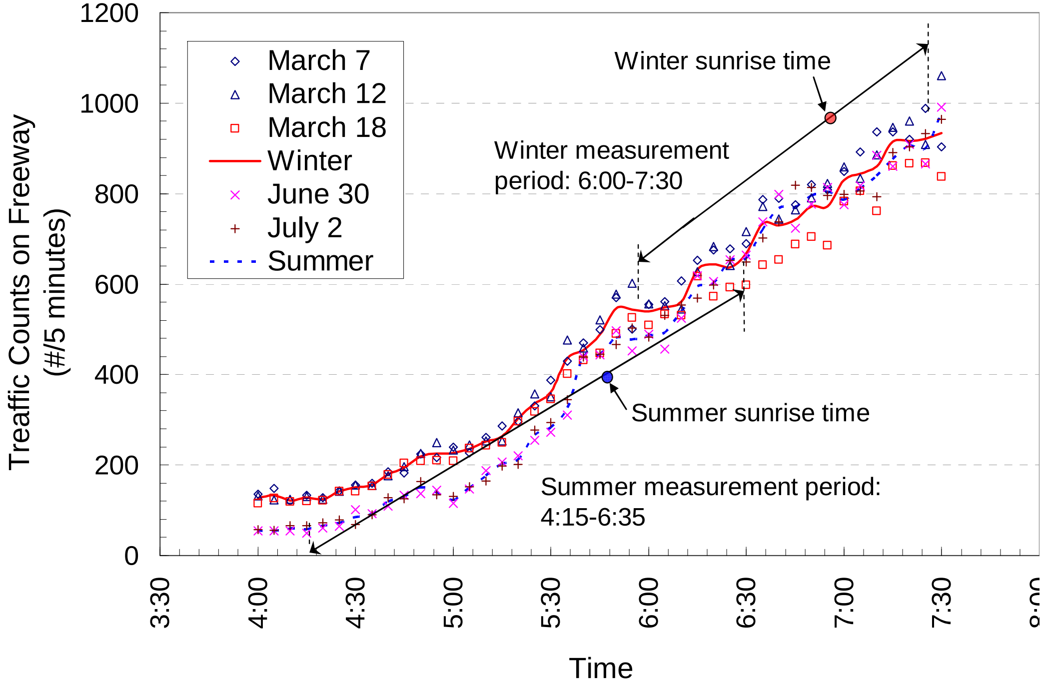
Santa Monica: Summer is Cleaner; why?



Hu et al., 2009

Traffic Counts Increase Rapidly in the Early AM

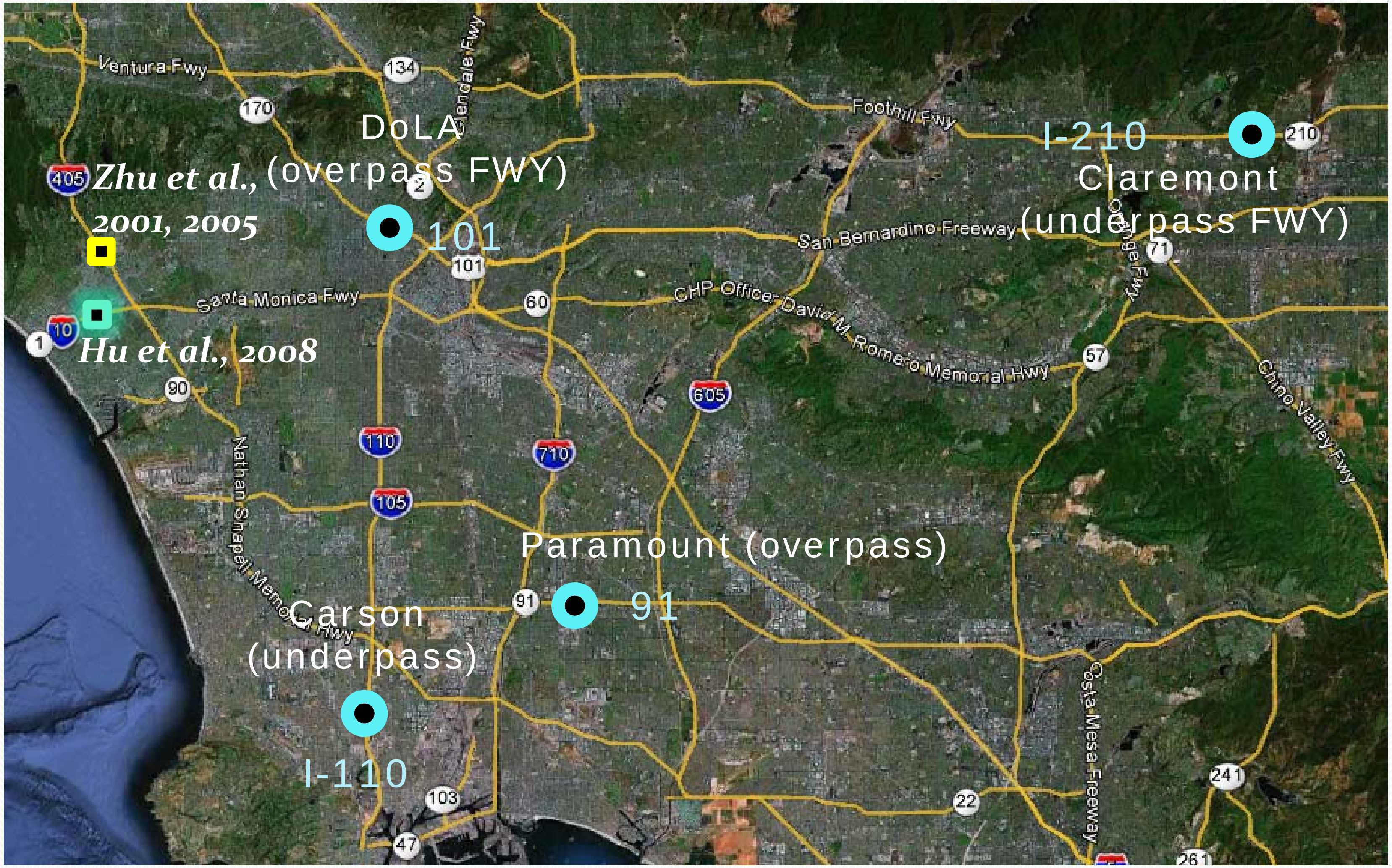




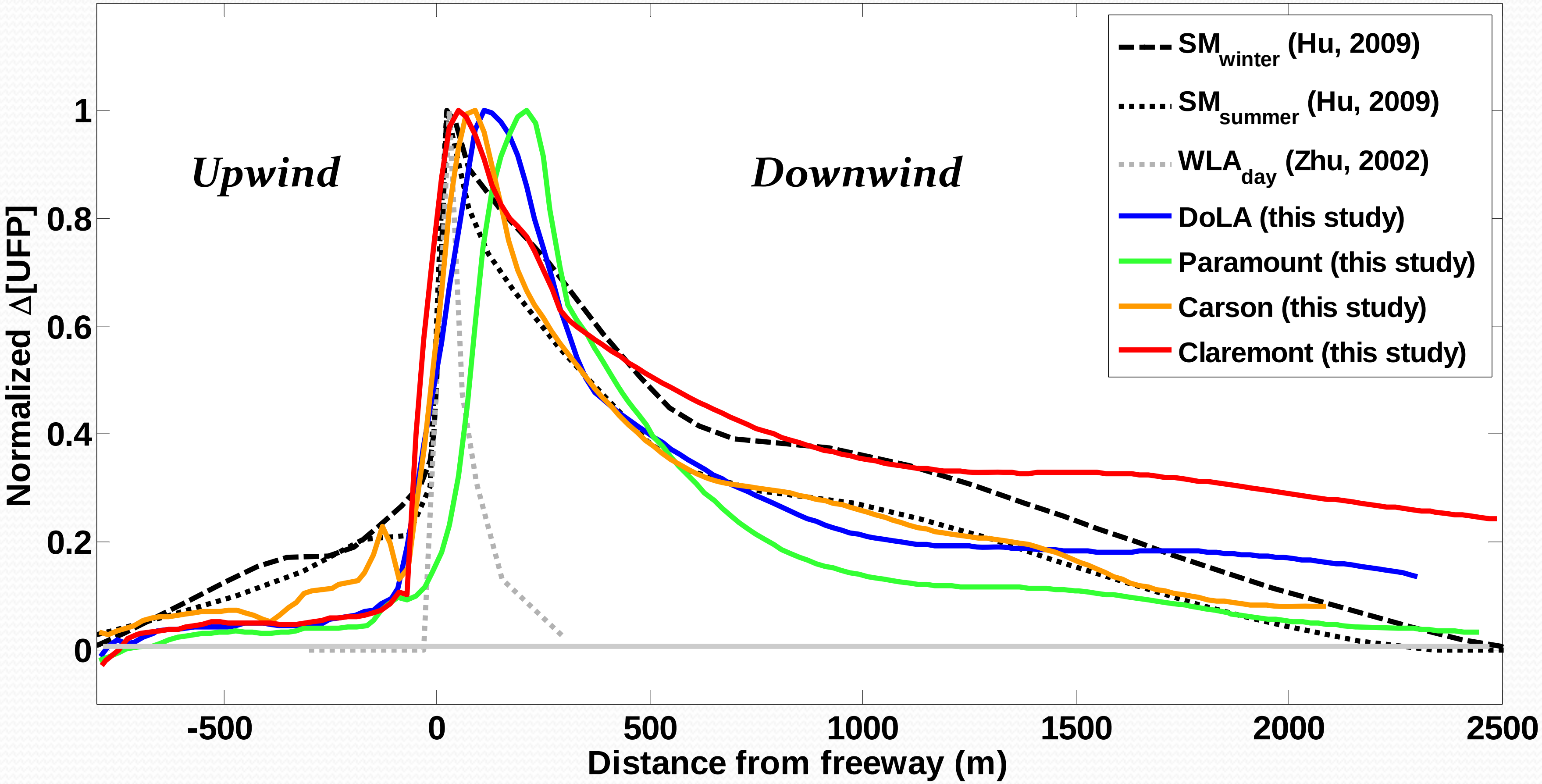
Summer is cleaner because there is less traffic during the pre-sunrise period

Hu et al., 2009

Sampling Area and Transects



Wide Impact Area Downwind of Freeways

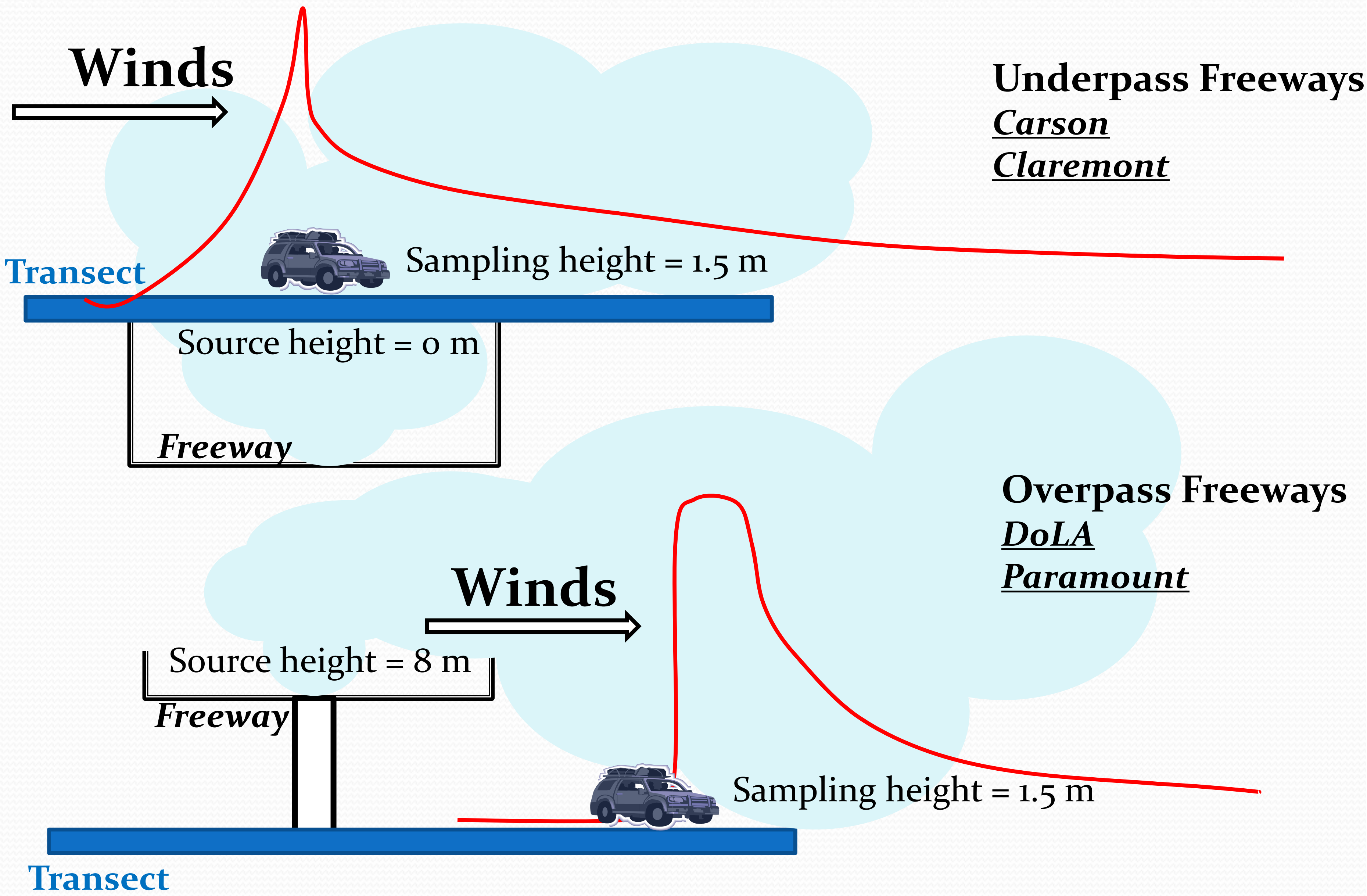


$$\Delta[UFP] = [UFP] - [UFP]_{bkgnd}$$

$$\text{Normalized } \Delta[UFP(x)] = \frac{\Delta[UFP(x)]}{\Delta[UFP]_{peak}}$$

[Choi et al., Atmos. Environ., 62, 318-327, 2012]

Freeway-Transect Geometry



Fits Model to Observed Profiles to Extract Emission Factor and Dispersion Coefficients

[Choi et al., submitted]

Gaussian Plume Dispersion model

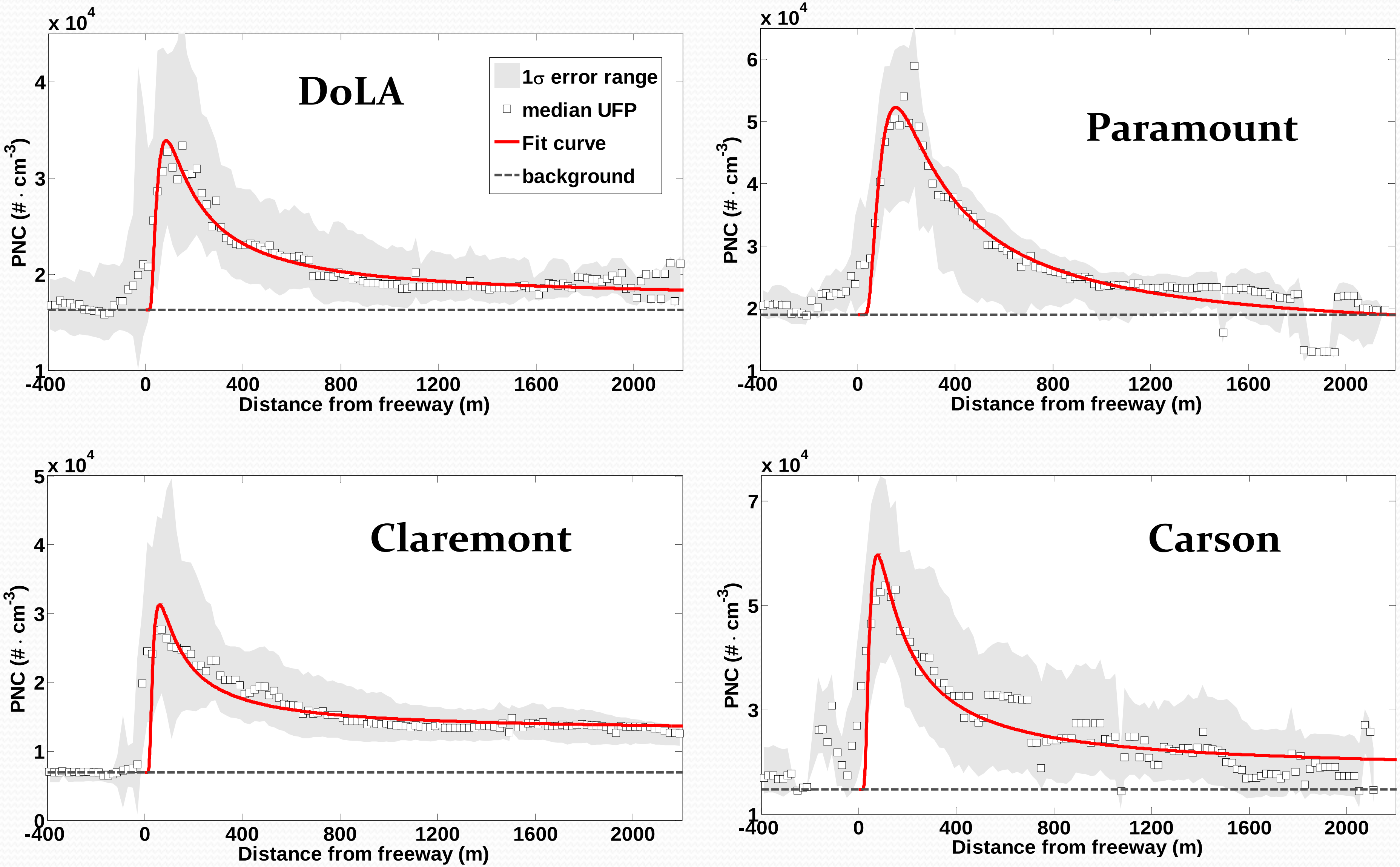
Q_c = *Emission rate corrected with wind speeds*
H = *Source height*
1.5m = *Measurement height*
σ_z = *Dispersion parameter*
x = *Horizontal distance from the source*

$$C(x,1.5m) = \frac{Q_c}{\sigma_z}\left[\exp\left(-\frac{(1.5m+H)^2}{2\sigma_z^2}\right)+\exp\left(-\frac{(1.5m-H)^2}{2\sigma_z^2}\right)\right]$$

References	Equation form	Land use	Stability Class	Dispersion coefficients
Briggs (1973)	$\sigma_z = \frac{\alpha \cdot x}{(1+\beta \cdot x)}$ (*distance*: x)	Rural	E[a] (slightly stable)	α = 0.03 β = 0.3×10^{-3}
			F[a] (moderately stable)	α = 0.016 β = 0.3×10^{-3}
		Urban	E – F[a] (stable)	α = 0.08 β = 1.5×10^{-3}

Dispersion Parameter

The Model Fits the Observations Well *($R^2 > 0.9$)*



Estimating the Particle Number Emission Factor

$$Q_c = \frac{q_{veh} \times (\text{Traffic flow})}{2\sqrt{2\pi}U_e}$$

Q_c = *Wind speed-corrected Emission rate* $(\# \cdot m \cdot cm^{-3})$

q_{veh} = *Particle number emission factor (PNEF)* $(\# \cdot mile^{-1} \cdot vehicle^{-1})$

Traffic flow = $vehicles \cdot s^{-1}$

U_e = *Effective wind speeds* *[Chock, AE, 1978]*
(wind speed + speed correction factor due to traffic wake)

$$q_{veh} = \frac{\sqrt{2\pi}Q_c \cdot U_e}{(\text{traffic flow})}$$

with the mean values obtained from observations

$$= \frac{\sqrt{2\pi} \times (8.12 \times 10^4) \times (0.64\,m/s + 0.2\,m/s) \times 10^6\, cm^3/m^3 \times 300\, s/5\min}{(680.2\, vehicles/5\min)}$$

= 7×10^{13} particles·mi^{-1}·vehicle^{-1}

This is 15% of the Particle Emission Factor measured in West LA in 2001

4.9×10^{14} particles·mi^{-1}·vehicle^{-1} in 2001 *[Zhu and Hinds, AE, 2005]*

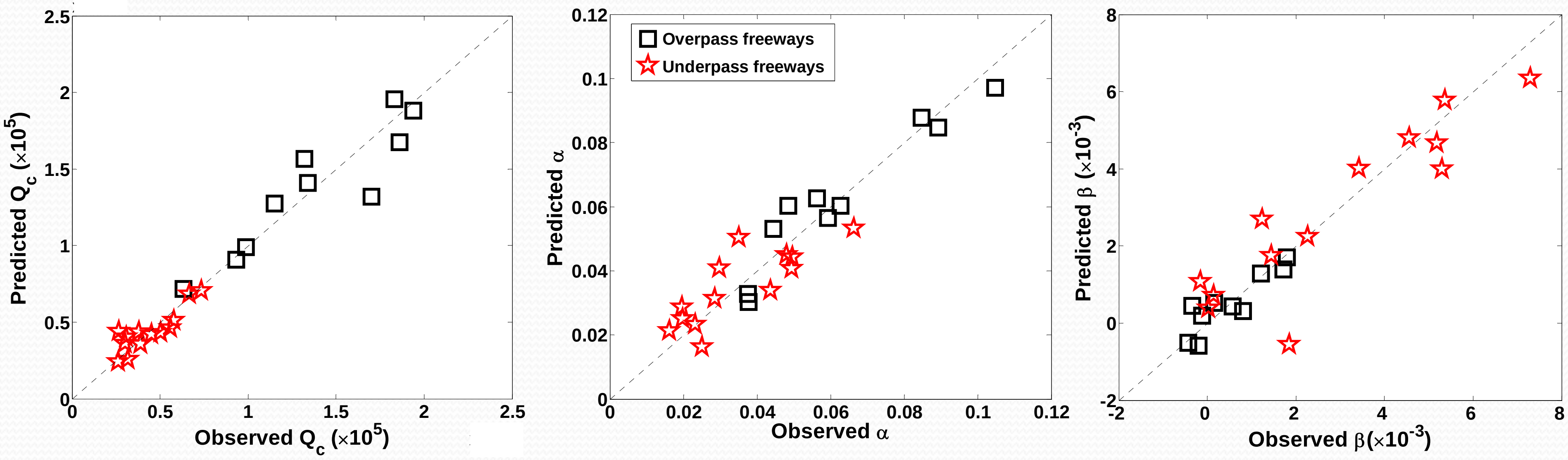
Prediction of Dispersion Coefficients

Multivariate Regression Model

Q_c : emission rate factor
WD_{rel}: relative wind direction to freeway
T : temperature
WSR : vector mean resultant wind speed
RH : relative humidity
C : correction factor

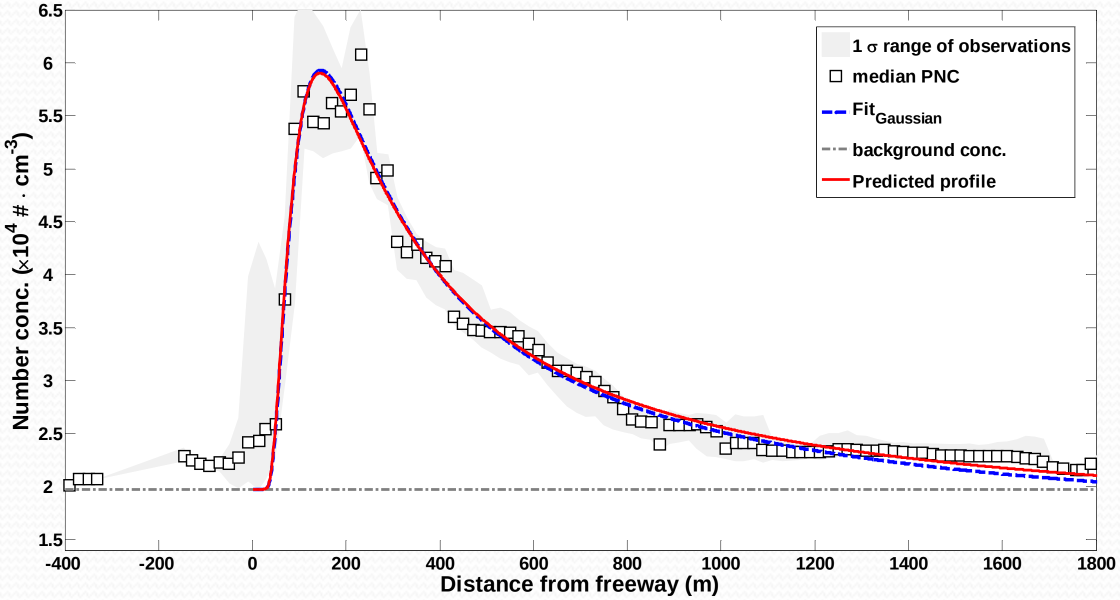
$$Q_{c,j} = c_1 \cdot Traffic_j + c_2 \cdot \left|WD_{rel,j}\right| + c_3 \cdot T_j + c_4 \cdot WSR_j + c_5 \cdot RH_j + C$$

$$\alpha_j \text{ or } \beta_j = c_1 \cdot Q_{c,j} + c_2 \cdot \left|WD_{rel,j}\right| + c_3 \cdot T_j + c_4 \cdot WSR_j + c_5 \cdot RH_j + C \qquad (j = 1,2,3,\ldots,k)$$

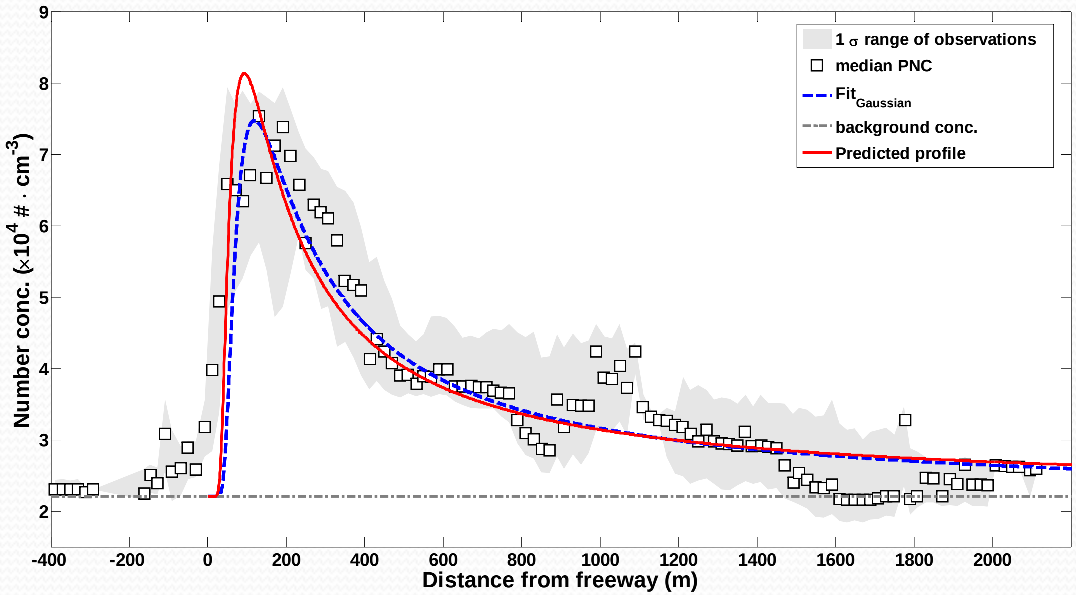


$R^2 = 0.95$ ***$R^2 = 0.88$*** ***$R^2 = 0.86$***

Predicted Profiles Match the Data Well

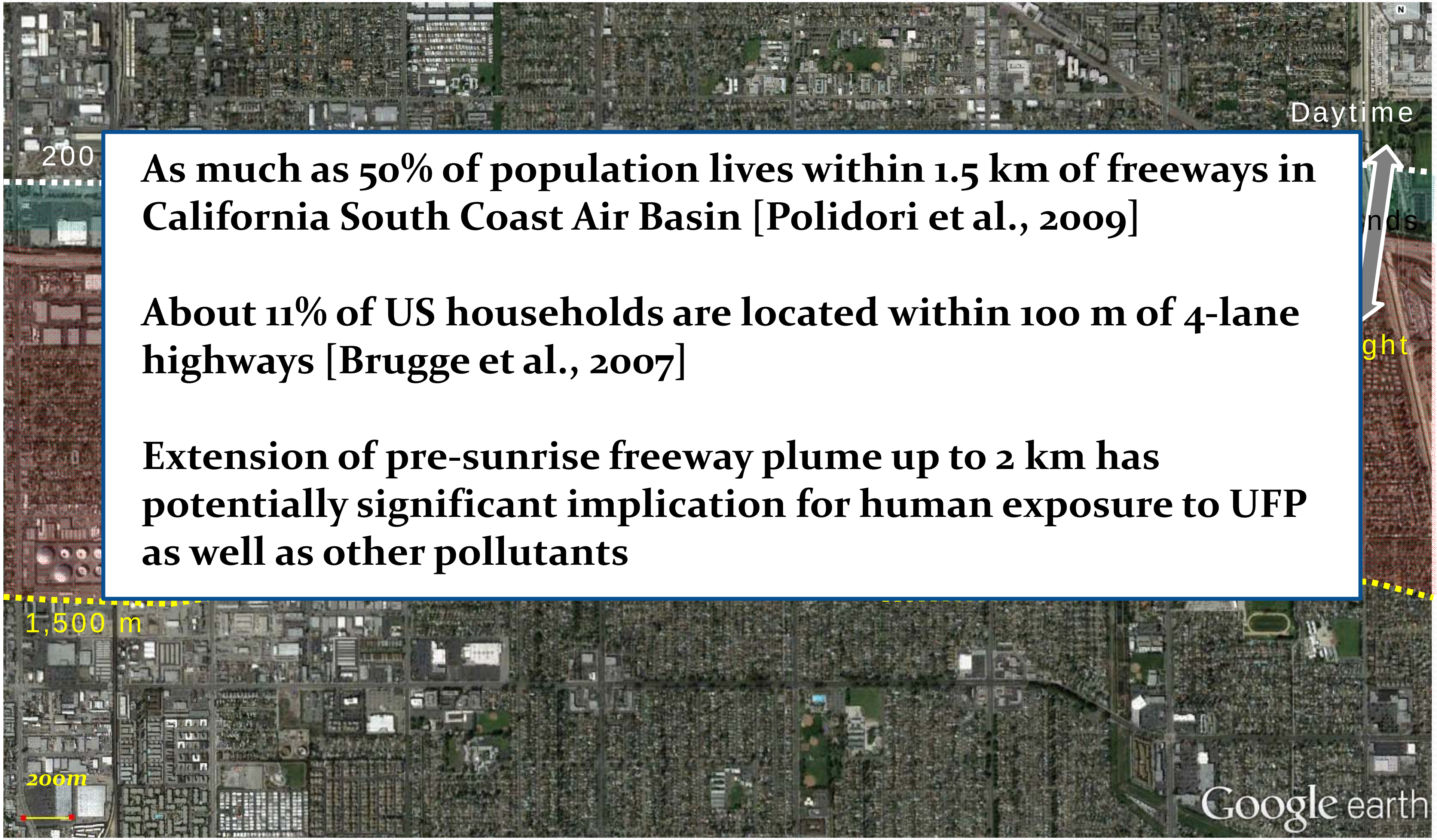


Paramount, March 18th, 2011



Carson, February 2nd, 2011

Night and Day



As much as 50% of population lives within 1.5 km of freeways in California South Coast Air Basin [Polidori et al., 2009]

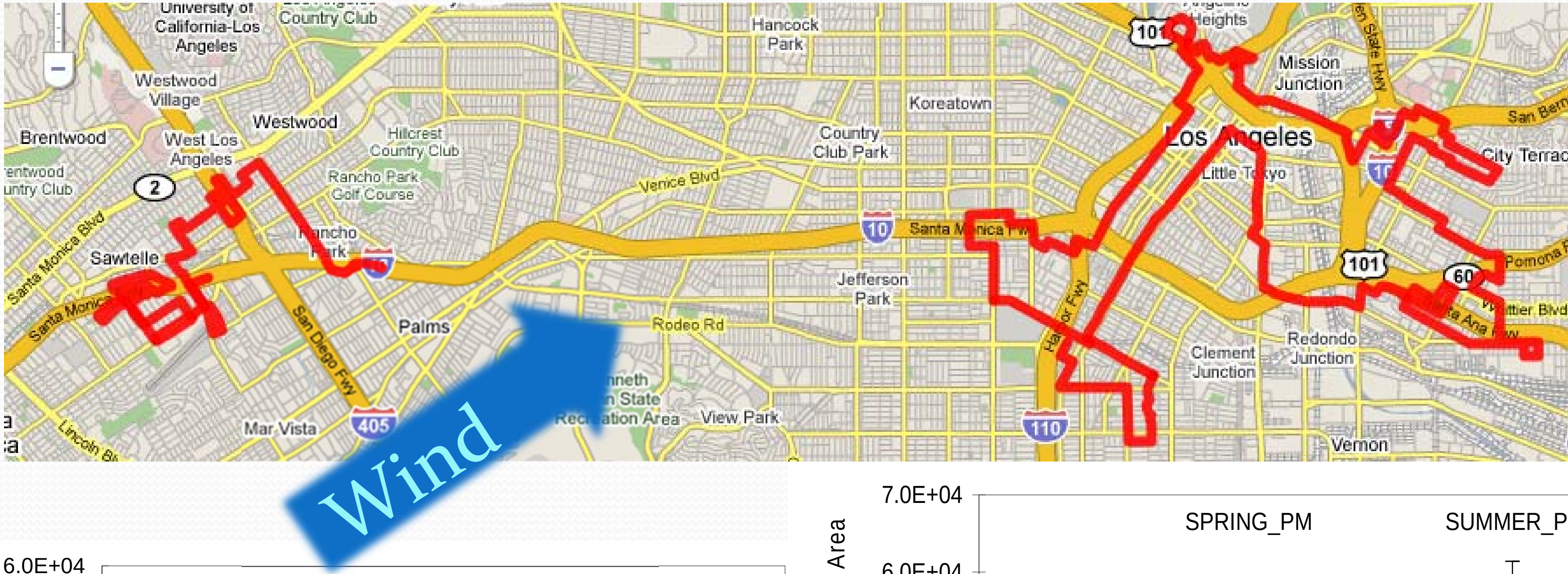
About 11% of US households are located within 100 m of 4-lane highways [Brugge et al., 2007]

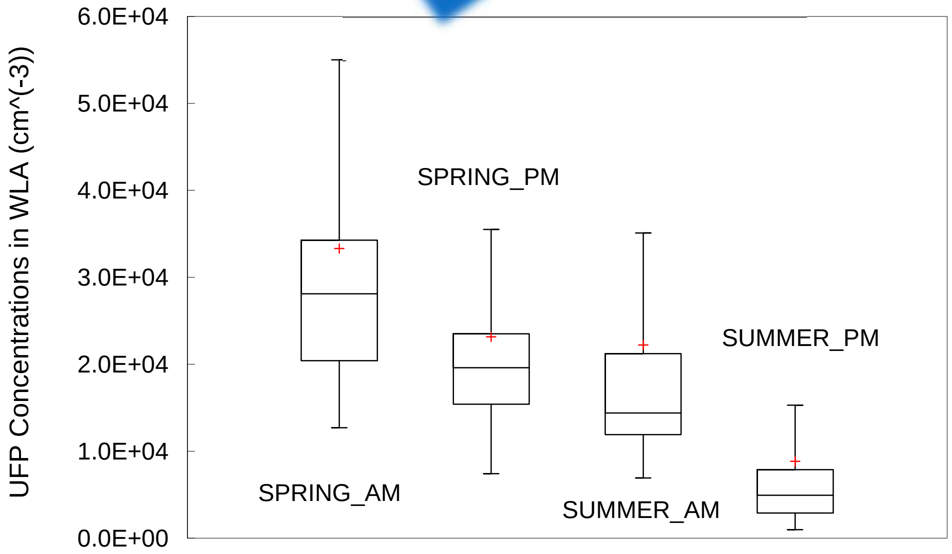
Extension of pre-sunrise freeway plume up to 2 km has potentially significant implication for human exposure to UFP as well as other pollutants

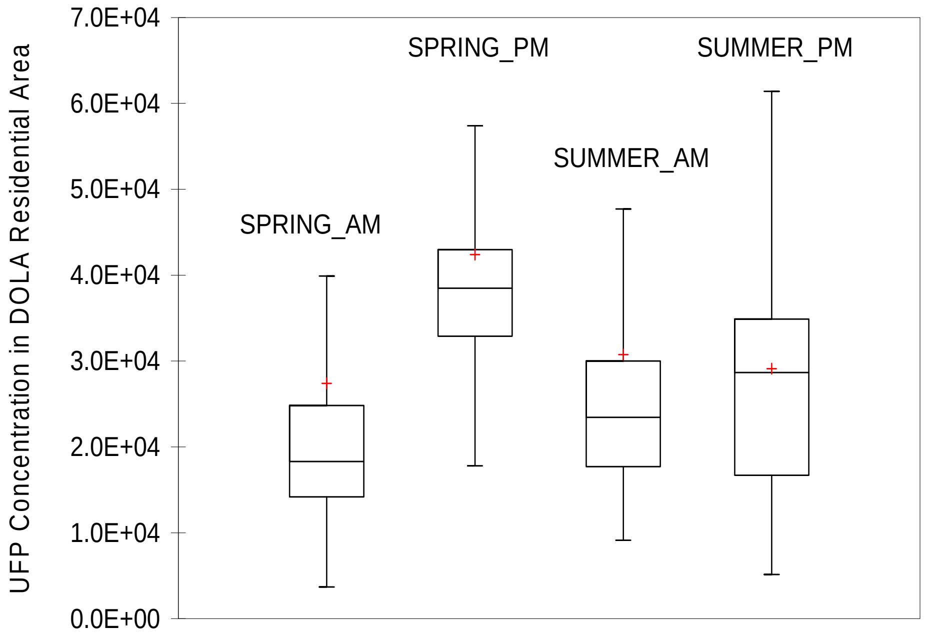
Paramount

Air Quality in Several Los Angeles Neighborhoods

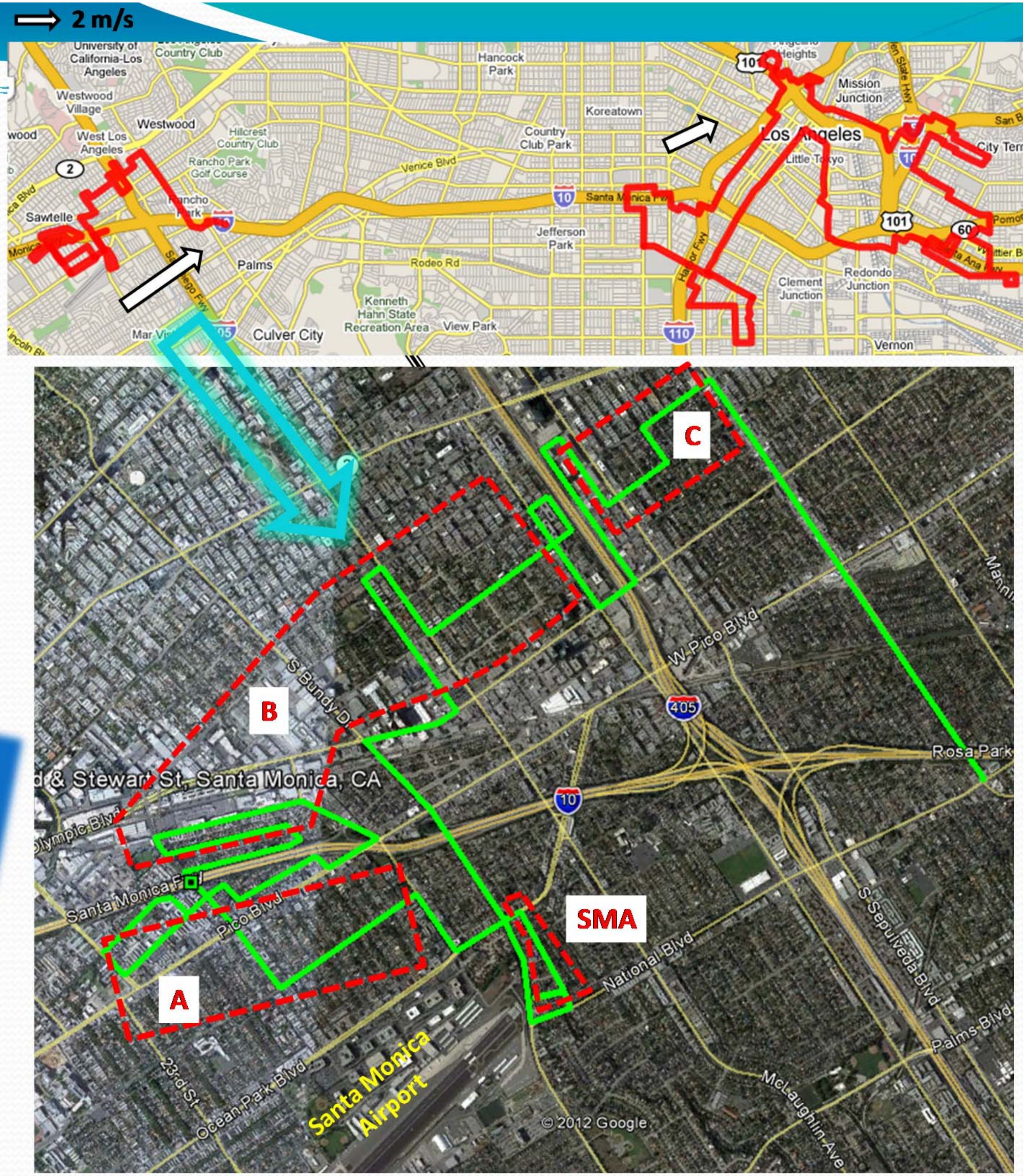
Temporal trends are quite different in different areas. Data are for residential areas only.



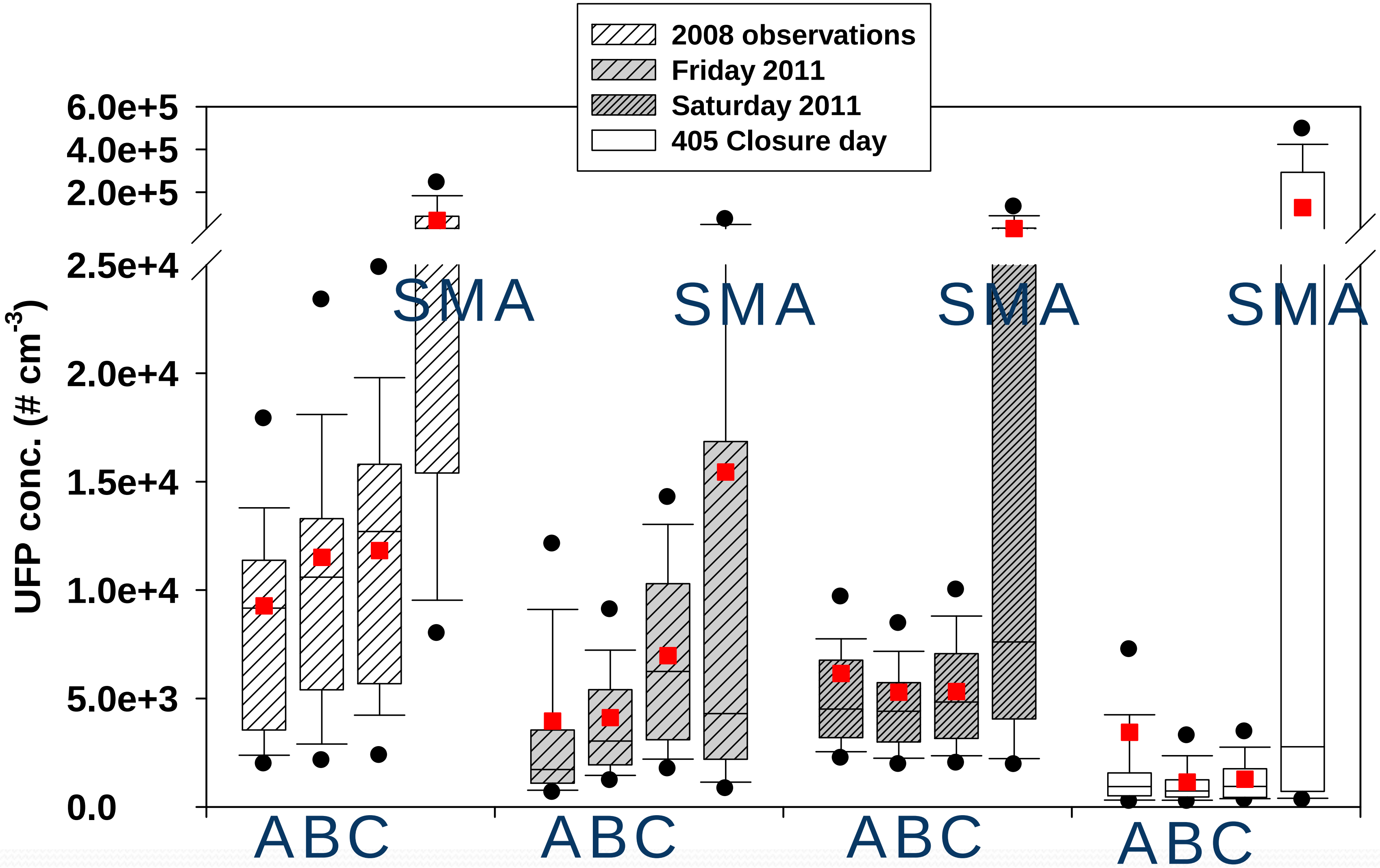




West Los Angeles measurement areas in 2008 and 2011



Ultrafine Particle Concentrations Vary Substantially between Neighborhoods

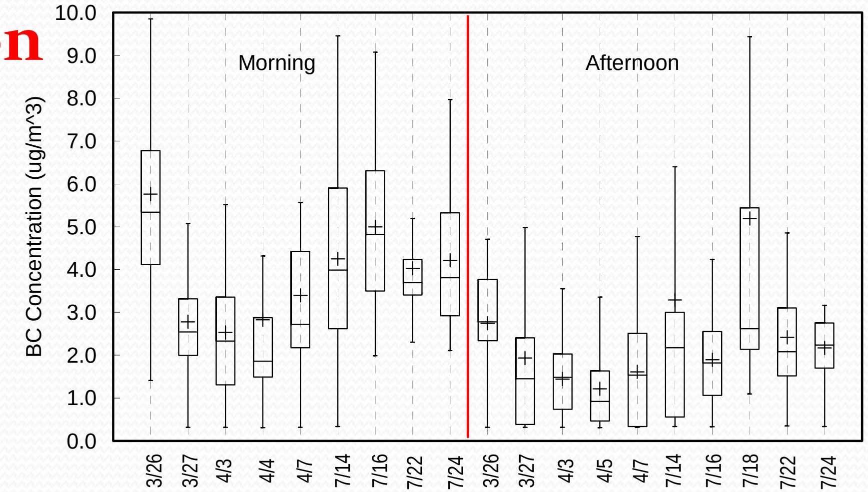


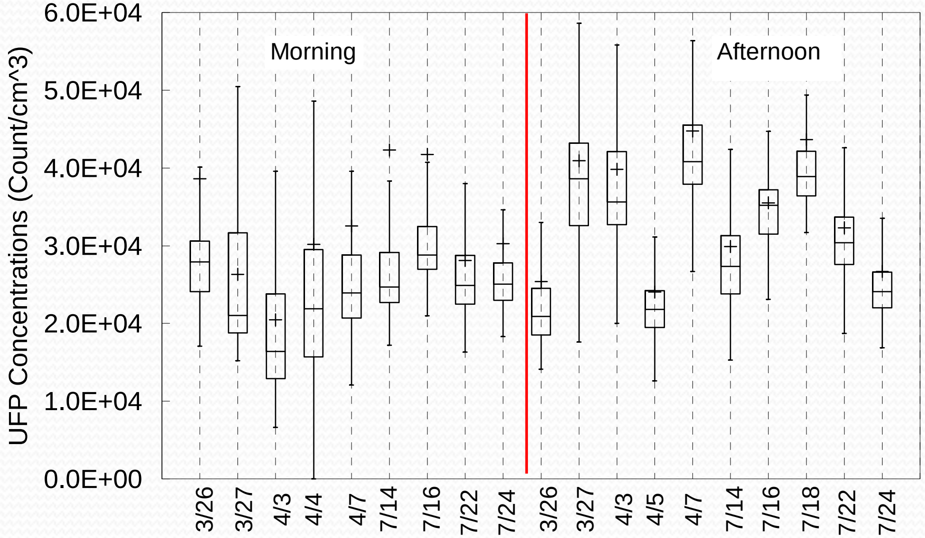
Summary

1. **Early morning extension of freeway plumes far downwind (> 2 km) is a general phenomenon in Southern California, and presumably most locations around the globe.**
2. Data indicate a strong drop in emissions of ultrafine particles over the past decade.
3. **Plume intensity as well as met. Parameters control pollutant plume lengths downwind of freeways.**
4. Plume shapes and areal impact can be predicted from routinely measurable parameters.
5. **Behavior of UFP concentrations in neighborhoods is sufficiently complex as to be easy to explain but somewhat difficult to predict.**

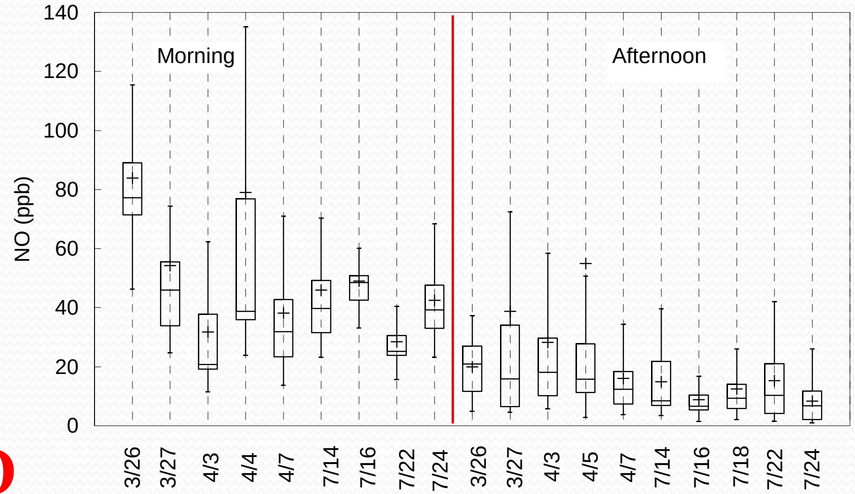
15 miles inland: UFP are higher in the afternoon while other 1° pollutants are lower

Black Carbon



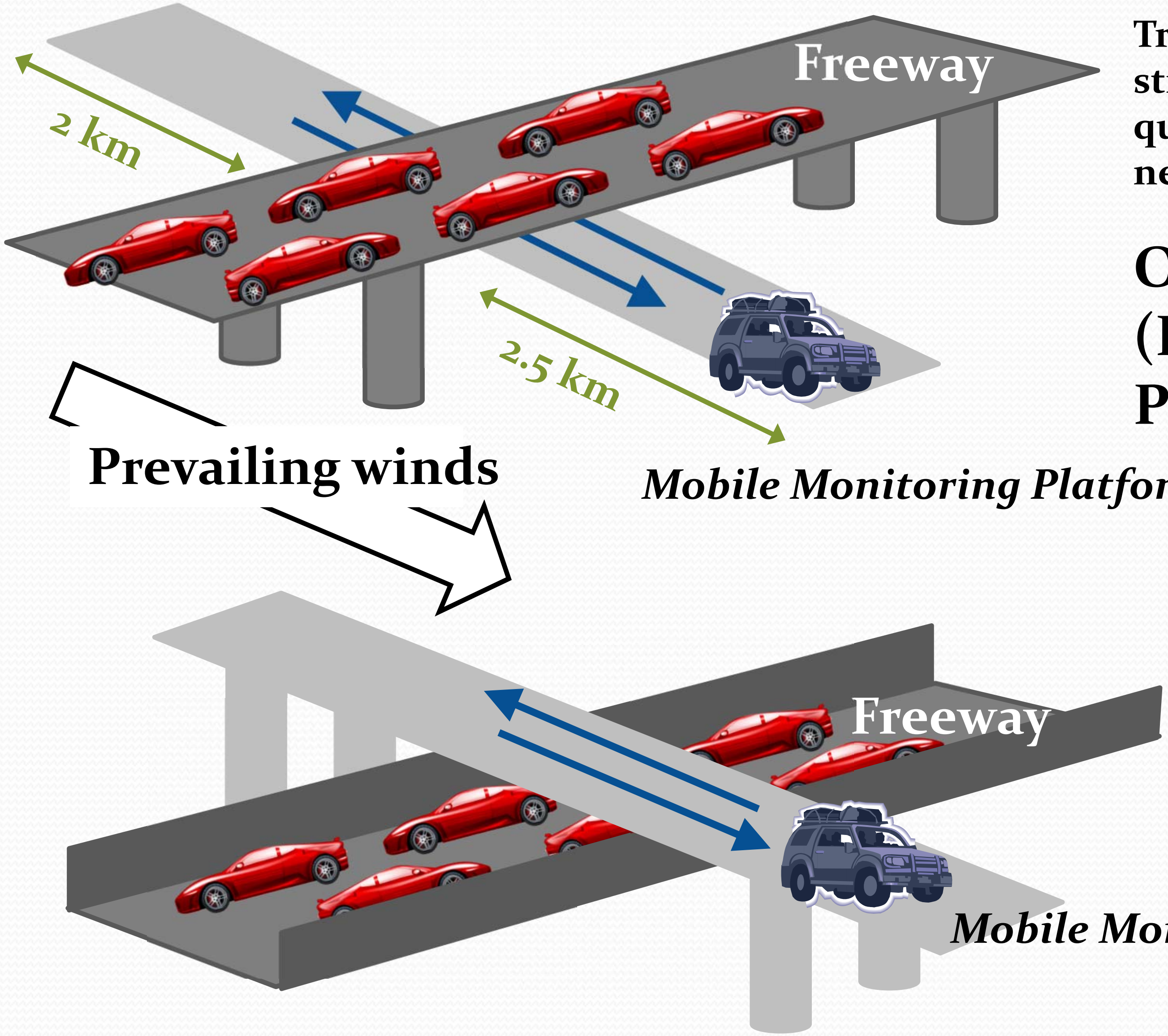


Ultra Fine Particles



NO

Transect Geometry Influences Plume Shape

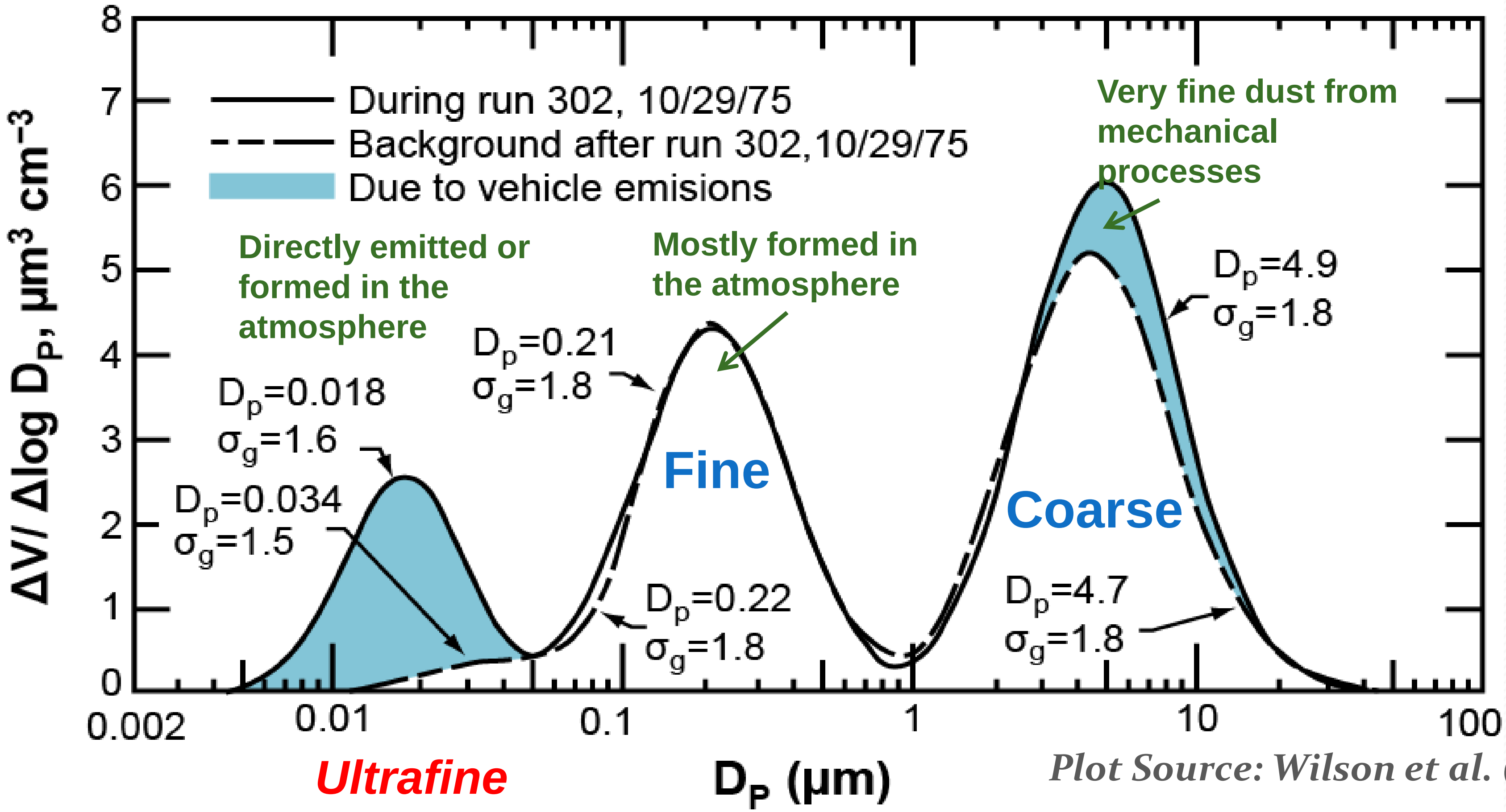


Transects are small 2-lane streets running through quiet residential neighborhoods

Overpass Freeway (DoLA and Paramount)

Underpass Freeway (Carson & Claremont)

Size Distribution of Atmospheric Particles



Plot Source: Wilson et al. (1977)

- **Mostly from vehicular emissions highly concentrated on UFP region: ~80% of the total number conc. but negligible in mass conc.** *[Kumar et al., 2010]*
- **Formed generally by condensation in the diluting exhaust plume (semi-volatile hydrocarbons and hydrated sulfuric acid)** *[Shi et al., 2000]*