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**Statistical Methods for Time Series of Ultrafine
Particle Distributions**

A dissertation submitted in partial satisfaction
of the requirements for the degree
Doctor of Philosophy in Biostatistics

by

Heidi Jean Fischer

2015

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ABSTRACT OF THE DISSERTATION

Statistical Methods for Time Series of Ultrafine Particle Distributions

by

Heidi Jean Fischer

Doctor of Philosophy in Biostatistics

University of California, Los Angeles, 2015

Professor Robert E. Weiss, Chair

We develop statistical methods to model ultrafine particle (UFP) counts as a function of size and time using Bayesian functional time-series methods. Because of their small diameter and relatively large surface area, UFPs can penetrate the lung, enter the circulatory system, and deposit in the brain. Health effects of UFPs may be strongly linked to particle size as this characteristic determines which region in the lung the particles deposit. UFP counts can increase by orders of magnitude above natural levels in urban environments from anthropogenic processes such as the operation of combustion engines. Understanding how UFP counts of various sizes change over time near combustion engines and high traffic roadways is crucial in setting idling, vehicle reduction, and vehicle emissions policy.

We first propose Bayesian longitudinal functional time series models to understand the impact of engine idling on UFP counts inside school buses. Our approach is a varying coefficient model and we model UFP counts over size at a given time with a cubic B-spline basis. UFP counts across a range of sizes were collected over time during multiple experiments while the bus engines were off and then turned on, making the data multivariate longitudinal. Steady meteorological and background traffic conditions during individual experiments imply that

UFP counts over size do not vary greatly with time before the engine is on, but variation in engine-off UFP counts is observed between experiments. We model this using a spline random effect model across all UFP sizes. We allow counts to increase over time at a size dependent rate once the engine is turned on. We explore alternate models for the engine-on increase: a possible jump in counts at engine-on followed by either a quadratic or bent line time trend. A random jump by experiment is also explored. Residuals are modeled with an autoregressive model over time to accommodate correlation over time. To account for larger count variance for smaller particles relative to larger particles, the log residual variance over size bin is modeled using a quadratic B-spline basis.

We next propose distinct Bayesian functional time series models to model the how UFP counts change over time near the I-405 roadway. As with the bus experiment, we model UFP counts over size at a given time with a cubic B-spline basis. Our methods allow UFP counts to then change over time at a size bin dependent rate by letting B-spline coefficients evolve over time following a random walk model. This model smooths UFP counts both over size and over time. To account for larger count variance for smaller particles relative to larger particle size bins, the log residual variance over size bin is again modeled using a quadratic B-spline basis. We also explore modeling residuals with an autoregressive model to accommodate correlation over time.

In both studies, we provide statistical summaries of how particle counts evolve over time as a function of size bin to estimate the contribution of emissions to UFP counts. We utilize plots to aid in the interpretation of model inferences and make model output interpretable to non-statisticians. Graphs also aid in diagnosis of lack of fit and can help suggest model improvements. Taken jointly, our methods can be used to understand the behavior of UFP counts over size and over time near combustion engines and high traffic roadways.

The dissertation of Heidi Jean Fischer is approved.

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Robert E. Weiss, Committee Chair

University of California, Los Angeles

2015

I would like to dedicate this thesis to Stefan, my mother, my father, and Emily
who have supported me and believed in me all these years.

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CHAPTER 1

Introduction

Aerosol particles are airborne solid or liquid particles. Found everywhere in the earth's atmosphere, the number and size distribution of aerosol particles in a given environment is governed by dynamic processes. Existing aerosol particles combine to grow in diameter and separate to shrink in diameter. Particles can also desorb to return to a gaseous state, form from a gaseous state, or deposit on solid surfaces. The rates of these dynamic processes change with temperature, wind speed, and other background conditions (Whitby et al., 1991).

Ultrafine particles (UFPs) are a subset of aerosol particles which have diameters less than 100 nm. Researchers have suggested that UFPs are more toxic to laboratory animals and humans than larger aerosol particles per unit mass (Alessandrini et al., 2006; Delfino et al., 2005; Ferin et al., 1990; Frampton et al., 2006). Because of their small particle size and relatively large surface area, UFPs can penetrate the lung, enter the circulatory system, and deposit in the brain (Oberdorster et al., 2004; Samet et al., 2009). Health effects of UFPs may be strongly linked to particle size as this characteristic determines which region in the lung the particles deposit (Morawska et al., 2008).

UFPs are always present in the air due to natural sources and processes. UFP counts can increase by orders of magnitude above natural levels in urban environments from anthropogenic processes such as the operation of combustion engines (Whitby et al., 1991; Morawska et al., 2008), particularly for the smallest size bins. Given the negative health effects of the UFP particles produced by

automobiles, buses and trucks, understanding how UFP counts of various sizes change over time near combustion engines and high traffic roadways is crucial.

Researchers use Scanning Mobility Particle Sizers (SMPS) to count the number of aerosol particles per cubic centimeter. The SMPS counts particles in 102 *size bins* grouped by diameter ranging from the first bin containing particles of the smallest diameters, 7.37–7.64 nm, to the last bin containing sizes of 269.0–278.8 nm. The ordered collection of counts in these 102 size bins at a single point in time is called a *UFP size distribution*, even though (i) technically particles with diameters greater than 100 nm are too big to be UFPs and (ii) the counts are not a distribution in the statistical sense. Size bin widths are approximately equally spaced on a log scale, so UFP size distributions have more bins for the smaller UFP particles of interest. The SMPS takes a full UFP size distribution measurement every two minutes.

Numerous mathematical representations have been used to describe size distributions over size bin and time, typically via *modal methods* (Whitby, 1978; Whitby et al., 1991). Modal methods model the particle size distribution as a mixture of densities (Hussein et al., 2005; Whitby et al., 1991), ignoring the total number of particles. More recently, Wraith et al. (2009, 2014) model time series of UFP size distributions using time-varying non-parameteric Bayesian mixture models.

Modal methods standardize particle counts: only information about the relative composition of particle size bins is retained. In contrast, in modeling vehicle emissions and in setting vehicle emissions policy, understanding actual particle counts is crucial; thus modal methods are insufficient for this application. Modal methods have also not accounted for sampling variances in observed UFP counts. UFP counts in smaller size bins have larger variances than those in larger size bins, as they tend to be more unstable than larger particles, following dynamic processes which have them desorb, deposit, or combine to form larger particles at

fairly rapid rates (Kulmala et al., 2004; Kittelson et al., 2006).

In urban environments UFP counts vary for certain particle size bins more than others because they are created by anthropogenic processes which fluctuate over time. Additionally the SMPS machine has higher measurement error for particles in these smaller size bins. Statistical methods which model mean UFP counts by particle size bin and over time must account for increased residual variance for smaller particle size bins.

In this thesis, we discuss statistical methods which model the behavior of UFP size distributions using functional time-series methods, which allow for inference on particle counts while accounting for differences in residual variance across particle size bin.

In Chapter 2 we propose Bayesian longitudinal functional time series models to understand the impact of engine idling on UFP counts inside school buses. In the U.S., roughly 25 million children ride school buses daily. About 90 percent of buses are diesel powered, emitting particles primarily in the UFP size range (EPA1, 2002; EPA2, 2014). As school buses idle at bus stops, UFPs from diesel emissions penetrate into cabins through cracks, doors, and windows. This so-called self-pollution increases the exposure to UFPs of children on board (Zhang et al., 2012). How UFP counts vary by particle size as school buses idle over time and under different idling conditions is not yet well understood.

UFP distributions were collected over time during multiple experiments, or *runs*, making the data multivariate longitudinal. Our approach is a varying coefficient model as in Hastie and Tibshirani (1993) or Lang and Brezger (2004). We model UFP size distributions at a given time with a cubic B-spline basis (de Boor, 1978) and allow counts to increase over time at a size bin dependent rate once the engine is turned on. We explore alternate models for the engine-on increase: a possible jump in counts at engine-on followed by either a quadratic or bent line time trend. For models which include an engine-on jump, we also consider a

random jump.

Steady meteorological and background traffic conditions during individual runs implies that UFP size distributions before the engine is on do not vary greatly with time, but variation in *baseline* UFP size distributions is observed between runs. Spline random effect models have been used numerous times in the literature to describe non-parametric processes with longitudinal study designs (e.g. Ruppert et al., 2003; Shi et al., 1996), however our model is different in that B-splines model UFP size distributions, not time trends. We use an autoregressive model for residuals to accommodate correlation over time. To account for larger count variance for smaller particles relative to larger particle size bins, the log residual variance over size bin is modeled using a quadratic B-spline basis.

In Chapter 3, we propose Bayesian functional time series models to assess the how UFP counts change over time near the I-405 roadway. Elevated concentrations of UFPs are commonly found near roadways and exposure to traffic-related air pollutants has been linked to adverse health effects by epidemiological cohort studies (Beelen et al., 2007; Laden et al., 2006; Zhu et al., 2002). Due to the significant contribution of air pollutants from traffic emissions, several studies have examined how traffic calming policies such as traffic restriction could be a rational strategy to mitigate urban air pollutants (Friedman et al., 2001; Levy et al., 2006; Wang et al., 2009; Whitlow et al., 2011), however how UFP counts vary by particle size near major roadways over time under different traffic conditions is not yet well understood.

In July 2011, a section of one of the busiest freeways in United States, the I-405, was scheduled for a two-day closure as part of a freeway improvement project. The closure event, dubbed *Carmageddon*, provided a valuable opportunity to conduct a natural air quality experiment by allowing comparison of UFP counts under high and low traffic conditions and to see how these evolve over time. Researchers collected particle counts near the I-405 during Saturdays pre-

ceeding, during, and after the 2011 scheduled closure event, as well as on Fridays and Sundays preceding and during the closure event.

As with the bus experiment, we model UFP size distributions at a given time with a cubic B-spline basis (de Boor, 1978). UFP counts then change over time at a size bin dependent rate by letting B-spline coefficients evolve over time following a random walk model (Prado and West, 2010; West and Harrison, 1997). This model smooths UFP counts both over size bin and over time, allowing for inference on count trends over time across various size bins. To account for larger count variance for smaller particles relative to larger particle size bins, the log residual variance over size bin is again modeled using a quadratic B-spline basis. We also explore modeling residuals with an autoregressive model to accommodate correlation over time.

In Chapter 2, interest centers on how mean particle counts change after the engine turns on as a function of size bin, while for Chapter 3, the focus is on how particle counts evolve over time near the I-405 as a function of size bin. In both studies, researchers are interested in number of local modes of particle size bin counts, local mode heights, and how modes evolve over time. Because combustion engines emit particles of extremely small diameter and these particles are also found to be the most toxic, researchers are interested in which conditions produce local modes in the smallest particle sizes and when these reach their maximum heights. We utilize plots to aid in the interpretation of model inferences and make model output interpretable to non-statisticians. Graphs also aid in diagnosis of lack of fit and can help suggest model improvements. Chapters 2 and 3 are intended to be stand alone so there are introductions for each with some overlap in definitions and concepts. Notation is defined separately for each chapter.

Finally, we provide remarks and a discussion of extensions to the discussed methods in Chapter 4.

CHAPTER 2

Ultrafine Particle Distributions Inside School Buses

2.1 Introduction

Ultrafine particles (UFPs) are types of particulate matter with diameters less than 100 nm. UFPs' small size and large surface area allow them to penetrate the lung, enter the circulatory system, and deposit in the brain (Oberdorster et al., 2004; Samet et al., 2009) and it has been suggested that they are more toxic to humans than larger particles (Alessandrini et al., 2006; Delfino et al., 2005; Ferin et al., 1990; Frampton et al., 2006). The health effects of UFPs are linked to particle size which determines the region in the lung the particles deposit (Morawska et al., 2008). Children are more sensitive than adults to UFPs because their physiological and immunological systems are still developing (Bennett and Zeman, 1998).

UFPs are always present in the air due to natural sources and processes. UFP counts can increase by orders of magnitude above natural levels in urban environments from anthropogenic processes such as the operation of combustion engines (Whitby et al., 1991; Morawska et al., 2008), particularly for the smallest size bins. Diesel engines in particular emit particles primarily in the UFP size range (EPA1, 2002; EPA2, 2014).

In the U.S., roughly 25 million children ride school buses daily. About 90 percent of buses are diesel powered. As school buses idle at bus stops, UFPs from diesel emissions penetrate into cabins through cracks, doors, and windows. This

so-called self-pollution increases the exposure to UFPs of children on board (Zhang et al., 2012). How UFP counts vary by particle size as school buses idle over time and under different idling conditions is not yet well understood. Given the negative health effects of the UFP particles, particularly to children, understanding how UFP counts of various sizes change over time in idling school buses is crucial to establishing good practice idling practices in the future.

Researchers collected particle counts inside buses first with the engine-off and then with the engine turned on and idling. A Scanning Mobility Particle Sizer (SMPS) counted particles per cubic centimeter in 102 *size bins* that group particles by diameter ranging from the first size bin containing particles of the smallest diameters, 7.37–7.64 nm, to the last size bin containing sizes of 269.0–278.8 nm. The ordered collection of counts in these 102 size bins at a single point in time is called a *UFP size distribution*, even though (i) technically particles with diameters greater than 100 nm are too big to be UFPs and (ii) the counts are not a distribution in the statistical sense. Size bin widths are approximately equally spaced on a log scale, so UFP size distributions have more bins for the smaller UFP particles of interest. UFP distributions were collected over time during multiple experiments, or *runs*, making the data multivariate longitudinal.

Numerous mathematical representations have been used to describe size distributions over size bin and time, typically via *modal methods* (Whitby, 1978; Whitby et al., 1991). Modal methods model the particle size distribution as a mixture of densities (Hussein et al., 2005; Whitby et al., 1991), ignoring the total number of particles. More recently, Wraith et al. (2009, 2014) have modeled time series of UFP size distributions using time-varying non-parameteric Bayesian mixture models. Modal methods standardize particle counts: only information about the relative composition of particle size bins is retained. In contrast, in modeling vehicle emissions and in setting vehicle emissions policy, understanding actual particle counts is crucial; thus modal methods are insufficient for this application.

Modal methods have also not accounted for sampling variances in observed UFP counts. UFP counts in smaller size bins have larger variances than those in larger size bins, as they tend to be more unstable than larger particles, following dynamic processes that have them desorb, deposit, or combine to form larger particles at fairly rapid rates (Kulmala et al., 2004; Kittelson et al., 2006). In urban environments UFP counts vary for certain particle size bins more than others because they are created by anthropogenic processes that fluctuate over time. Statistical methods that model mean UFP counts by particle size bin and over time must account for increased residual variance for smaller particle size bins.

Modeling UFP size distributions using functional time-series methods allows for inference on particle counts while accounting for differences in residual variance across particle size bin. It is also an improvement on the methods in Zhang et al. (2012), who modeled UFP counts inside idling school buses over time using separate univariate longitudinal models for each particle size bin. As we expect neighboring bins to have similar counts, the univariate longitudinal approach does not fully utilize the information in the data.

We propose Bayesian longitudinal functional time series models to assess the impact of engine idling on UFP counts inside school buses. Our approach is a varying coefficient model as in Hastie and Tibshirani (1993) or Lang and Brezger (2004). We model UFP size distributions at a given time with a cubic B-spline basis (de Boor, 1978) and allow counts to increase over time at a size bin dependent rate once the engine is turned on. We explore alternate models for the engine-on increase: a possible jump in counts at engine-on followed by either a quadratic or bent line time trend.

Steady meteorological and background traffic conditions during individual runs implies that UFP size distributions before the engine is on do not vary greatly with time, but variation in *baseline* UFP size distributions is observed between runs. Spline random effect models have been used numerous times in the literature to

describe non-parametric processes with longitudinal study designs (e.g. Ruppert et al., 2003; Shi et al., 1996); however, our model is unusual in that B-splines model UFP size distributions, not time trends. We use an autoregressive model to accommodate correlation over time. To account for larger count variance for smaller particles relative to larger particle size bins, the log residual variance over size bin is modeled using a quadratic B-spline basis.

Interest centers on how mean particle counts change after the engine turns on as a function of size bin. Researchers are also interested in the mode of particle size counts, the mode height, and how both evolve after the engine turns on. Because combustion engines emit particles of extremely small diameter and these particles are also found to be the most toxic, researchers are interested in which conditions produce local modes in the smallest particle sizes, how high these local modes are, and when these reach their maximum heights. We provide summaries of how the mode and mode height evolve as the engine idles. Plots are presented to aid in the interpretation of model inferences and make model output interpretable to non-statisticians. Graphs also aid in diagnosis of lack of fit and can help suggest model improvements.

In Section 2.2 we describe the dataset, Section 2.3 presents our model and Section 2.4 gives results. Finally, Section 2.5 is discussion.

2.2 Data Description

UFP size distribution measurements were collected inside the bus every 2 minutes and the current analysis considers measurements taken during the time period between 15 minutes before the engine was turned on and 20 minutes afterwards. A set of UFP size distributions collected over this time period defines one run, though a few runs are shorter than the defined time. For certain runs, measurements occurred at odd numbered minutes while for other runs measurements occurred at even numbers. Runs took place under one of two window positions: (1) all windows-closed, although some windows could not be closed tightly; and (2) eight rear windows, four on each side, open 20 cm. There are 21 runs in this dataset: 12 for windows-open and 9 for windows-closed. The study was conducted in an open space under stable meteorological conditions without nearby UFP emission sources in Los Angeles, CA (Zhang et al., 2012).

Figure 2.1 shows examples of engine-off UFP size distributions from different runs showing variation in particle counts by run. Particle counts are shown as a bar chart with labels below the x-axis indicating size bin number and labels just above the x-axis indicating particle diameter (nm).

Figure 2.2 shows a plot of size distributions for a single run. Time is measured in minutes from when the engine is turned on and ranges from approximately -15 to 20 minutes. The 7 UFP size distributions collected before the engine is turned on are plotted as darker curves and the 9 UFP distributions collected after the engine is turned on are plotted as progressively lighter curves. UFP size distributions become increasingly more peaked the longer the engine runs. The rate of increase in particle counts after engine-on varies greatly by size bin, with little to no increase seen above bin 70, and much larger increases in the 10-60 size bin range. This particular set of UFP size distributions has only one mode, and that mode occurs at smaller size bins as the engine runs while the height nearly

triples in magnitude.

Figure 2.3(a) and Figure 2.3(b) plot particle counts and log particle counts over time for size bin 30 (20.9-21.7 nm) for all runs by window position. Each line is a separate run. Counts for size bin 30 generally increase sharply when the engine first turns on then continue to increase at slower rates thereafter, though in some cases increases are not seen, particularly for the window closed position. A log transformation allows for easier temporal modeling of counts (Whitby et al., 1991; Wraith et al., 2009, 2014).

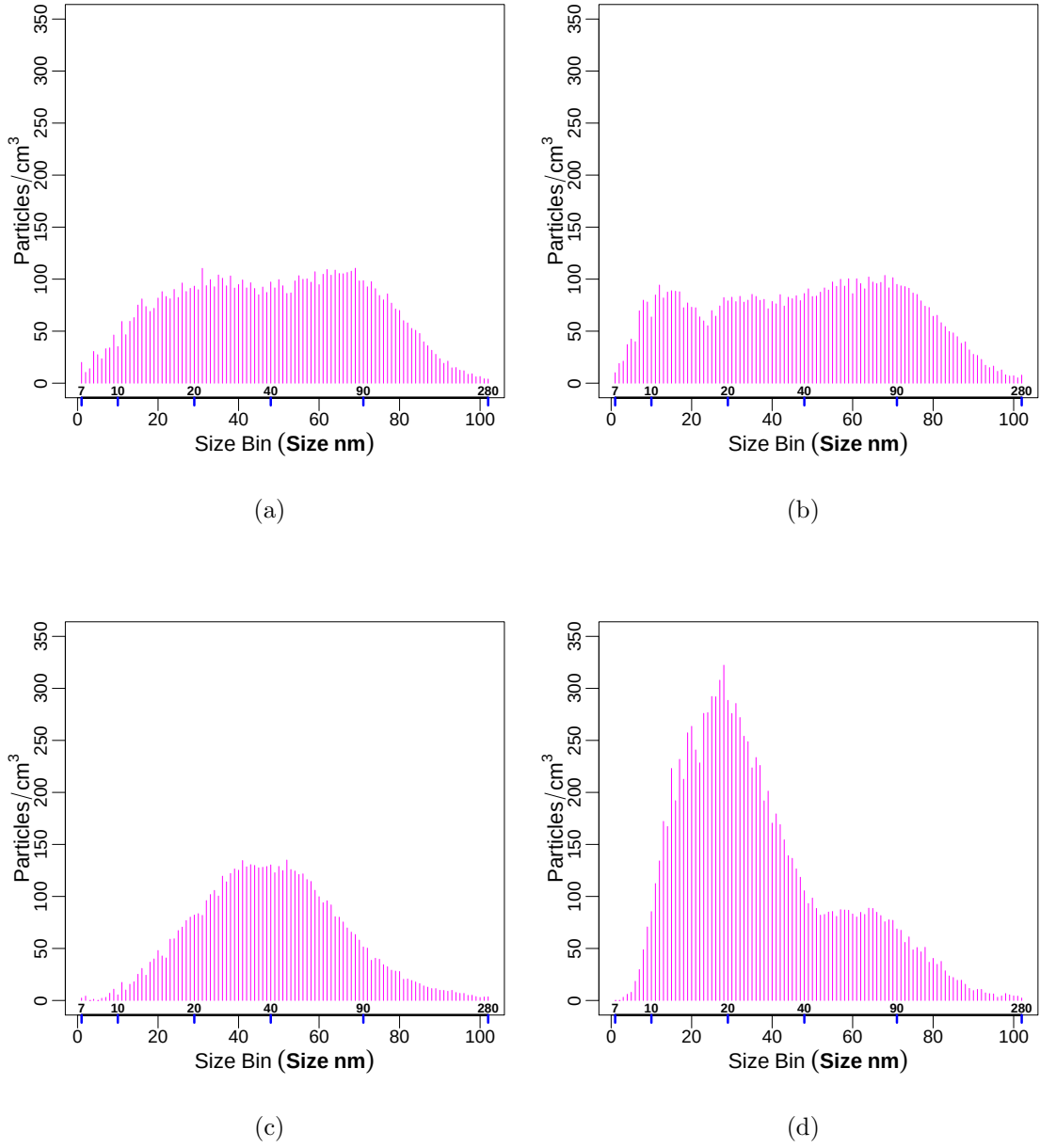


Figure 2.1: Figures a) through d) show examples of engine-off UFP size distributions from different runs showing variation in particle counts by run. Particle counts are shown as a bar chart with labels below the x-axis indicating size bin number and above the x-axis indicating approximate particle diameter (nm).

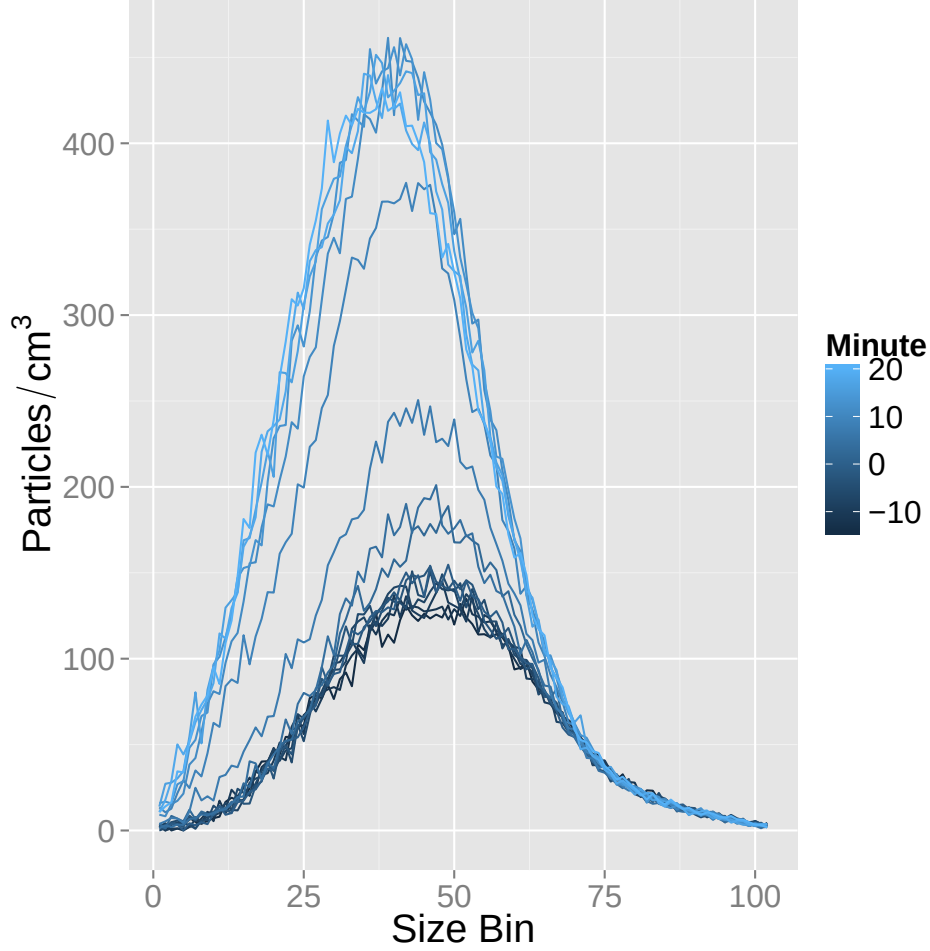
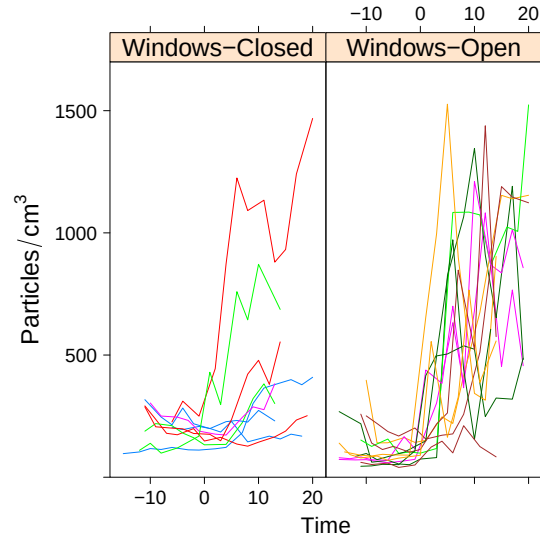
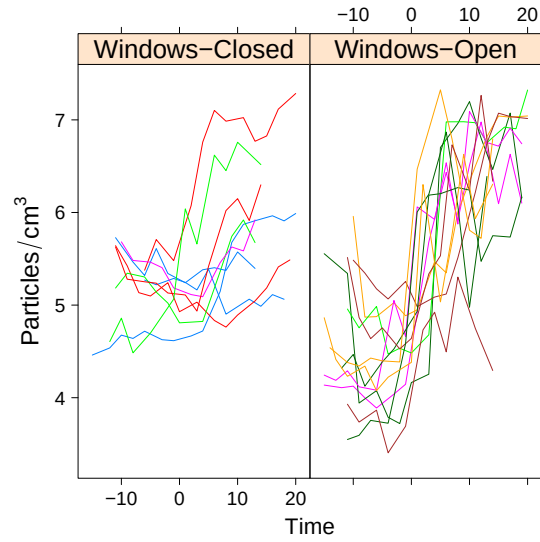


Figure 2.2: Plot of all UFP size distributions for one sample run. Time is measured in minutes from when the engine is turned on meaning measurement times range from approximately -15 to 20 minutes. The 7 UFP size distributions collected before the engine is turned on are plotted as darker curves and the 9 UFP distributions collected after the engine is turned on are plotted as progressively lighter curves. UFP size distributions become increasingly more peaked the longer the engine runs. The rate of increase in particle counts after the engine turns on varies greatly by size bin, with little to no increase seen above bin 70, and much larger increases in the 10-60 size bin range. This particular UFP size distribution has only one mode which occurs at smaller size bins as the engine continues to run while nearly tripling in magnitude.



(a)



(b)

Figure 2.3: Figures (a) and (b) show particle counts and log particle counts over time, respectively, for size bin 30 (20.9-21.7 nm) for all runs by window position. Each line is a separate run. Counts for size bin 30 generally increase sharply when the engine first turns on then continue to increase at slower rates thereafter, though in some cases increases are not seen, particularly for the window closed position. A log transformation allows for easier temporal modeling of counts.

2.3 A Time Series Semi-Parametric Model for UFP Size Distributions on School Buses

Let i index run, where $i = 1, \dots, R$ and for our data $R = 21$. Let s index particle size bin, with $s = 1, \dots, S$ and for our data $S = 102$. Time, t , has a run dependent range of $t_{\min,i}$ to $t_{\max,i}$. Time is measured in minutes and defined so that usually $t_{\min,i} = -14$ or -15 , always the engine is turned on at $t = 0$, and usually $t_{\max,i} = 19$ or 20 ; there is modest variation by run for $t_{\min,i}$ and $t_{\max,i}$. Baseline refers to time before engine-on, when $t < 0$. Let $z(i)$ be an indicator of window position where $z(i) = 1$ corresponds to windows-open and $z(i) = 0$ to corresponds to windows-closed. We write $z \equiv z(i)$ to simplify notation. The window position should only affect measurements after $t = 0$, not before. Outcome y_{ist} is the natural log of particle count plus 10 for run i , size bin s at time t . We add ten to the natural log to further smooth counts near zero.

2.3.1 Model

Before engine-on, baseline mean log counts are expected to be constant over time and are described by a hierarchical model with random run intercepts which vary as a function of size bin s . At baseline, we expect y_{ist} to have a size bin specific population mean, α_s , and size bin specific random intercept, γ_{is} . For $t < 0$, y_{ist} is modeled as

$$y_{ist} = \alpha_s + \gamma_{is} + u_{ist}. \quad (2.1)$$

Residuals u_{ist} are discussed shortly.

After engine-on, the y_{ist} increase additively from baseline levels. Let $\mathbf{f}(t)$ be a $J \times 1$ vector of functions of t , with j -th element

$$f_j(t) = \begin{cases} 0 & t < 0 \\ m_j(t) & t \geq 0. \end{cases} \quad (2.2)$$

For a model with quadratic trend, $J = 2$, $m_1(t) = t$ and $m_2(t) = t^2$. Other choices for $\mathbf{f}(t)$ are discussed in Section 2.3.2.

Engine-on increases in mean vary by window position z . Let δ_{zs} be a $J \times 1$ vector of size bin and window position specific regression coefficients for $\mathbf{f}(t)$. Then, for given s and z , the increase in mean log particle count from baseline is $\delta_{zs}^T \mathbf{f}(t)$.

Residuals u_{ist} are modeled with an autoregressive process over time, conditioning on the first observation. The model for any time t is

$$y_{ist} = \alpha_s + \gamma_{is} + \delta_{zs}^T \mathbf{f}(t) + u_{ist} \quad (2.3)$$

$$u_{ist} = \theta u_{i,s,t-1} + \epsilon_{ist}, \quad (2.4)$$

$$\epsilon_{ist} \sim N(0, \sigma_s^2) \quad (2.5)$$

where θ is the lag one correlation, and σ_s^2 is the variance of ϵ_{ist} given the previous residual $u_{i,s,t-1}$.

We want the baseline engine-off population mean, α_s , baseline random intercept, γ_{is} , and the coefficients for the time trends, δ_{zs} , to vary smoothly by size bin s so we model them as cubic B-spline functions of s . Let $\mathbf{B}(s)$ be a cubic $(K \times 1)$ B-spline basis over size bin s with $(K - 4)$ knots. Let $\boldsymbol{\alpha}$ be the coefficients of $\mathbf{B}(s)$ for the population mean, then

$$\alpha_s = \boldsymbol{\alpha}^T \mathbf{B}(s),$$

and let γ_i be the coefficients of $\mathbf{B}(s)$ for random intercept for run i , then

$$\gamma_{is} = \boldsymbol{\gamma}_i^T \mathbf{B}(s).$$

The vectors of random coefficients describing baseline UFP size distribution variation by run, $\boldsymbol{\gamma}_i$, have distribution

$$\boldsymbol{\gamma}_i \mid \mathbf{D} \sim N_K(0, \mathbf{D}), \quad (2.6)$$

for, $i = 1, \dots, R$. Together $\alpha^T \mathbf{B}(\mathbf{s}) + \gamma_i^T \mathbf{B}(\mathbf{s})$ model the baseline UFP size distribution for run i and size bin s .

Let δ_{zsj} be the j -th element of δ_{zs} . Set $\delta_{zsj} = \Delta_{zj}^T \mathbf{B}(\mathbf{s})$. We define Δ_z so that Δ_{zj} is the j -th row of Δ_z , $j = 1, \dots, J$. Then

$$\mathbf{f}(t)^T \Delta_z \mathbf{B}(\mathbf{s}) = \delta_{zs}^T \mathbf{f}(t) \quad (2.7)$$

There is one Δ_z , $J \times K$, for each window position, closed, Δ_0 , and open, Δ_1 .

The error variance σ_s^2 , is not constant and needs to vary smoothly over size bin. The log variance is modeled with a separate quadratic ($L \times 1$) B-spline basis over size bin, $\mathbf{B}_{\text{err}}(\mathbf{s})$, with $(L - 3)$ knots. Log variance is modeled

$$\log(\sigma_s^2) = \boldsymbol{\eta}^T \mathbf{B}_{\text{err}}(\mathbf{s}) + w_s \quad (2.8)$$

$$w_s \sim N(0, \tau_\eta^2) \quad (2.9)$$

where the w_s are small normal residual terms with known variance τ_η^2 added to $\boldsymbol{\eta}^T \mathbf{B}_{\text{err}}(\mathbf{s})$ to enable efficient MCMC sampling (Crainiceanu et al., 2007; Baladandayuthapani et al., 2005; Hadfield, 2010). We set τ_η^2 to a small number to not add too much variation.

2.3.2 Time Trend Models

Because the parametric form of count increases after the engine turns on has not been explored in current research, we consider several models for the time trend after engine-on.

2.3.2.1 Quadratic Time Trend

For the model with quadratic trend, $J = 2$, $m_1(t) = t$, $m_2(t) = t^2$ and Δ_z is a $2 \times K$ matrix with rows of coefficients of $\mathbf{B}(\mathbf{s})$ for the linear and quadratic time trend terms.

2.3.2.2 Bent Line Time Trend

In this model, the change in counts after the engine turns on follows a linear spline with single knot, allowing the slope to change at time t_Ω . Here $J = 2$, $m_1(t) = t$, and $m_2(t) = \max[0, t - t_\Omega]$. We consider knots at $t_\Omega = 8, 10$ or 12 .

2.3.2.3 Jump Models with a Quadratic or Linear Spline Time Trend

Next, we consider an immediate jump in counts when the engine first turns on followed by counts that change following either a quadratic or bent line time trend. The same knot times $t_\Omega = 8, 10, 12$ are considered. For example, the jump model with bent line time trend and knot at $t_\Omega = 8$ has $J = 3$, $m_1(t) = 1$ and $m_2(t) = t$ and $m_3(t) = \max[0, t - 8]$.

2.3.3 Random Jumps for the Time Trend

Initial log count increases modeled by the jump trend varied across run in part because the engine was turned on at different times relative to time t across runs. For the jump models, we considered random jumps as part of the time trend to determine if further improvements in model fit were possible. The random jump time trend uses the same B-spline basis $\mathbf{B}(\mathbf{s})$ to vary time trend coefficients by size bin. Let $\mathbf{g}(t)$ be a $G \times 1$ vector of functions of t , defined analogously to $\mathbf{f}(t)$ in Equation (2.2). Let Υ_i be defined as Δ_z in Equation (2.7), except Υ_i varies by run, not window position, and $\Upsilon_i \mid \mathbf{W} \sim N(0, \mathbf{W})$. A model with a random jump would have $\mathbf{g}(t) = 1$, $G = 1$, and Υ_i be a $K \times 1$ random vector. This notation allows for more complex random effects after engine-on. However, residual analysis did not indicate a need for additional random effects, see Figures 2.9, 2.10, and 2.11.

The model now takes the form

$$y_{ist} = \boldsymbol{\alpha}^T \mathbf{B}(\mathbf{s}) + \boldsymbol{\gamma}_i^T \mathbf{B}(\mathbf{s}) + \mathbf{f}(t)^T \boldsymbol{\Delta}_z \mathbf{B}(\mathbf{s}) + \mathbf{g}(t)^T \boldsymbol{\Upsilon}_i \mathbf{B}(\mathbf{s}) + u_{ist}, \quad (2.10)$$

along with Equations (2.4), (2.5), (2.6), (2.8), and (2.9).

Models with only quadratic or bent line time trends are referred to as *non-jump* models, while models with an engine-on jump are referred to as *jump* models. These models are described by Equation 2.10 without the term $\mathbf{g}(t)^T \boldsymbol{\Upsilon}_i \mathbf{B}(\mathbf{s})$. Models with the engine-on random jump are referred to as *random jump* models and are described by Equation 2.10.

2.3.4 Priors

Proper priors are used on all parameters.

Priors for the fixed effects $\boldsymbol{\alpha}$ and $\boldsymbol{\Delta}_{zj}$, $j = 1 \dots J$, are

$$\boldsymbol{\alpha} \mid \tau_\alpha \sim N_K(0, \tau_\alpha^2 \mathbf{I}_K), \quad (2.11)$$

$$\boldsymbol{\Delta}_{zj} \mid \tau_\delta \sim N_K(0, \tau_\delta^2 \mathbf{I}_K), \quad (2.12)$$

where $\mathbf{I}_K$ is the $K \times K$ identity matrix. Both τ_α^2 and τ_δ^2 are set to large values so that the priors are non-informative.

The prior for the correlation parameter θ is taken to be truncated normal as conjugacy improves computational speed

$$\theta \mid \mu_\theta, \sigma_\theta^2 \sim N(\mu_\theta, \sigma_\theta^2) \mathbf{I}[\theta \in (-1, .9)] \quad (2.13)$$

with the indicator function giving the lower -1 and upper $.9$ truncation points. The upper limit of 0.9 was chosen to avoid singularity in the likelihood (Palmer and Pettit, 1996). Covariance matrices $\mathbf{D}$ and $\mathbf{W}$ are given Inverse-Wishart priors

$$\mathbf{D} \sim \text{inverse Wishart}(n_D, (n_D - K - 1) * \mathbf{M}), \quad (2.14)$$

$$\mathbf{W} \sim \text{inverse Wishart}(G * K + 1, \mathbf{I}_{G*K}), \quad (2.15)$$

where the known $K \times K$ scale matrix $\mathbf{M}$ uses information from 65 other engine-off experiments reported in Zhang et al. (2012) but not used in the current analysis. Specifically, a linear mixed effects model using $\mathbf{B}(\mathbf{s})$ for the fixed and random UFP size distribution effects was run using log counts from this additional data. The estimated covariance matrix for the random effects from this fit was used as the scale matrix $\mathbf{M}$ in the Inverse-Wishart prior and we set $n_D = 65$. This allowed information available about typical correlations in UFP size distribution shapes from urban environments to be used, supplementing the sparse information available in the current dataset with only 21 runs and a $K = 7$ dimensional B-spline basis. Unfortunately there was no prior information as easily available for $\mathbf{W}$, so we set the scale matrix to give marginal uniform distributions to the correlation coefficients with low degrees of freedom and a prior mean of one for the variances.

The prior for the coefficients of the log variance is taken to be

$$\boldsymbol{\eta} \sim N_L(\log(0.4), g^2 \mathbf{I}_L) \quad (2.16)$$

where 0.4 was the mean residual error reported in the experiments of Zhang et al. (2012) that are not considered in this analysis, $\mathbf{I}_L$ is the $L \times L$ identity matrix, the hyperparameter g is set to put a 95 probability around residual variances between 0.2 and 0.7. The prior is centered at a constant residual across size bins.

2.3.5 Computing Overview

Posterior estimates are obtained via Markov Chain Monte Carlo (MCMC) simulations (Hastings, 1970; Gelfand and Smith, 1990) using JAGS 3.3.0 (Plummer, 2003). The MCMC convergence and mixing properties were assessed by visual inspection of the autocorrelation and chain histories of individual parameters. Convergence appeared to be immediate for all parameters.

2.3.6 Estimates of Interest

Let $\mathbf{Y}$ be the vector of all y_{ist} . To investigate model fit, we plotted subsets of Equation 2.10: baseline UFP size distributions, $\boldsymbol{\alpha}^T \mathbf{B}(\mathbf{s}) + \boldsymbol{\gamma}_i^T \mathbf{B}(\mathbf{s})$ and engine-on posterior time trend changes, $\mathbf{f}(t)^T \boldsymbol{\Delta}_z \mathbf{B}(\mathbf{s})$. Integrating out random engine-off and engine-on effects, predicted mean log counts at size bin s and time t are $\psi_{st} = \boldsymbol{\alpha}^T \mathbf{B}(\mathbf{s}) + \mathbf{f}(t)^T \boldsymbol{\Delta}_z \mathbf{B}(\mathbf{s})$

For emission monitoring purposes, posterior predictions must be transformed back to their original scale. Conditional on run i and random effects, predicted counts have marginal variance, $\lambda_s^2 \equiv \text{var}(y_{ist} \mid \boldsymbol{\gamma}_i, \boldsymbol{\Upsilon}_i)$ that is the same for all times t . Then, $\text{var}(u_{ist} \mid \boldsymbol{\gamma}_i, \boldsymbol{\Upsilon}_i) = \text{var}(\theta u_{i,s,t-1} + \epsilon_{ist} \mid \boldsymbol{\gamma}_i, \boldsymbol{\Upsilon}_i)$ so that $\lambda_s^2 = \theta^2 \lambda_s^2 + \sigma_s^2$ to give $\lambda_s^2 = \sigma_s^2 / (1 - \theta^2)$. Defining ρ_s as $\mathbf{B}(\mathbf{s})^t (\mathbf{D} + \mathbf{W}) \mathbf{B}(\mathbf{s})$, this implies a marginal predictive variance $\zeta_s^2 \equiv \rho_s + \lambda_s^2$ for size s at new run i for random jump models. No-jump, jump and random jump models define marginal predictive variance equivalently, but no-jump and jump models remove $\mathbf{W}$ from the definition of ρ_s . The back transformed mean of predicted counts for size bin s at time t is then

$$\mu_{st} = \exp(\psi_{st} + \zeta_s^2/2) \quad (2.17)$$

We define s_t as the modal size bin and the modal particle count as $h_t \equiv \text{argmax}(\mu_{st})$, where μ_{st} is defined in (2.17).

Residuals can be calculated as

$$e_{ist} = y_{ist} - (\boldsymbol{\alpha}^T \mathbf{B}(\mathbf{s}) + \boldsymbol{\gamma}_i^T \mathbf{B}(\mathbf{s}) + \mathbf{f}(t)^T \boldsymbol{\Delta}_z \mathbf{B}(\mathbf{s}) + \mathbf{g}(t)^T \boldsymbol{\Upsilon}_i \mathbf{B}(\mathbf{s})), \quad (2.18)$$

removing both fixed and random effects (Chaloner and Brant, 1988; Chaloner, 1994). Autoregressive error was not removed from Equation (2.18) to better visualize lack of fit in the models that may be picked up by the autoregressive term θ . Residuals for no-jump and jump models are calculated removing $\mathbf{g}(t)^T \boldsymbol{\Upsilon}_i \mathbf{B}(\mathbf{s})$.

2.4 Results

We present results graphically to convey important model inferences as meaningful representations of conclusions are difficult to convey in table form.

2.4.1 Model Selection

Figure 2.4 presents window-open posterior predictive means for μ_{st} as functions of s for no-jump, jump, and random jump models at $t = 5, 15$ and 20 minutes after the engine is turned on: each sub-figure has estimates for the quadratic and the three knot models. Predictive mean counts for the knot and quadratic models differ within no-jump models, but are very similar within the jump models. Predictive mean counts are very similar within the random jump models as well. The non-jump quadratic model differs from knot models in that it predicts counts to eventually decrease by time $t = 20$. However, this decrease is diminished with the addition of the fixed jump, and disappears with the random jump. The random jump models give the highest predicted means across size bin for all time points. Credible intervals are not plotted to ease visual interpretation, but there is overlap in these intervals for all models.

Mean predictive posterior counts for the windows-closed position for all models after the engine has been idling are presented in Figure 2.5 for $t = 5$, row 1(a)(b)(c); $t = 15$, row 2(d)(e)(f); and $t = 20$, row 3(g)(h)(i). Non-jump models are in the left column (a)(b)(c), jump models are in the middle column (d)(e)(f), and random jump models are in the right column (g)(h)(i). Unlike for the windows-open position, windows-closed results are very similar across all model types, including models with and without jumps and random jumps.

Model choice was through a combination of DIC (Spiegelhalter et al., 2002) and graphical examination of residuals, which included both narrowing the range of mean posterior residuals and the number of outliers. DIC and components for

each model are presented in Table 3.2. D bar measures how well the model fits the data, the smaller it is the better fits the data, while pD is the effective number of parameters, and acts a penalization for over complicated models. DIC is the sum of these two components.

Table 2.1: DIC Calculations for School Bus Models

Model	DIC	DIC bar	pD
Quadratic	3,082.3	2,858.7	223.6
Knot 8	3,509.7	3,285.8	223.9
Knot 10	3,715.9	3,492.4	223.4
Knot 12	3,913.6	3,690.9	222.7
Jump Quadratic	1,587.2	1,348.6	238.6
Jump Knot 8	1,640.8	1,402.3	238.6
Jump Knot 10	1,624.4	1,386.0	238.4
Jump Knot 12	1,614.5	1,376.1	238.4
Random Jump Quadratic	-3,928.3	-4,291.0	362.7
Random Jump Knot 8	-3,887.6	-4,250.2	362.6
Random Jump Knot 10	-3,903.6	-4,265.6	362.1
Random Jump Knot 12	-3,896.3	-4,258.8	362.4

The model with the lowest DIC is the preferred model. The random jump models have much lower DICs than jump models, which have much lower DICs than the non-jump models.

The posterior median of the AR parameter $\hat{\theta}$ for the jump quadratic model without any random effect was .90 with a 95 percent credible interval (.89, .90), recall the prior was truncated at .9. Adding engine off random effects reduced this to 0.55 with credible interval (.54, .56), while adding the engine on random jump effect further reduced the estimate to .37 with credible interval (.35, .38).

Residual estimates and AR estimates for other models show similar patterns.

The random jump quadratic model had the best (lowest) DIC score and, along with all random jump models, the lowest estimates for the AR parameter.

2.4.1.1 Selected Residual Checks

Models were further evaluated by plotting mean posterior residuals, $\bar{e}_{ist} \equiv E[e_{ist} | \mathbf{Y}]$, by time and by size. Figure 2.6 presents residuals $\bar{e}_{ist}$ for the quadratic, jump quadratic, and random jump models plotted against size in (a)(c)(e) and against time in (b)(d)(f). Adding the jump component to the model narrowed the range of mean posterior residuals and reduced outliers. Adding the random jump component reduced these to an even greater extent.

Figure 2.7 shows improvement in fit following the addition first of engine-off random effects, and then also jump random effects to a fixed effect only model using the quadratic trend. Figures 2.7(a) and (d) plot a subset of residuals colored by run and show residuals were extremely correlated by run before any random curves were included. Figures 2.7(b) and (e) plot the same subset of residuals after engine-off random curves by run are added to the model. Residuals by run are more mixed on the plot, indicating correlation by run is removed to a large extent, though it is still present. Figures 2.7(c) and (f) plot residuals after the random jump is added. Residual mixing is further increased. The residual variance is decreasing from Figures 2.7(a)(d) to (b)(e) to (c)(f) with the further addition of random curves due to the removal of variation.

Figure 2.8 plots posterior mean baseline UFP size distributions $E(\boldsymbol{\alpha}^T \mathbf{B}(\mathbf{s}) + \boldsymbol{\gamma}_i^T \mathbf{B}(\mathbf{s}) | \mathbf{Y})$, observed log counts y_{ist} , and residuals $\bar{e}_{ist}$ for two sample runs from when the engine is off. Posterior means are plotted with green curves, observed log counts are plotted with red dots, and residuals are plotted in black. The baseline UFP size distributions on the left, Figures 2.8(a) and (c), are fit with the jump

quadratic model, while those on the right, Figures 2.8(b) and (d) are quadratic random jump. The improvement in fit from Figure 2.8(a) to (b) and from Figure 2.8(c) to (d) shows the value of adding the engine-on random jump.

Figures 2.9, 2.10, and 2.11 show residual profile plots over time for select size bins for the jump quadratic (left column) and random jump quadratic (right column) models. The portion of the plot to the right of the vertical black line plots engine-on. Adding the random jump narrows residual range for engine-off for the entire range of sizes. Residual range for engine-on is also narrowed to a lesser extent. Because of the lowest DIC score, AR θ estimate, and superior fit via residual examination, the random jump quadratic model was selected to present further inferences.

2.4.2 Results for Inferences of Interest

Residual variance shows heteroskedasticity over size bin, as shown in Figure 2.6, and Figure 2.12 plots posterior median and 95 percent credible intervals for σ_s^2 by bin size.

Figure 2.13 plots baseline posterior mean log counts by size bin for $t < 0$ for all runs i . The plots show significant variation in baseline UFP size distributions across run.

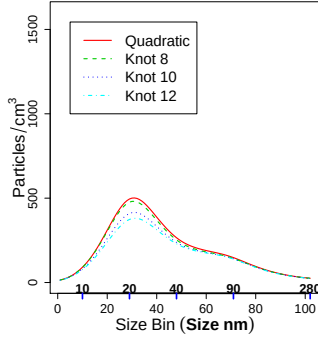
Subsequent inferences are discussed in terms of approximate particle sizes rather than size bins to facilitate interpretation of results. Time trend components are presented at $t = 15$ in Figure 2.14. Figure 2.14(a) plots the jump component, $\Delta_{z1}^T \mathbf{B}(s)$, (b) plots 15 times the linear component, $15 * \Delta_{z2}^T \mathbf{B}(s)$, and (c) plots 15^2 times the quadratic component $15^2 \Delta_{z3}^T \mathbf{B}(s)$ for both window positions. Adding these curves together gives the net log particle count increase after 15 minutes. The jump component is negligible for the window closed position, but there is an initial jump estimated for the window open position for all

size bins, with the greatest increase for particles between 10 and 40nm. Credible intervals do not overlap between window positions for sizes between 15 and 30 nm. For particle sizes between 10 and 90 nm, the linear component is larger for the window open position than the window closed position as credible intervals do not overlap. Negative values across size bin for both window positions in the quadratic component implies that that particle counts increase at slower rates for all bins as the engine continues to run, though these rate decreases are smaller across size for the window closed position.

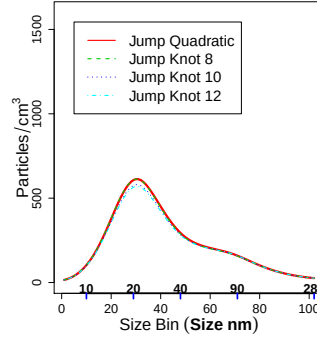
In Figure 2.15, UFP size bin distributions, μ_{st} , are plotted for engine-off and window open and closed after the engine has been idling for (a) 5, (b) 10, (c) 15, and (d) 20 minutes for the random jump quadratic model. Adding the random jump both increased predicted means and widened credible intervals compared to models without it. The window open position has higher predicted counts for sizes between about 15 and 40 nm at all time points compared to the window closed position and also becomes much more peaked over time as its mode gets higher and higher. The windows-closed position predictions increase over time at a slower, more constant rate. While for the first 10 minutes credible intervals between the engine-off and window closed curves overlap slightly, at 15 minutes particles of sizes less than 30nm show increases from the engine-off state which increase further at 20 minutes. Closing the windows hinders self pollution over time but does not prevent it.

Figure 2.16 plots medians of the posterior mode locations, s_t and heights, h_t . The location of the mode in the engine-off state is uncertain, with 95 percent credible intervals (32, 66), equivalent to a size range of 22 – 76nm (full credible interval not shown on plot). In 2.16(a), the window-open mode location decreases in size and the credible interval rapidly narrows to finally reach (24, 29) at 20 minutes. The window-closed position mode location in 2.16(b) decreases at a slower rate and with more uncertainty: at 20 minutes the 95 percent credible

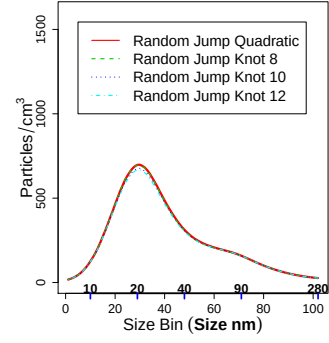
interval is $(22, 32)$. In Figure (c), posterior medians of the heights h_t reach counts of 1,475 for window open at $t = 20$ compared to about 390 for window-closed. The window-closed position has posterior mode heights that increase at an almost linear rate over time that is slower than for the windows-open position. Credible intervals do not overlap, but get wider over time for both window positions.



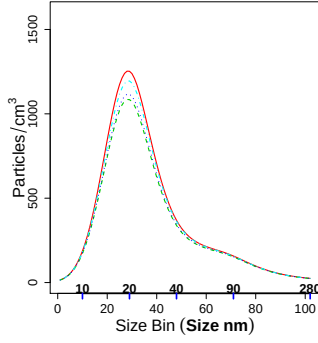
(a) 5 minutes, Non-jump



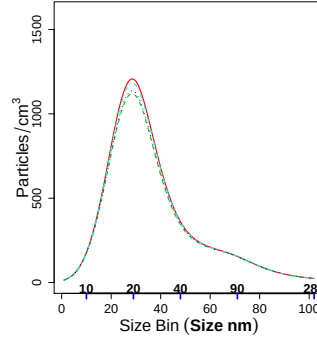
(b) 5 minutes, Jump



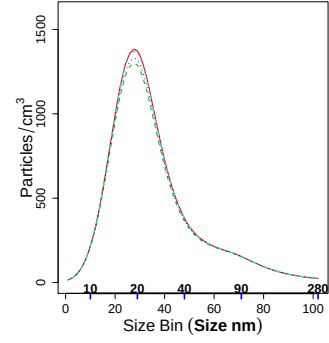
(c) 5 minutes, Random Jump



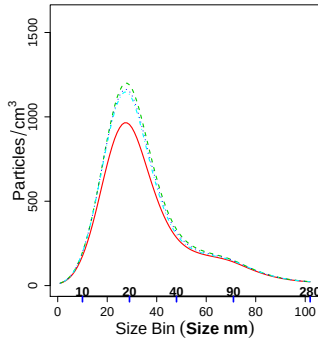
(d) 15 minutes, Non-jump



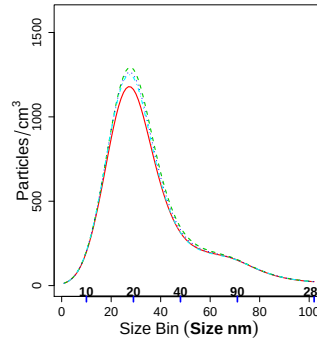
(e) 15 minutes, Jump



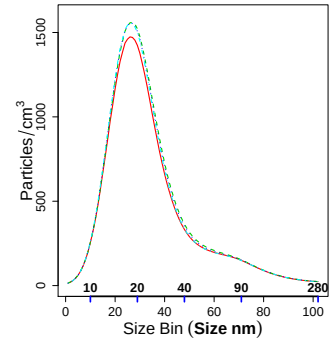
(f) 15 minutes, Random Jump



(g) 20 minutes, Non-jump

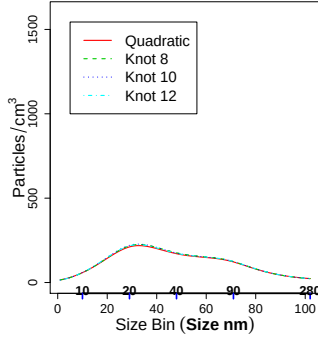


(h) 20 minutes, Jump

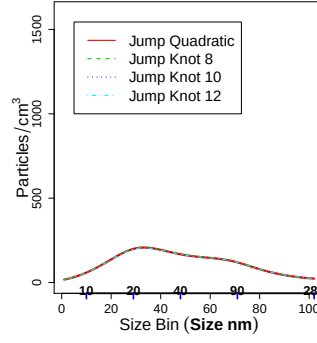


(i) 20 minutes, Random Jump

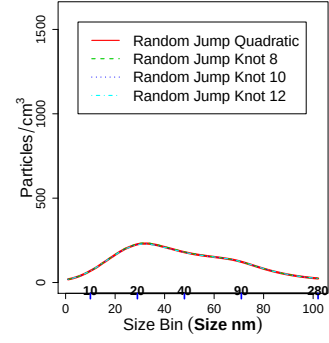
Figure 2.4: Mean predictive posterior counts for the windows-open position for all models after the engine has been idling for $t = 5$, row 1(a)(b)(c); $t = 15$, row 2(d)(e)(f); and $t = 20$, row 3(g)(h)(i). Non-jump models are in the left column (a)(c)(g), jump models are in the middle column (b)(d)(e)(f)(h), and random jump models are in the right column (c)(f)(i).



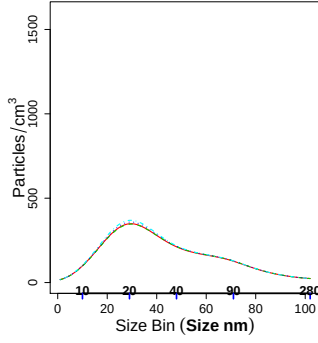
(a) 5 minutes, Non-jump



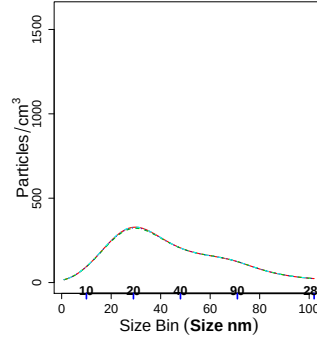
(b) 5 minutes, Jump



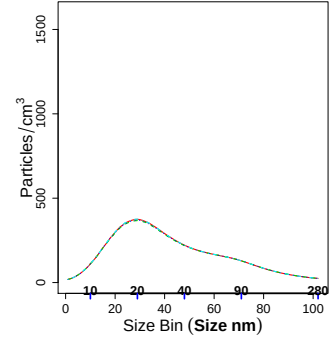
(c) 5 minutes, Random Jump



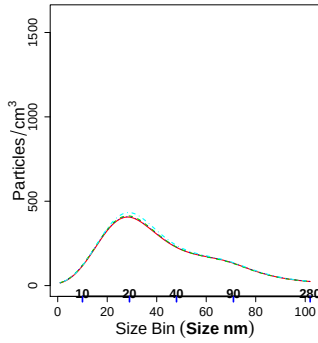
(d) 15 minutes, Non-jump



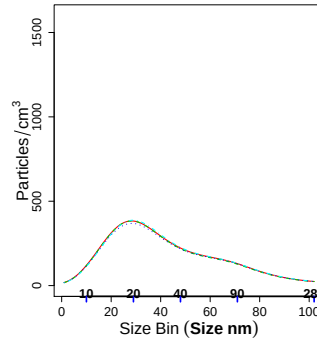
(e) 15 minutes, Jump



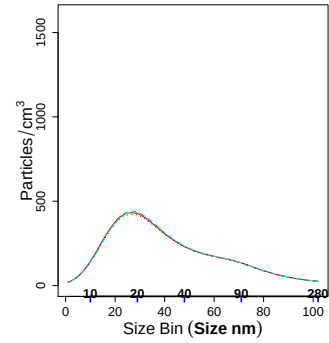
(f) 15 minutes, Random Jump



(g) 20 minutes, Non-jump

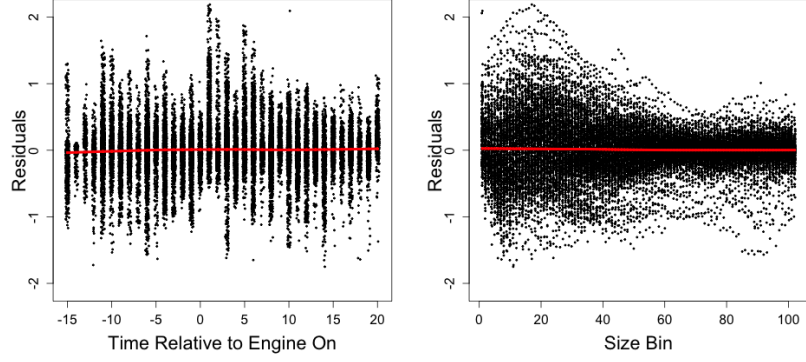


(h) 20 minutes, Jump

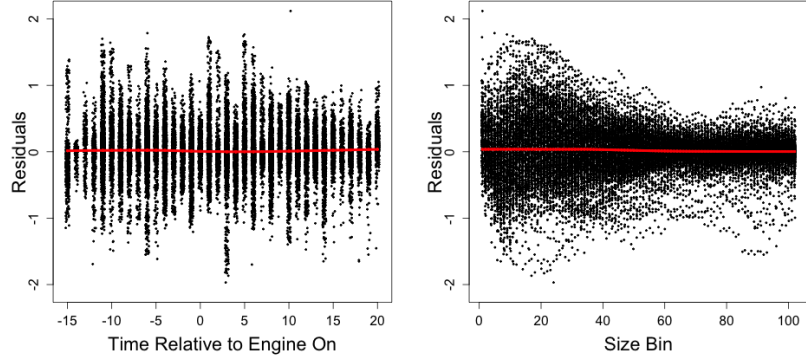


(i) 20 minutes, Random Jump

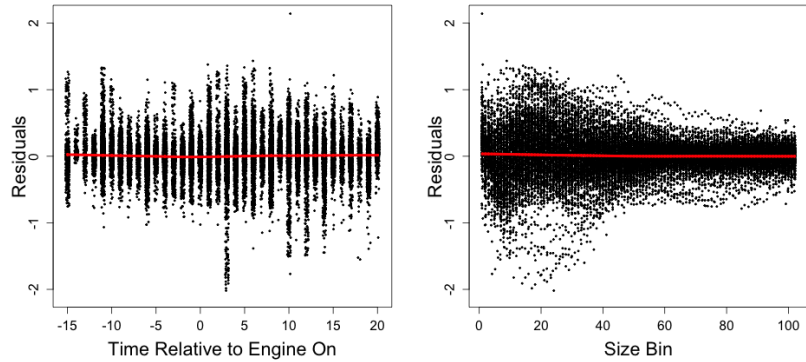
Figure 2.5: Mean predictive posterior counts for the windows-closed position for all models after the engine has been idling for $t = 5$, row 1(a)(b)(c); $t = 15$, row 2(d)(e)(f); and $t = 20$, row 3(g)(h)(i). Non-jump models are in the left column (a)(c)(g), jump models are in the middle column (b)(d)(e)(f)(h), and random jump models are in the right column (c)(f)(i).



(a) Quadratic $\bar{e}_{ist}$ Over Time (b) Quadratic $\bar{e}_{ist}$ Over Size



(c) Jump Quadratic $\bar{e}_{ist}$ Over Time (d) Jump Quadratic $\bar{e}_{ist}$ Over Size



(e) Rand. Jump Quad. Over Time (f) Rand. Jump Quad. Over Size

Figure 2.6: Residuals $\bar{e}_{ist}$ for the quadratic, jump quadratic, and random jump models plotted against time and size bin.

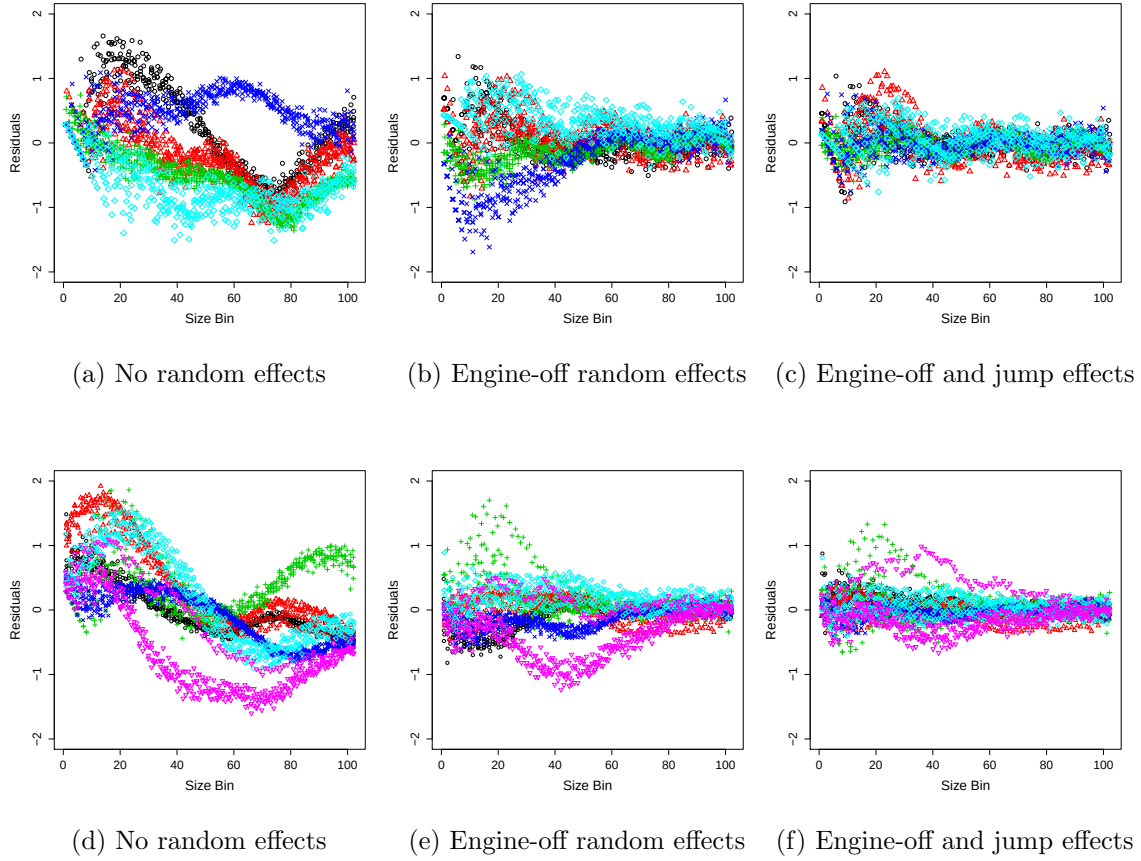


Figure 2.7: Improvement in fit following the addition of engine-off random effects, and then both engine-off and jump random effects to a fixed effect only model. Figures (a) and (d) plot a subset of residuals colored by run and show residuals were extremely correlated by run before any random curves were included. Figures (b) and (e) plot the same subset of residuals after engine-off random curves by run are added to the model. Residuals by run are more mixed on the plot, indicating correlation by run is removed to a large extent, though it is still present. Figures (c) and (f) plot residuals after the random jump is added. Residual mixing is further increased. The variance of residuals is decreasing from Figures (a)(d) to (b)(e) to (c)(f) with the further addition of random curves due to the removal of variation.

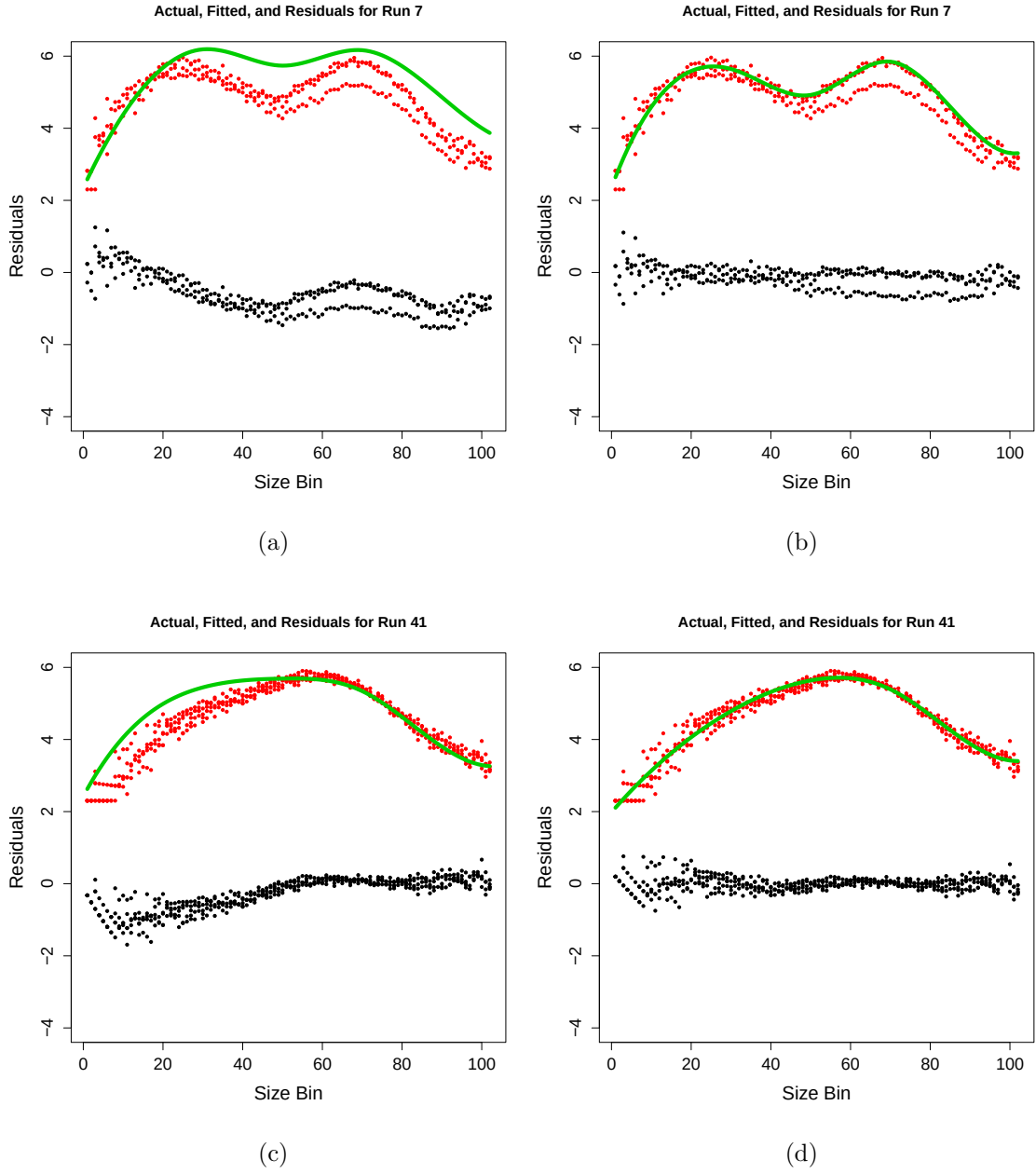
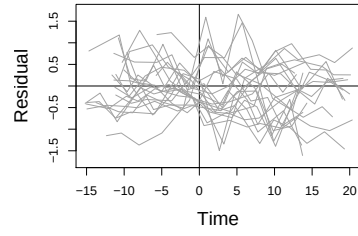
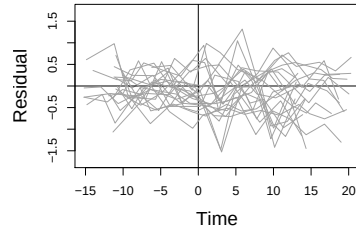


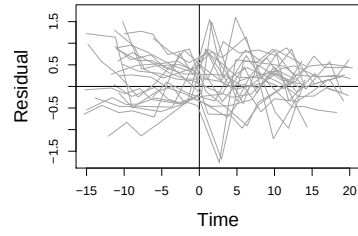
Figure 2.8: Posterior mean baseline UFP size distributions $E(\alpha^T \mathbf{B}(s) + \gamma_i^T \mathbf{B}(s) \mid \mathbf{Y})$, observed log counts y_{ist} , and residuals $\bar{e}_{ist}$ for two sample runs from when the engine is off. Posterior means are plotted with green curves, observed log counts are plotted with red dots, and residuals are plotted in black. The baseline UFP size distributions on the left, Figures (a) and (c), are fit with the jump quadratic model, while those on the right, Figures (b) and (d) are fit with the quadratic random jump model.



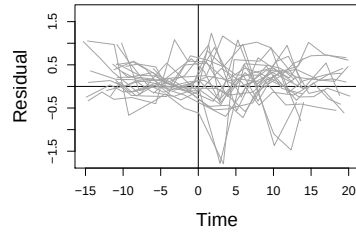
(a) Size 10, no random jump



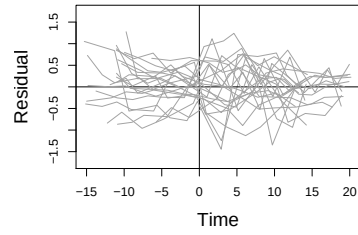
(b) Size 10, random jump



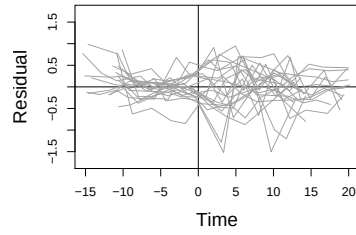
(c) Size 20, no random jump



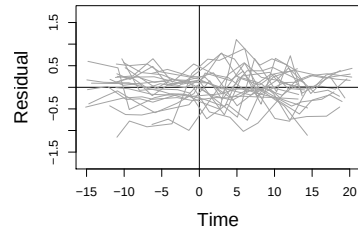
(d) Size 20, random jump



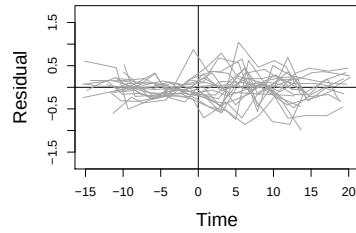
(e) Size 30, no random jump



(f) Size 30, random jump

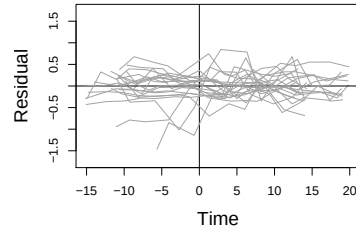


(g) Size 40, no random jump

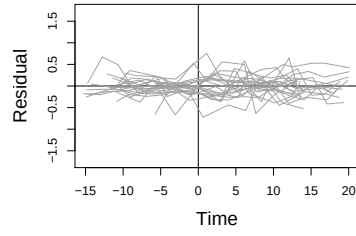


(h) Size 40, random jump

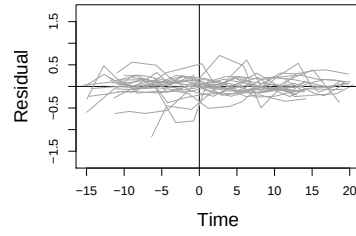
Figure 2.9: Residual profile plots over time for size bins 10, 20, 30, and 40 for the jump quadratic (left column) and random jump quadratic models (right column). The vertical black line indicates engine-on.



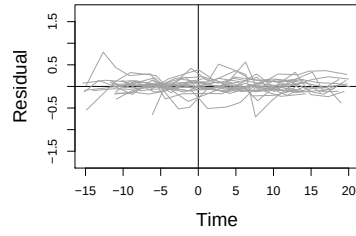
(a) Size 50, no random jump



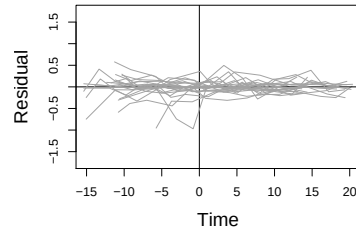
(b) Size 50, random jump



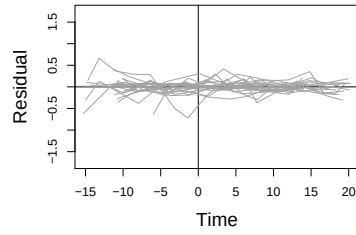
(c) Size 60, no random jump



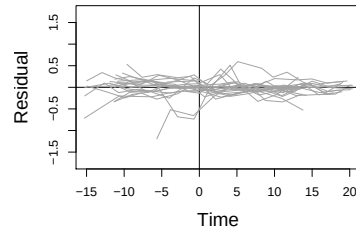
(d) Size 60, random jump



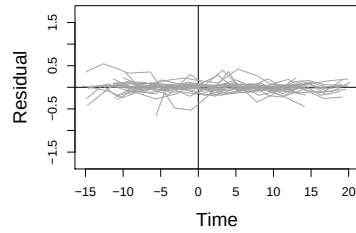
(e) Size 70, no random jump



(f) Size 70, random jump



(g) Size 80, no random jump



(h) Size 80, random jump

Figure 2.10: Residual profile plots over time for size bins 50, 60, 70, and 80 for the jump quadratic (left column) and random jump quadratic models (right column). The vertical black line indicates engine-on.

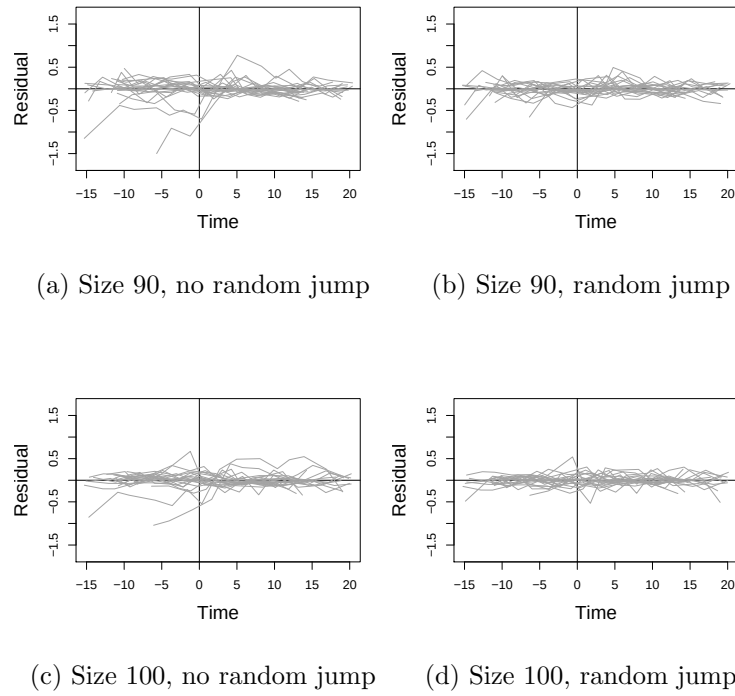


Figure 2.11: Residual profile plots over time for size bins 90 and 100 for the jump quadratic (left column) and random jump quadratic models (right column). The vertical black line indicates engine-on.

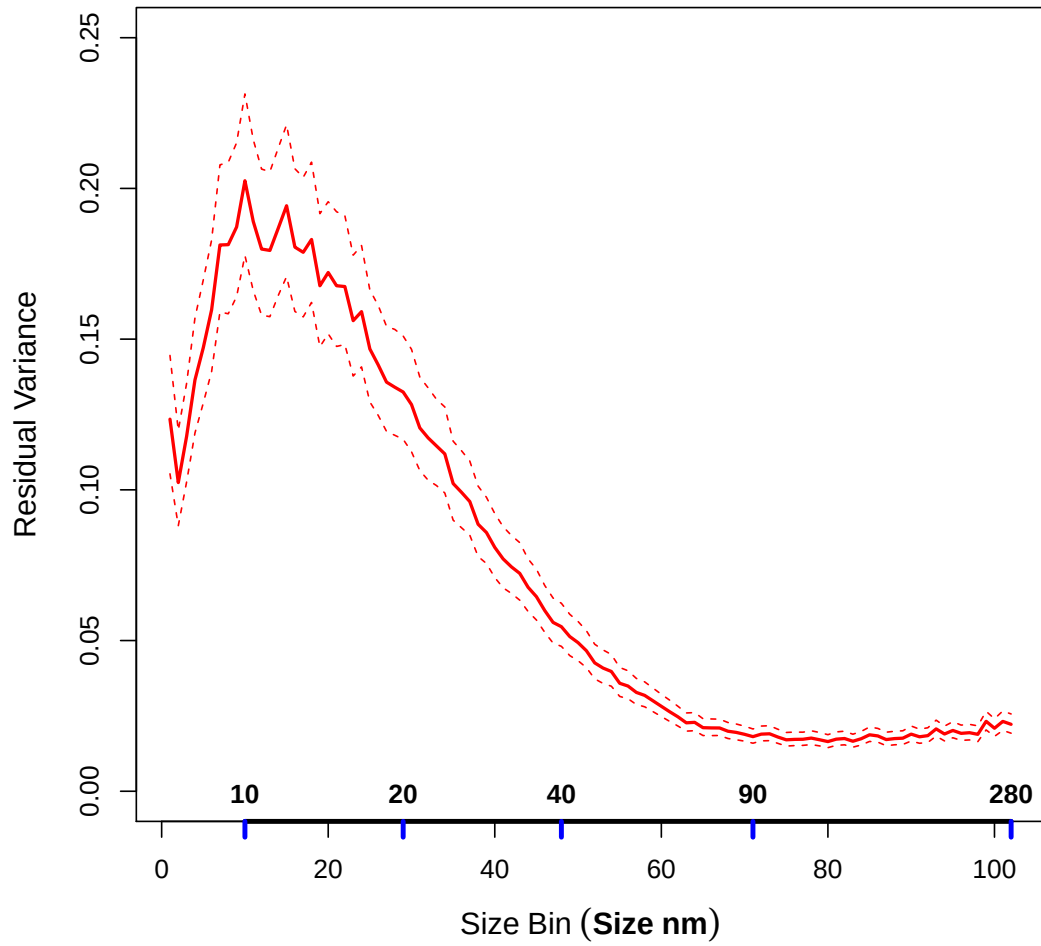


Figure 2.12: Posterior medians and 95 percent credible intervals for σ_s^2 as a function of size bin.

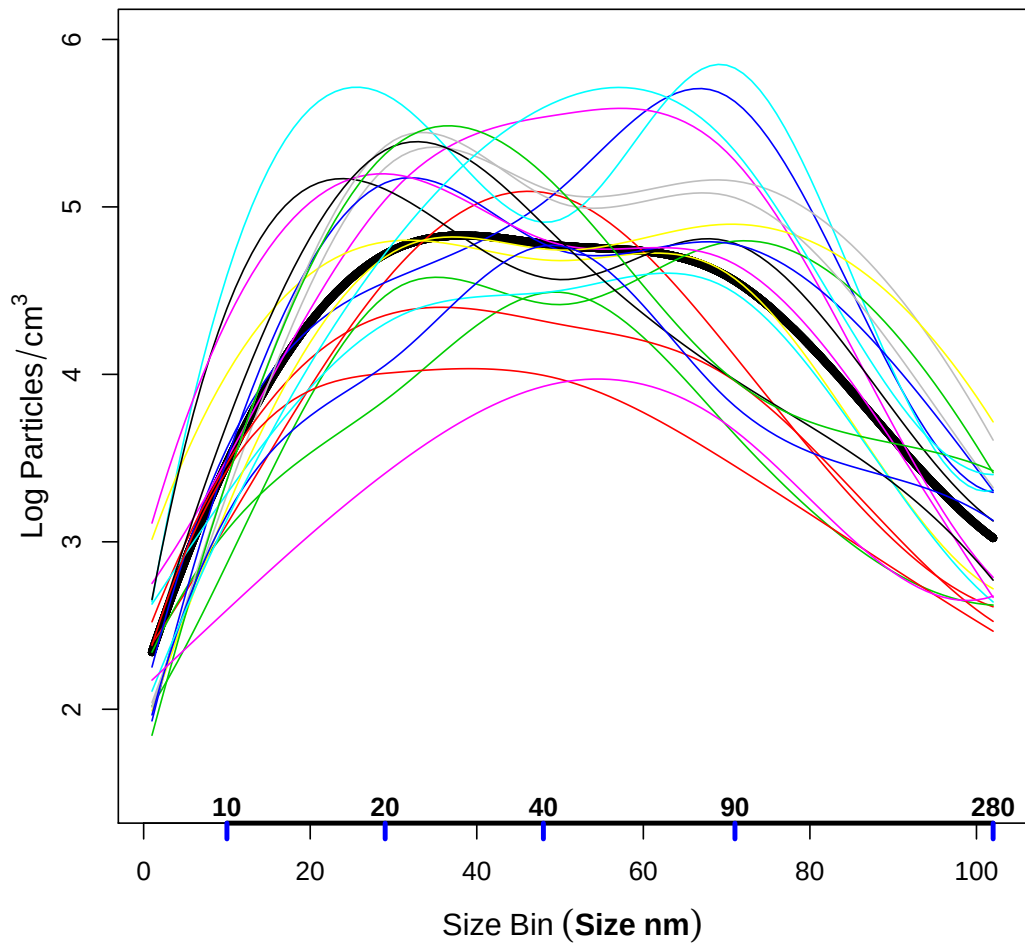
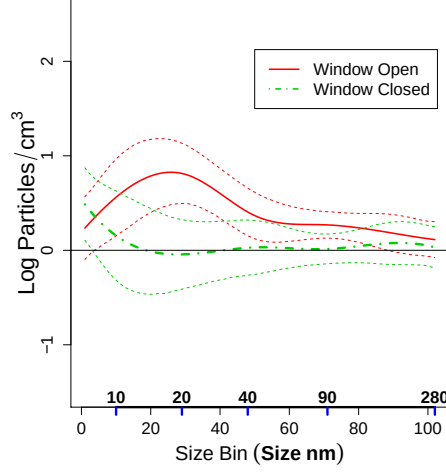
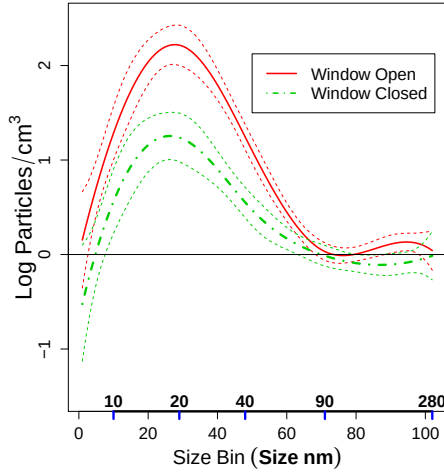


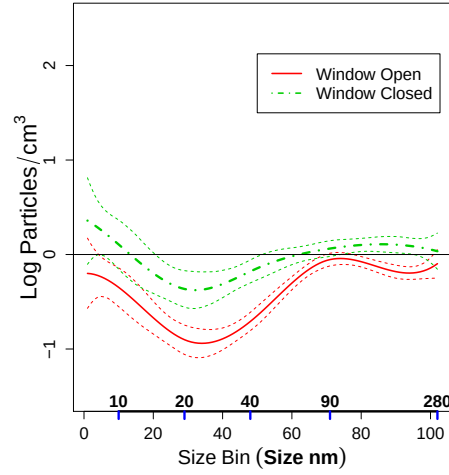
Figure 2.13: Posterior mean log baseline counts by size bin for $t < 0$. The thick black curve is the posterior population mean and the lighter curves show posterior mean log counts by size bin for individual run i .



(a) Initial Engine-On Jump

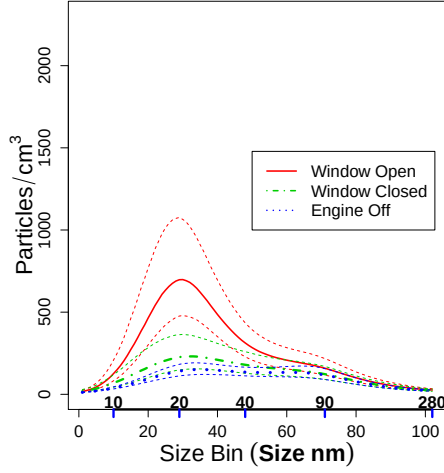


(b) Linear Trend

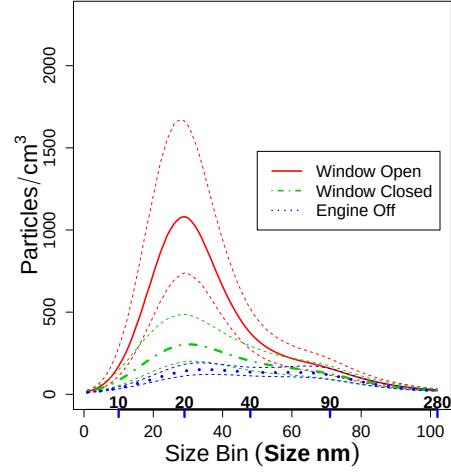


(c) Quadratic Trend

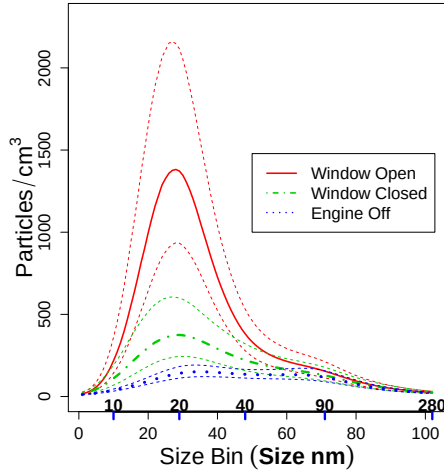
Figure 2.14: Fixed time trend components on the log scale. Figure (a) plots the jump component, $\Delta_{z1}^T \mathbf{B}(s)$, (b) plots the linear component, $15 * \Delta_{z2}^T \mathbf{B}(s)$, and (c) plots the quadratic component $15^2 \Delta_{z3}^T \mathbf{B}(s)$ for both window positions. Adding these curves together gives the net log particle count increase after 15 minutes. Dotted lines display ninety-five percent posterior intervals.



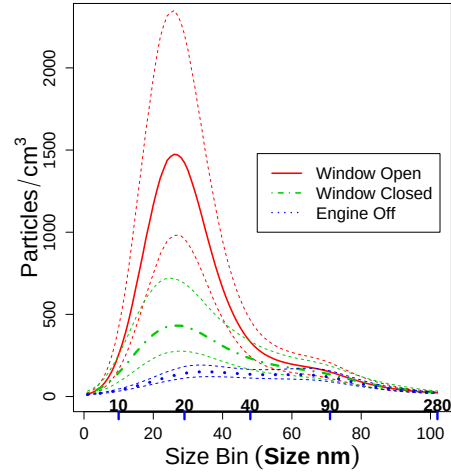
(a) Idling 5 minutes



(b) Idling 10 minutes



(c) Idling 15 minutes



(d) Idling 20 minutes

Figure 2.15: UFP size distributions, μ_{st} for engine-off is plotted in all figures; window open and closed after the engine has been idling for (a) 5, (b) 10, (c) 15, and (d) 20 minutes for the random jump quadratic model. Solid lines are medians and dashed are 95 percent credible intervals. Adding the random jump both increased predicted means and widened credible intervals.

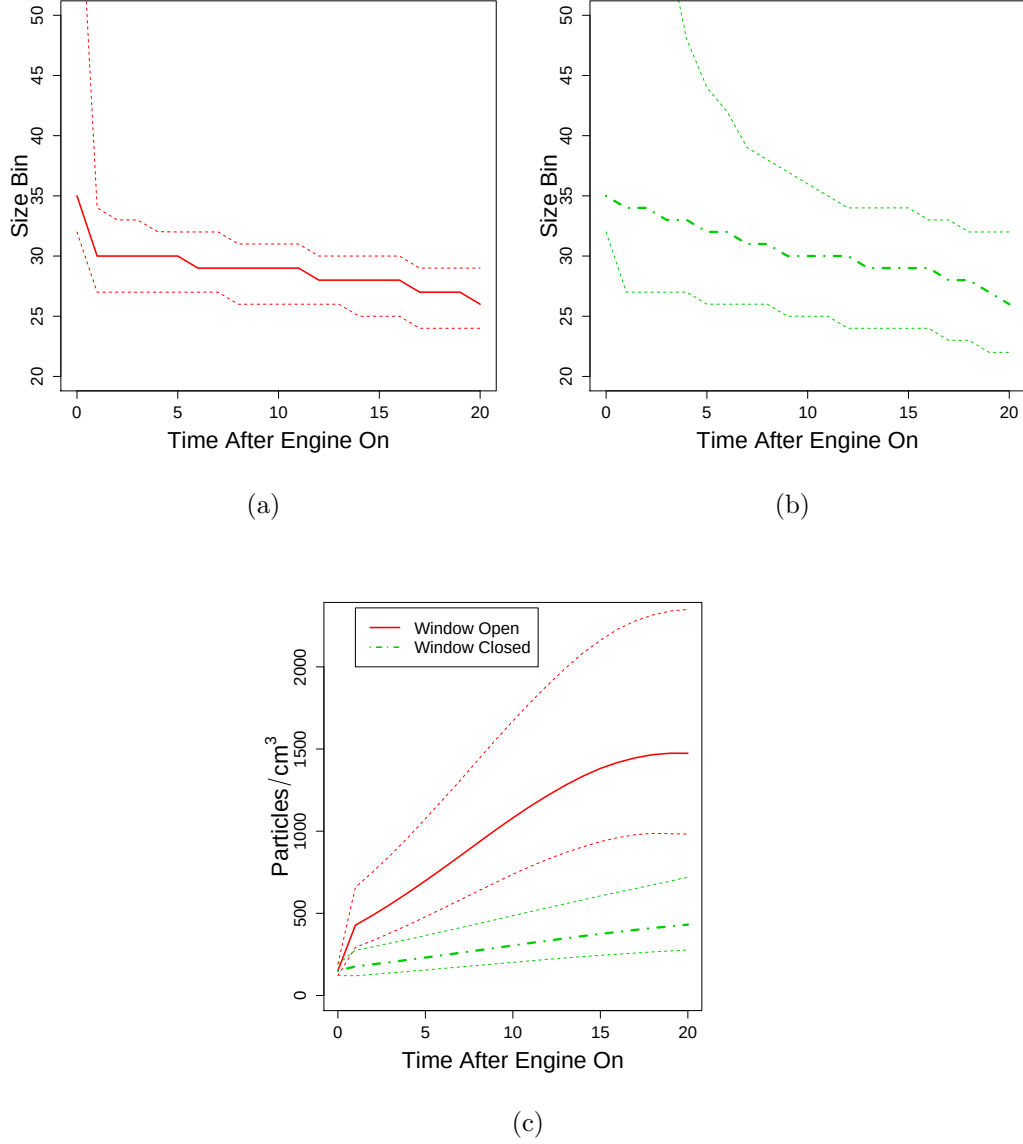


Figure 2.16: Median posterior mode locations s_t for (a) window-open and (b) window-closed and (c) heights over time h_t for both window positions with 95 percent credible intervals. The location of the mode in the engine-off state is uncertain, with 95 percent credible intervals (32, 66), equivalent to a size range of 22 – 76nm (full credible interval not shown on plot). In Figure (a), the window-open mode location rapidly narrows while in (b) the window-closed position mode location decreases at a slower rate and with more uncertainty. Posterior mode heights h_t in Figure (c) for the window-closed position increase at a slower almost constant linear rate over time.

2.5 Discussion

Our functional time-series methods greatly reduce the size of confidence intervals compared to the non-functional univariate longitudinal methods used in Zhang et al. (2012) that show overlap for both window positions at all size bins. Our methods permit inference about actual particle counts, which are not available using modal methods.

At all time points the windows-open position had higher predictions for particles between about 15 and 40 nm than windows-closed, but predictions for the windows-closed position continued to increase over time. Windows-closed credible intervals did not overlap with engine-off credible intervals after the engine had been idling for 10 minutes. Having the window closed does not prevent UFPs from entering the bus cabin, particularly for particles less than 40 nm. From a public health perspective, this supports anti-idling practices which limit bus idling to 5 minutes or less. After that time period, harmful UFPs of the smallest sizes are entering into the bus cabin, possibly causing exposure to the children inside.

A major benefit of these methods are the ease in which model fit can be evaluated, both through the use of DIC and through residual checks allowing visual inspection of model fit across size bin and time. Graphical techniques also allow for the evaluation of the fit of random effects. Our methods are easily adapted to different model specifications, such as a different form for the time trends or engine-on random effects and other variance structures.

Our methods allow uncertainty in predictions to be quantified and communicated graphically in ways easily interpretable to a non-statistician. We provide statistical summaries of useful quantities currently used by researchers such as mode locations and heights. Other quantities that researchers may find interesting in the future, along with their uncertainties, can also be calculated with ease.

Using our methods, we show that even with windows closed, engine idling allows UFP particles into the school bus cabin. This can be used as support for anti-idling policies currently in place. Additionally, vehicle emission standards are currently based on particulate matter mass measurements that fail to capture exposure to UFPs. There is growing concern about the suitability of a single metric based on particle mass as the standard for regulation (Ning and Sioutas, 2010). Our methods provide a basis to monitor exposure to UFPs based on particle counts separated into particles of different groups of size bins. Given UFPs' toxic nature with health effects strongly related to particle size bin, these methods provide information needed for regulating vehicle emissions to minimize UFP exposure in the future. Our methods allow evaluation and comparison of emissions by particle size for UFPs not captured in current emission standards.

CHAPTER 3

Carmageddon

3.1 Introduction

Ultrafine particles (UFPs) are particulate matter with diameters less than 100 nm. UFPs' small size and large surface area allow them to penetrate the lung, enter the circulatory system, and deposit in the brain (Oberdorster et al., 2004; Samet et al., 2009) and it has been suggested that they are more toxic to humans than larger particles (Alessandrini et al., 2006; Delfino et al., 2005; Ferin et al., 1990; Frampton et al., 2006). Health effects of UFPs are linked to particle size which determines the region in the lung the particles deposit (Morawska et al., 2008).

Elevated concentrations of UFPs are commonly found near roadways and exposure to traffic-related air pollutants has been linked to adverse health effects by epidemiological cohort studies (Beelen et al., 2007; Laden et al., 2006; Zhu et al., 2002). Due to the significant contribution of air pollutants from traffic emissions, several studies have examined how traffic calming policies such as traffic restriction could be a rational strategy to mitigate urban air pollutants (Friedman et al., 2001; Levy et al., 2006; Wang et al., 2009; Whitlow et al., 2011). However, how UFP counts vary by particle size near major roadways over time under different traffic conditions is not yet well understood. In July 2011, one of the busiest freeways in United States, the I-405, was scheduled for a two-day closure as part of a freeway improvement project. The closure event, dubbed *Carmageddon* provided

a valuable opportunity to conduct a natural air quality experiment by allowing comparison of UFP counts under high and low traffic conditions and to see how these evolve over time.

Researchers collected particle counts near the I-405 during Saturdays preceeding, during, and after the 2011 scheduled closure event, as well as on Fridays and Sundays preceeding and during the closure event. Scanning Mobility Particle Sizers (SMPS) counted particles per cubic centimeter in 102 *size bins* that group particles by diameter ranging from the first size bin containing particles of the smallest diameters, 7.37–7.64 nm, to the last size bin containing sizes of 269.0–278.8 nm. The ordered collection of counts in these 102 size bins at a single point in time is called a *UFP size distribution*, even though (i) technically particles with diameters greater than 100 nm are too big to be UFPs and (ii) the counts are not a distribution in the statistical sense. Size bin widths are approximately equally spaced on a log scale, so UFP size distributions have more bins for the smaller UFP particles of interest. UFP size distributions were collected at two minute intervals over the course of a day.

Numerous mathematical representations have been used to describe size distributions over size bin and time, typically via *modal methods* (Whitby, 1978; Whitby et al., 1991). Modal methods model the particle size distribution as a mixture of densities (Hussein et al., 2005; Whitby et al., 1991), ignoring the total number of particles. More recently, Wraith et al. (2009, 2014) model time series of UFP size distributions using time-varying non-parameteric Bayesian mixture models. Modal methods standardize particle counts: only information about the relative composition of particle size bins is retained. In contrast, in modeling UFP exposure near roadways, understanding actual particle counts is crucial in order to properly model exposure; thus modal methods are insufficient for this application. Modal methods have also not accounted for sampling variances in observed UFP counts. UFP counts in smaller size bins have larger variability than

those in larger size bins, as they tend to be more unstable than larger particles, following dynamic processes which have them desorb, deposit, or combine to form larger particles at fairly rapid rates (Kulmala et al., 2004; Kittelson et al., 2006). In urban environments UFP counts vary for certain particle size bins more than others because they are created by anthropogenic processes that fluctuate over time. Statistical methods that model mean UFP counts by particle size bin and over time must account for increased residual variance for smaller particle size bins.

Modeling UFP size distributions using functional time-series methods allows for inference on particle counts while accounting for differences in residual variance across particle size bin. It is also an improvement on the methods in Quiros et al. (2013), who considered only day-long summaries of UFP counts near the I-405. As we are interested in how these counts evolve over time under different traffic conditions, the time series methods developed in this analysis will greatly aid future research.

We propose Bayesian functional time series models to model how UFP counts change over time near the I-405 roadway. We model UFP size distributions at a given time with a cubic B-spline basis (de Boor, 1978). UFP counts then change over time at a size bin dependent rate by letting B-spline coefficients evolve over time following a random walk model (Prado and West, 2010; West and Harrison, 1997). This model smooths UFP counts both over size bin and over time, allowing for inference on count trends over time across various size bins.

To account for larger count variance for smaller particles relative to larger particle size bins, the log residual variance over size bin is modeled using a quadratic B-spline basis. We also explore modeling residuals with an autoregressive model to accommodate correlation over time.

Interest centers on how particle counts evolve over time near the I-405 as a function of size bin. Researchers are also interested in number of local modes of

particle size bin counts, local mode heights, and how modes evolve over time near high traffic roadways. Because combustion engines emit particles of extremely small diameter and these particles are also found to be the most toxic, researchers are interested in which conditions produce local modes in the smallest particle sizes, how high these local modes are, and when these reach their maximum heights. We provide summaries of how local modes and mode heights evolve near the I-405 and compare these under different traffic conditions. Plots are presented to aid in the interpretation of model inferences and make model output interpretable to non-statisticians. Graphs also aid in diagnosis of lack of fit and can help suggest model improvements.

In Section 3.2 we describe the dataset, Section 3.3 presents our model and Sections 3.4 and 3.5 give results. Finally, Section 3.6 is discussion.

3.2 Data Description

The I-405 closure event began midnight of Friday July 15 and remained in effect for 36 hours until 12 noon on Sunday July 17. UFP counts were measured by two SMPS machines between 10am and 8pm for three consecutive weekends: Friday, July 8 through Sunday, July 10th, the *pre-closure period*, Friday, July 15 through Sunday, July 17, the *closure period*, and Saturday July 23, the *post-closure period*. The machines were not functioning on Friday, July 22 nor Sunday, July 24. We refer to the pre- and post-closure periods collectively as *non-closure*.

Figure 3.1 shows a map of the study location taken from Quiros et al. (2013). SMPS measurements took place on Constitution Avenue, a perpendicular cross street passing beneath the I-405. One SMPS machine was located *upwind* of the 405, meaning opposite of wind direction so emissions from passing traffic would not be blown to this side. The other SMPS machine was located *downwind* of the 405, meaning traffic emissions would be blown to this side. During closure, northbound lanes of the 405 were completely closed and one lane of traffic was permitted southbound. The freeway was completely closed in both directions about a mile north of the measurement site. Sepulveda remained open during closure, but Constitution was closed to local access. The sampling time from 10am to 8pm corresponded to consistent sea breeze conditions and the time of peak weekend traffic flow. Downwind measurements were matched to upwind measurements so that they occurred between 45 and 103 seconds after upwind measurements. Ninety-five percent of downwind and upwind measurements occurred between 45 and 74 seconds apart, and the median time was 46 seconds, meaning the distribution of time between upwind and downwind measurements is skewed right. Both the upwind and downwind SMPS machines record measurements every two minutes.

Figure 3.2 plots upwind counts for select size bins from 10am to 8pm on the Saturday of the pre-closure period. As counts can change suddenly by or-

ders of magnitude, a log transformation allows for easier temporal modeling of counts (Whitby et al., 1991; Wraith et al., 2009, 2014; Fischer et al., 2014). We transform counts by taking the natural log plus ten, to further smooth counts near zero.

A log transformation exposes another issue with the data: inaccurate zeros over time recorded by the SMPS upwind machine. Figures 3.3(a)(b) show log upwind and downwind counts for the closure period Saturday. Log upwind counts for size bin 10 are at similar levels as downwind counts with the exception of frequent zeros over time for the upwind counts. Figures 3.3(c)(d) display UFP size distributions between 11-12pm on Saturday July 16. Curves are plotted progressively lighter as time increases and show the upwind zeros occur erroneously for UFP size bins smaller than 20 and to a certain extent for size bins smaller than 5 for the downwind counts. This is due to a malfunction in the machines and is not a measurement of actual counts. As such, zero counts for both the upwind and downwind SMPS machines are treated as missing values for this analysis.

Figures 3.4(a)(b)(c)(d) show plots of logged counts during the pre-closure Saturday with zeros removed. Sampling variance is larger for upwind counts for smaller size bins than for downwind counts, demonstrated by the jagged measurements of the UFP size distributions for smaller size bins. As smaller counts may be more likely to be given erroneous zero readings by the machine, this introduces the possibility of bias for model estimates. This will be discussed further in the results section. Downwind UFP counts are higher than upwind UFP counts for size bins between 5 and 40: downwind UFP size distributions typically look like upwind distributions with an additional local mode for particle size bins less than 40.

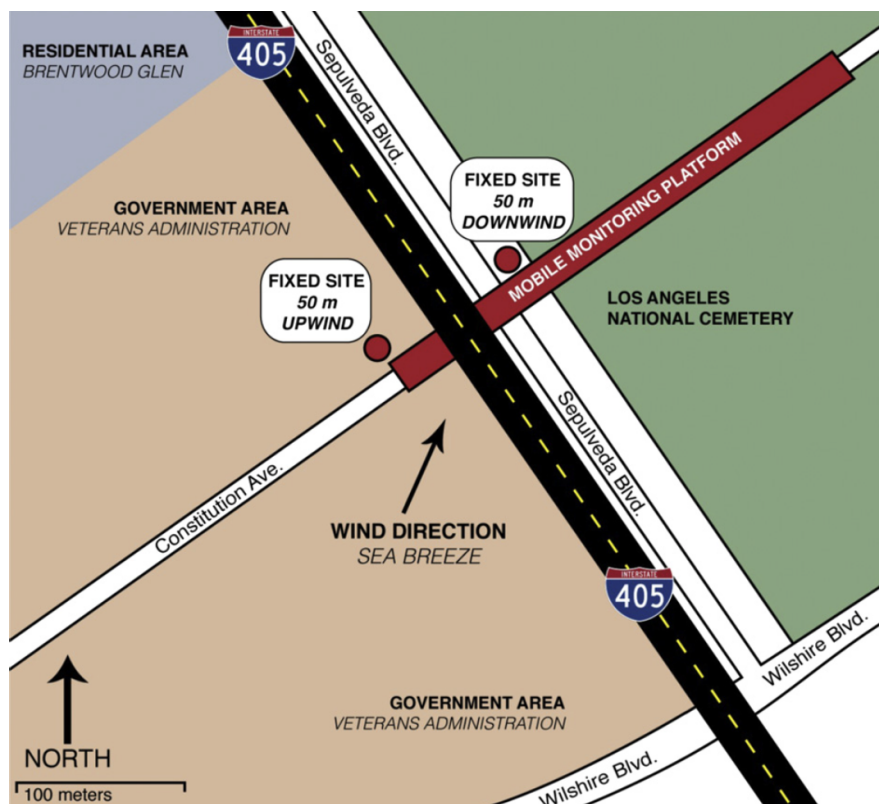


Figure 3.1: A map of the study location. SMPS measurements took place on Constitution Avenue, a perpendicular cross street passing beneath I-405. One SMPS machine was located *upwind* (west) of the 405, meaning opposite of wind direction so emissions from passing traffic would not be blown to this side. The other SMPS machine was located *downwind* (east) of the 405, meaning traffic emissions would be blown to this side. Figure from Quiros et al. (2013)

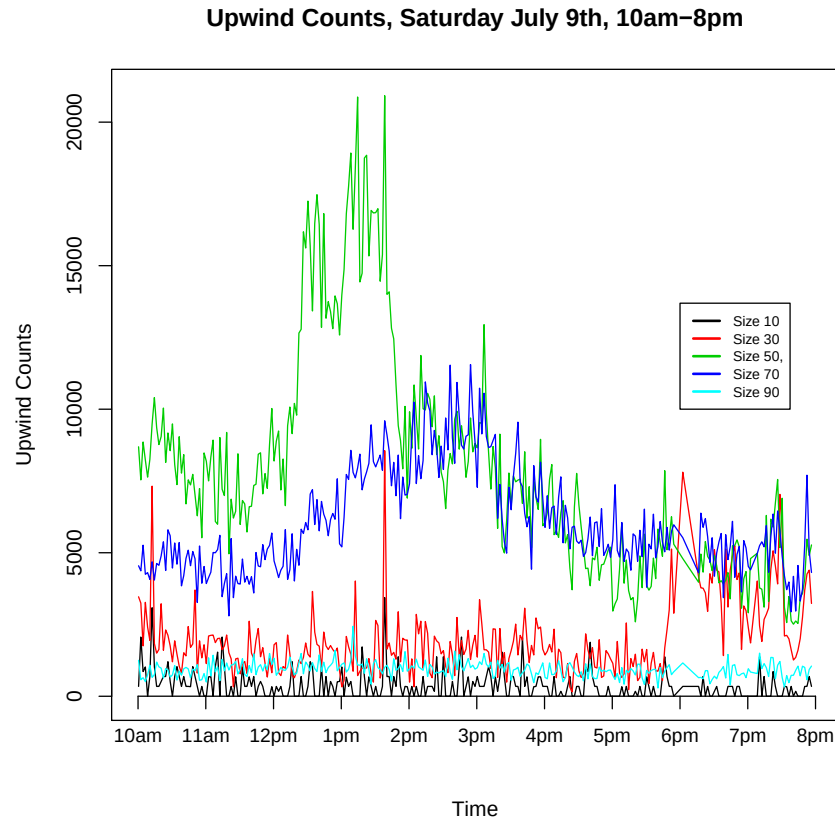


Figure 3.2: Upwind counts for select size bins from 10am to 8pm during the Saturday of the pre-closure period. As counts can change suddenly by orders of magnitude, a log transformation allows for easier modeling of counts (Whitby et al., 1991; Wraith et al., 2009, 2014; Fischer et al., 2014)

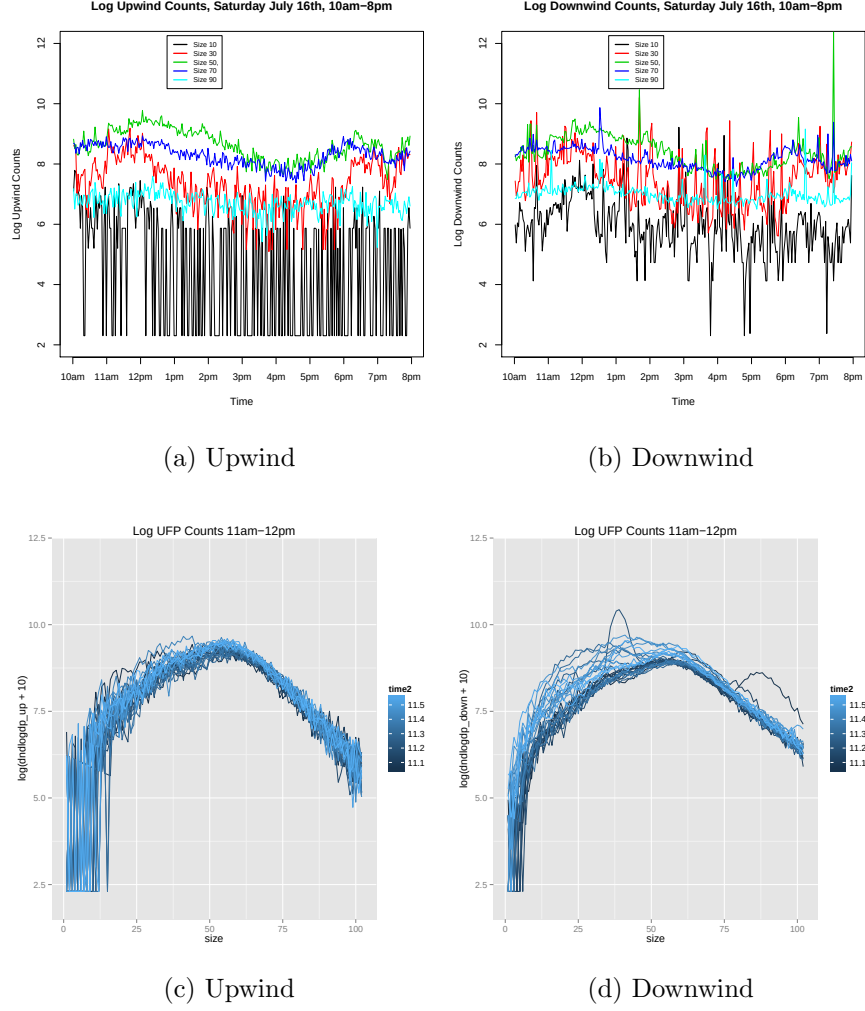


Figure 3.3: Figures (a)(b) show log upwind and downwind counts for the Saturday during the closure period. Log upwind counts for size bin 10 are at similar levels as downwind counts with the exception of frequent zeros over time for the upwind counts. Figures (c)(d) display UFP size distributions between 11-12pm on Saturday July 16. UFP distributions are plotted progressively lighter as time increases and show the upwind zeros occur excessively for UFP size bins smaller than 20 and to a certain extent for size bins smaller than 5 for the downwind counts. This is due to a malfunction in the machines, not a measurement of actual counts.

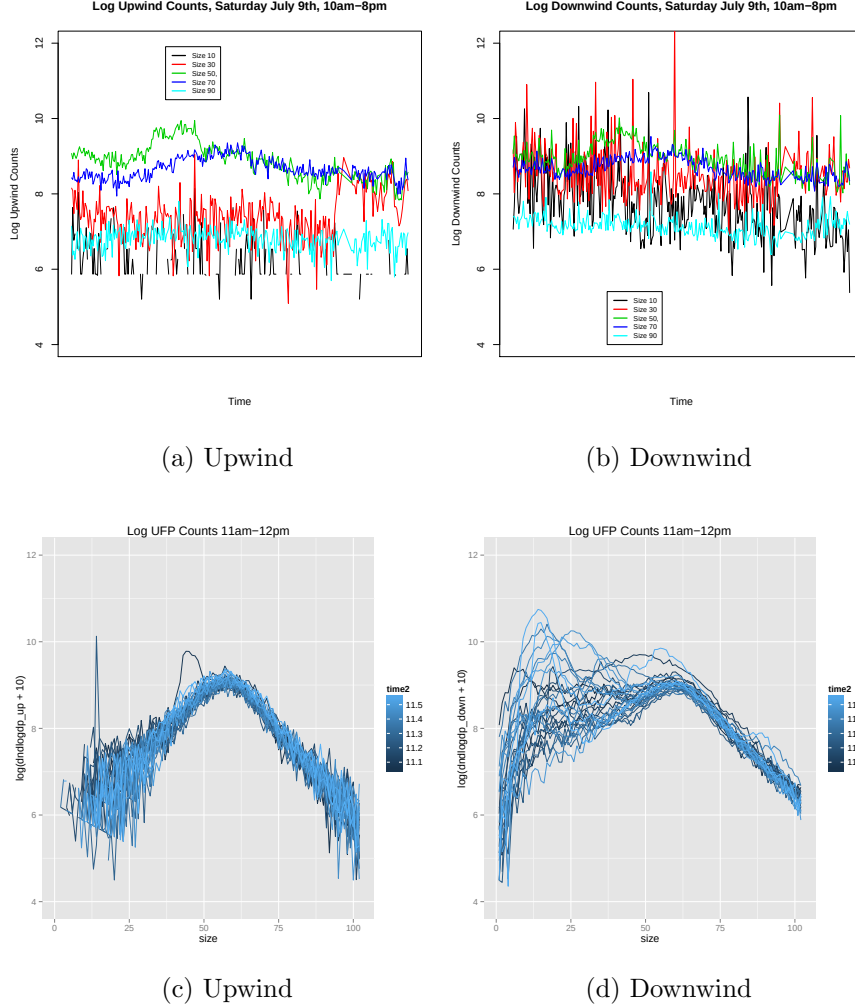


Figure 3.4: Figures(a)(b)(c)(d) show plots for logged counts during the pre-closure Saturday with zeros removed. Sampling variance is much larger for upwind counts for smaller size bins, demonstrated by the jagged measurements of the UFP size distributions for smaller size bins. As smaller counts may be more likely to be given erroneous zero readings by the machine, this introduces the possibility of bias for model estimates. This will be discussed further in the results section. Downwind UFP counts are higher than upwind UFP counts for size bins between 5 and 40: downwind UFP size distributions typically look like upwind distributions with an additional local mode for particle size bins less than 40.

3.3 A Time Series Semi-Parametric Model for UFP Size Distributions

Let s index particle size bin, with $s = 1, \dots, S$, for our data $S = 102$. Let i index pre-closure Friday, Saturday, Sunday, closure Friday, Saturday, Sunday, and post-closure Saturday, so that $i = 1, \dots, D$ and $D = 7$. Let z be an indicator of upwind and downwind location, where $z = 1$ corresponds to upwind and $z = 2$ corresponds to downwind. Time, t , is measured in seconds. Time has a day i and location z dependent range of $t_{\min,i,z}$ to $t_{\max,i,z}$, but the longest time frame possible is $t_{i,z,\min} = 0$ and $t_{i,z,\max} = 36,000$ seconds, corresponding to a time frame of 10am to 8pm. The set of all timepoints for which we have data on day i and location z is called T_{iz} . Because measurements are always 120 seconds apart, the elements of T_{iz} will depend on $t_{i,z,\min}$ for day i and location z . Outcome y_{izst} is the natural log of particle count plus 10 for day i , location z , size bin s at time t .

3.3.1 Model

Mean log upwind and downwind counts are expected to be non-stationary over time at levels dependent on day, location and size bin. We model y_{izst} as

$$y_{izst} = \rho_{izst} + u_{izst}, \quad (3.1)$$

where ρ_{izts} is the day i , location z and size bin s specific mean at time t . We let ρ_{izst} evolve smoothly over size bin and time following processes described shortly. We consider two models for residuals. The first is an independent residual model

$$u_{izst} \sim N(0, \sigma_{izs}^2) \quad (3.2)$$

where σ_{izs}^2 is the variance of u_{izst} . The second is an autoregressive process over time

$$u_{izst} = \theta_{iz} u_{i,z,s,t-120} + \epsilon_{izst}, \quad (3.3)$$

$$\epsilon_{izst} \sim N(0, \sigma_{izs}^2) \quad (3.4)$$

where lag one correlation is θ_{iz} , and σ_{izs}^2 is the variance of ϵ_{izst} given the previous residual $u_{i,z,s,t-120}$.

We want the upwind and downwind mean counts ρ_{izst} , to vary smoothly by size bin s and we model them as cubic B-spline functions of s . Let $\mathbf{B}(s)$ be a cubic $(K \times 1)$ B-spline basis over size bin s with $(K - 4)$ knots. Let $\boldsymbol{\alpha}_{izt}$ be the vector of coefficients of $\mathbf{B}(s)$ for the population mean at time t and location z . Then

$$\rho_{izst} = \boldsymbol{\alpha}_{izt}^T \mathbf{B}(s),$$

where $\boldsymbol{\alpha}_{izt} = (\alpha_{iztk})$. Upwind and downwind mean counts vary as a function of time and we model each coefficient α_{iztk} with a random walk over time independent of the other elements

$$\alpha_{iztk} \sim N(\alpha_{i,z,t-120,k}, \tau_{\alpha}^2). \quad (3.5)$$

The upwind and downwind error variances σ_{izs}^2 are a function of size bin and modeled with a separate quadratic $(L \times 1)$ B-spline basis over size bin, $\mathbf{B}_{\text{err}}(s)$, with $(L - 3)$ knots. Log variance is modeled

$$\log(\sigma_{izs}^2) = \boldsymbol{\eta}_{iz}^T \mathbf{B}_{\text{err}}(s) + w_{izs} \quad (3.6)$$

$$w_{izs} \sim N(0, \tau_{\eta}^2) \quad (3.7)$$

where the w_{izs} are small normal residual terms with known variance τ_{η}^2 added to $\boldsymbol{\eta}_{iz}^T \mathbf{B}_{\text{err}}(s)$ to enable efficient MCMC sampling (Crainiceanu et al., 2007; Baladandayuthapani et al., 2005; Hadfield, 2010). We set τ_{η}^2 to a small number to not add too much variation.

The full model is

$$y_{izst} = \boldsymbol{\alpha}_{izt}^T \mathbf{B}(\mathbf{s}) + u_{izst}, \quad (3.8)$$

together with Equations (3.2), (3.6), and (3.7) for the model with independent error and Equations (3.3), (3.4), (3.6), and (3.7) for the model with autoregressive error. We refer to the models that model upwind and downwind counts separately as the *separate* models. We refer to the model with autoregressive error as the *separate autoregressive error* model and the model with independent error structure as the *separate independent error* model.

3.3.1.1 Joint Model

The model above is fit to upwind and downwind data separately for each day i . The resulting smoothed estimates from the separate upwind and downwind models can be compared to determine differences in upwind and downwind estimates over time. An alternate way to compare upwind and downwind estimate is to estimate their differences in a single model. Here, upwind counts are modeled as in the separate independent error model in Equations (3.2), (3.6), (3.7) and (3.8). Then, defining the upwind fitted value at day i , size bin s , and time t as $\Upsilon_{ist} = \boldsymbol{\alpha}_{i1t}^T \mathbf{B}(\mathbf{s})$, where $z = 1$ for upwind counts, the downwind log counts are modeled as

$$y_{i2st} = \Upsilon_{ist} + \boldsymbol{\delta}_{it}^T \mathbf{B}(\mathbf{s}) + u_{i2st}, \quad (3.9)$$

$$u_{i2st} \sim N(0, \sigma_{i2s}^2) \quad (3.10)$$

where $\mathbf{B}(\mathbf{s})$ is the same cubic ($K \times 1$) B-spline basis used for $\boldsymbol{\alpha}_{i1t}$ in Equation (3.8) and $\boldsymbol{\delta}_{it}^T$ are the time varying coefficients of $\mathbf{B}(\mathbf{s})$ for the change from upwind to downwind UFP size distributions at time point t . This model implies changes from the upwind to downwind UFP size distributions are additive on the log scale, which is not a restriction since all counts are positive. The k th element δ_{itk} of

$\boldsymbol{\delta}_{it}^T$ follows a random walk over time as in Equation (3.5),

$$\delta_{itk} \sim N(\delta_{i,t-120,k}, \tau_\delta^2) \quad (3.11)$$

so that each element δ_{itk} of $\boldsymbol{\delta}_{it}^T$ follows a random walk process independent of the other elements. We model δ_{itk} and α_{itk} independently of each other.

The variance of u_{i2st} is σ_{i2s}^2 . This also varies smoothly over size bin and is modeled as in Equation (2.8)

$$\log(\sigma_{i2s}^2) = \boldsymbol{\eta}_{i2}^T \mathbf{B}_{\text{err}}(\mathbf{s}) + w_{i2s}, \quad (3.12)$$

$$w_{i2s} \sim N(0, \tau_\eta^2). \quad (3.13)$$

The log variances σ_{i1s}^2 and σ_{i2s}^2 are modeled independently. We only consider independent residuals for this model. We refer to this model as the *joint* model.

3.3.2 Priors

Proper priors are used on all parameters.

Both α_{itk} and δ_{itk} follow random walk processes with normal priors with means equal to $\alpha_{i,t-2,k}$ or $\delta_{i,t-1,k}$ $t - 1$ as described in Equations (3.5) and (3.11). Priors for time period $t_{i,z,\min}$, when there was no previous observation are,

$$\alpha_{izt_{\min}k} \sim N(0, \tau_\alpha^2) \quad (3.14)$$

$$\delta_{it_{\min}k} \sim N(0, \tau_\delta^2). \quad (3.15)$$

We give τ_α^2 and τ_δ^2 weakly informative priors

$$\tau_\alpha^2 \sim \text{gamma}(a_\alpha, b_\alpha) \quad (3.16)$$

$$\tau_\delta^2 \sim \text{gamma}(a_\delta, b_\delta). \quad (3.17)$$

The prior for the correlation parameter θ_{iz} is taken to be truncated normal as conjugacy improves computational speed and the likelihood for θ_{iz} is normal

$$\theta_{iz} \mid \mu_\theta, \sigma_\theta^2 \sim N(\mu_\theta, \sigma_\theta^2) \mathbb{I}[\theta \in (-1, .9)] \quad (3.18)$$

with the indicator function giving the lower -1 and upper $.9$ truncation points. The upper limit of 0.9 was chosen to avoid singularity in the likelihood (Palmer and Pettit, 1996).

The prior for the coefficients of the log variance is taken to be

$$\boldsymbol{\eta} \sim N_L(\log(0.4), g^2 \mathbf{I}_L) \quad (3.19)$$

where 0.4 was the mean residual error reported fitting UFP size distributions in Zhang et al. (2012), $\mathbf{I}_L$ is the $L \times L$ identity matrix, the hyperparameter g is set to 3.27 to put a 95 probability around residual variances between 0.2 and 0.7 . The prior is centered at a constant residual across size bins.

3.3.3 Computing Overview

Posterior estimates are obtained via Markov Chain Monte Carlo (MCMC) simulations (Hastings, 1970; Gelfand and Smith, 1990) using JAGS 3.3.0 (Plummer, 2003). MCMC convergence and mixing properties were assessed by inspecting autocorrelation and chain histories of individual parameters. Convergence appeared to be immediate for all parameters.

Missing observations were imputed as part of the MCMC algorithm, an automatic aspect of the JAGS model.

3.3.4 Estimates of Interest

Let $\mathbf{Y}_{iz}$ be the vector of all y_{izst} for day i and position z . Define the fitted log UFP counts for the separate model in Equation (3.8) as $\psi_{izst} = \boldsymbol{\alpha}_{izt}^T \mathbf{B}(\mathbf{s})$ and for the joint model defined in Equation (3.9) as $\psi_{izst} = \Upsilon_{ist} + \boldsymbol{\delta}_{it}^T \mathbf{B}(\mathbf{s}) * \mathbf{1}\{z = 2\}$.

Residuals can be calculated as

$$\hat{e}_{izst} = E[y_{izst} - \psi_{izst} \mid \mathbf{Y}_{iz}] \quad (3.20)$$

for both models (Chaloner and Brant, 1988; Chaloner, 1994). Autoregressive error

was not removed to better visualize lack of fit in the models that may be picked up by $\theta_{iz}y_{i,z,s,t-120}$.

For emission monitoring purposes, posterior predictions must be transformed back to their original scale. Conditional on day i and position z , predicted counts have marginal variance, $\lambda_{izs}^2 \equiv \text{var}(y_{izst} \mid \psi_{izst})$ that is the same for all times t . For the independent error structure we have simply $\lambda_{izs}^2 = \sigma_{izs}^2$. For the autoregressive error structure in Equation (3.3), the variance is

$$\lambda_{izs}^2 \equiv \text{var}(y_{izst} \mid \boldsymbol{\alpha}_{izt}^T) = \text{var}(\theta_{iz}u_{i,z,s,t-1} + \epsilon_{izst} \mid \boldsymbol{\alpha}_{i,z,t-1}) = \theta_{iz}^2\lambda_s^2 + \sigma_{izs}^2 \quad (3.21)$$

which solving for λ_{izs} gives

$$\lambda_{izs}^2 = \sigma_{izs}^2 / (1 - \theta_{iz}^2). \quad (3.22)$$

The back transformed mean of predicted counts for size bin s at time t is then

$$\mu_{izst} = \exp(\psi_{izst} + \lambda_{izs}^2/2). \quad (3.23)$$

Since σ_{izs}^2 is used in the calculation of λ_{izs} for both error structures, it is important that the variance captured by σ_{izs}^2 describes the actual movement of UFP particles in the air and not measurement error by the SMPS machine. To the extent that σ_{izs}^2 is measurement error, its inclusion in the calculation of the back transformed mean can misinform inferences. This will be discussed further in the results section. The back transformed median is simply

$$\xi_{izst} = \exp(\psi_{izst}). \quad (3.24)$$

We define the *ratio* UFP size distribution as $\gamma_{ist} = \exp(\boldsymbol{\delta}_{it}^T \mathbf{B}(\mathbf{s}))$, which is the ratio of the downwind to upwind counts.

It is of interest to understand how upwind and downwind UFP size distributions evolve over time, how the difference between the two changes over time, and how this varies across day. Because UFP size distributions near major roadways

are subject to short term fluctuations from minute to minute, looking at changes over the short 2-minute time intervals contained in the dataset may distort the gradual changes across hours that concern researchers. We therefore examine UFP size distributions averaged across time within one hour non-overlapping time periods.

To define this formally, we must first introduce some new notation. Let $h = 1, \dots, H$, $H = 10$, index hour, where $h = 1$ indexes to the time period between $t = 0$ and $t = 3,599$ (or between 10:00:00-10:59:59am). We define t_{izh} to be the initial time point t in hour h , position z , and day i , where by definition $t_{iz1} = t_{i,z,\min}$. Let $T_{izh} = \{t : t \in T_{iz}, t_{i,z,h-1} \leq t < t_{izh}\}$ and define $|T_{izh}|$ as the number of these elements. Then the hour mean for hour h , size s , position z , and day i takes the form

$$S_{izsh} = \frac{\sum_{t \in T_{izh}} \mu_{izst}}{|T_{izh}|}. \quad (3.25)$$

The hour mean can alternatively be defined using the median ξ_{izst}

$$S_{izsh} = \frac{\sum_{t \in T_{izh}} \xi_{izst}}{|T_{izh}|}. \quad (3.26)$$

We then define the *smoothing hour* distribution, $\mathbf{S}_{izh}$, as the $S \times 1$ size bin ordered vector of all S_{izsh} for day i , location z , and hour h . We define the *smoothing ratio hour* distribution $\mathbf{R}_{izh}$ as the $S \times 1$ size bin ordered vector of all

$$R_{izsh} = \frac{\sum_{t \in T_{izh}} \gamma_{ist}}{|T_{izh}|}. \quad (3.27)$$

Using smoothing hour $\mathbf{S}_{izh}$ and ratio hour $\mathbf{R}_{izh}$ distributions, we can compare upwind and downwind posterior estimates between one hour time intervals as well as make inferences about their evolution over time and across different days.

We define *additive hour difference* distributions $\mathbf{S}_{ih}$ as the difference between downwind, $z = 2$, and upwind, $z = 1$, smoothing hour UFP size distributions

S_{izh} during the same hour

$$S_{ih} = S_{i_2h} - S_{i_1h}. \quad (3.28)$$

The *difference of differences* S_h is the difference between two additive hour difference distributions S_{ih} for the same hour h but different day $i, i_0 \neq i_1$,

$$S_h = S_{i_1h} - S_{i_0h}. \quad (3.29)$$

The difference of differences is useful because we would like to compare additive hour differences between days i , helping researchers to understand the impact of freeway closure.

Define s_{izh} as the local modal size bin for smoothing hour distributions for day i , location z and hour h , where modes are only considered if counts to the left and to the right of s_{izh} are lower than s_{izh} . Define the modal particle count h_{izh} as the value of S_{izsh} at s_{izh} . The modal size bin s_{izh} and modal particle count h_{izh} provide researchers with a useful way to summarize the overall appearance of UFP size distributions by describing the location and heights of the size bins with the highest counts across hours over the course of the day.

3.4 Saturday Results

Because the separate and joint models produce near identical results for downwind and upwind estimates, we examine model fit as well as upwind and downwind inferences in this section using only the separate model. We then examine ratios between upwind and downwind counts using the joint model, and examine the difference of differences using the separate model. Saturdays are the focus of inference in this section both because there is data for pre, during, and post-closure periods, and because the closure Saturday was the only day the freeway was completely closed for the duration of measurements. Fridays and Sundays are examined in the subsequent section, however the freeway was never closed on the closure Friday and the freeway reopened at 12 noon on the closure Sunday.

3.4.1 Model Evaluation

We first evaluated whether the autoregressive residuals were a useful addition to our separate model through a combination of DIC (Spiegelhalter et al., 2002) and examination of posterior estimates for the AR parameter θ_{iz} . We considered both upwind and downwind locations on post-closure Saturday, July 23th.

Posterior median autocorrelation θ_{iz} estimates and 95 percent credible intervals for the autoregressive separate models applied to upwind and downwind data are presented in Table 3.1.

Table 3.1: Carmageddon Autocorrelation θ_{iz} Estimates

Model	Median	95% Credible Interval
Upwind	0.0029	(-0.0104,0.0163)
Downwind	0.0340	(0.0215,0.0466)

Posterior credible intervals for θ_{iz} overlap with zero for the separate model

fit to upwind data and are practically zero for the model fit to downwind data. DIC and components for the autoregressive and independent separate models fit to upwind and downwind data are presented in Table 3.2. Smaller DIC scores indicate better model fit.

Table 3.2: DIC Calculations for Carmaggedon Models

Model	DIC	DIC bar	pD
Upwind, Independent	7,497.9	4,874.5	2,623.4
Upwind, Autoregressive	7,470.4	4,839.7	2,630.8
Downwind, Independent	-42,810.8	-46,275.7	3,465.0
Downwind, Autoregressive	-42,830.6	-46,300.8	3,470.1

The DIC is much larger for the model fit to the upwind versus downwind data, regardless of error structure. Though DICs for models fit to different dependent variables cannot be compared directly, we note this is largely a consequence of the upwind SMPS machine having larger sampling variance than the downwind machine for smaller size bins, which removed the functional form typically seen in the UFP size distribution at smaller size bins.

DIC is lower for the autoregressive models than the independent models fit to the same data: it is about 27 lower for the model fit to the upwind data and about 20 lower for the model fit to the downwind data. It is difficult to determine what constitutes an important difference in DIC, and there is no one rule. One common threshold is a difference of 10 or more, however other factors must also be taken into consideration during model selection. Though these DIC differences surpass the threshold of 10, estimates for θ_{iz} were practically zero for both upwind and downwind models. The AR component θ_{iz} is unlikely to add valuable information to model inference and unnecessarily complicates model structure. We thus use the independent separate model going forward. All inferences are based on the

independent separate model unless otherwise specified.

As an example of model fit, Figure 3.5 shows fitted, actual and residual upwind and downwind log counts for post-closure Saturday, July 23th from 12-1pm. Curves are plotted in progressively lighter colors over the course of the hour. Plots show the separate model does a good job of fitting the actual data while smoothing counts over size bin. Increased sampling variance for the smaller upwind size bins causes higher residual variance for the upwind model for those size bins.

Figure 3.6 shows upwind and downwind residuals, $\hat{e}_{izst}$, over size bin for pre-closure, closure, and post-closure Saturdays. Figure 3.7 shows upwind and downwind residuals for these Saturdays over time. Residual variance for model fit to both upwind and downwind data is roughly constant over time on all days, however variance is not constant over size bin. Residual variance is larger for the model fit to upwind data than fit to downwind data, particularly for the smaller size bins. There are fewer upwind residuals plotted for smaller sizes because of the higher number of upwind missing observations.

Figure 3.8 plots posterior median σ_{zs}^2 estimates for pre-closure, closure and post-closure Saturdays. Dotted lines show 95 percent credible intervals. Downwind residual variance estimates are similar by size across day while upwind residual variance estimates differ to a larger extent. Upwind residual variance estimates are consistently larger than downwind residual variance estimates across all days for sizes smaller than 40nm.

3.4.2 Smoothing Hour Distributions

It was noted in Section 3.3.4 that to the extent that σ_{izs}^2 is measurement error, its inclusion in the calculation of the back transformed mean μ_{izst} as defined in Equation (3.23) can misinform inferences. To demonstrate the typical behavior of residuals from this analysis, Figures 3.9(a) and (b) plot upwind and downwind

residuals across size bin for Saturday, July 23 from 12-1pm, this time connecting residuals for the same time point with a line across size bin. Figures (c) and (d) are similar, but with only 2 timepoints plotted to allow better examination of the residual pattern. Jagged lines around zero indicate deviations from a smooth mean over size, and these are much larger for the upwind machine with larger sampling variance. These residuals are not a result of actual variation in particle counts in the air, but rather mostly machine measurement error. Calculating smoothing hour distributions $\mathbf{S}_{izt}$ using the back transformed mean μ_{izst} will lead to inaccurate inferences. Thus, we calculate $\mathbf{S}_{izt}$ using medians ξ_{zst} as defined in Equation (3.26).

Figure 3.10 plots smoothing hour distributions over the course of the day for pre-closure (Figures (a)(b)), closure (Figures (c)(d)), and post-closure (Figures (e)(f)) Saturdays for both upwind and downwind positions. Plots suggest that for all Saturdays, both upwind and downwind UFP size distributions tend to begin the day at lower levels across all size bins, reach their highest levels at midday, then decrease again into the evening. For non-closure Saturdays, upwind UFP size distributions tend to have one mode, while downwind counts tend to have 2 local modes. Both upwind and downwind positions tend to have one mode during the closure Saturday. Modes are explored in more detail shortly.

Figure 3.11 compares upwind and downwind smoothing hour distributions for 11am-12pm, 2pm-3pm, and 5pm-6pm for pre-closure (Figures (a)(b)(c)), closure (Figures(d)(e)(f)), and post-closure (Figures (g)(h)(i)) Saturdays. Ninety-five percent credible intervals are plotted with dotted lines. However, intervals are very narrow due to the high amount of data used to calculate smoothing hour distributions. Non-closure downwind smoothing hour distributions at all three selected hours tend to have an additional mode at smaller size bins reflecting contributions from freeway traffic. Closure upwind and downwind smoothing hour distributions look very similar, as little traffic was allowed in the area on that Saturday and

the freeway was completely closed. For particle size bins smaller than 5 (and on the closure Saturday smaller than 10), downwind estimates are often lower than upwind estimates. This could be due to bias in upwind estimates since so many observations were discarded due to machine reporting counts of zero.

3.4.3 Difference Estimates

Figure 3.12 plots the ratio hour distributions as defined by Equation (3.27) over time for pre-closure, closure, and post-closure Saturdays. These plots show the ratio between downwind and upwind smoothing hour distributions at different hours for all days, highlighting when downwind counts are higher than upwind counts. For pre and post-closure Saturdays, the ratios of downwind to upwind counts are very high for particles between 10 and 20nm, but that these ratios tend to decrease by the end of each day. These high ratios are not seen during the closure Saturday: the ratio was less than 5 during the closure Saturday compared to up to 20 during the non-closure Saturdays.

We next examine the difference of differences across days as defined in Equation (3.29) using the independent separate models for inference. Figure 3.13 plots the difference of differences for select hours comparing pre- and post-closure Saturdays to the closure period. Using the language of Equation (3.29), the closure weekend is always day i_0 . These plots show that in general the downwind–upwind differences were always higher during non-closure weekend than during the closure weekends. They also show that generally counts were higher for the post-closure weekend compared to the pre-closure weekend. The largest differences are seen in particles smaller than 40nm, reflecting contributions from vehicle emissions to downwind UFP size distributions and possibly meteorological conditions, which can vary greatly by day.

3.4.4 Local Modes

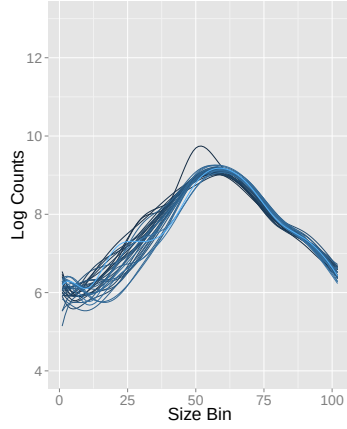
We now look at the temporal aspect of local modes. Following Quiros et al. (2013), we refer to local modes occurring below size bin 50 as *nucleation* modes, and local modes occurring at size bins greater than 50 as *larger UFP* modes. Nucleation modes are typically more variable and are caused by local emission sources, while larger UFP modes are more constant as they typically reflect background pollution levels.

Figures 3.14(a)(c)(e) plot the size bin locations of nucleation modes (dotted lines with triangles) and larger UFP models (solid lines with circles) over time for upwind (green) and downwind (red) locations on the three Saturdays. Figures (b)(d)(f) plot the heights of these of nucleation modes and larger UFP modes over time for the three Saturdays. Closure and post-closure Saturdays had a maximum of two modes for both the upwind and downwind locations, while on the pre-closure Saturday for the downwind location a third mode appeared (a second nucleation mode) during the hours of 10-11am and 5-6pm.

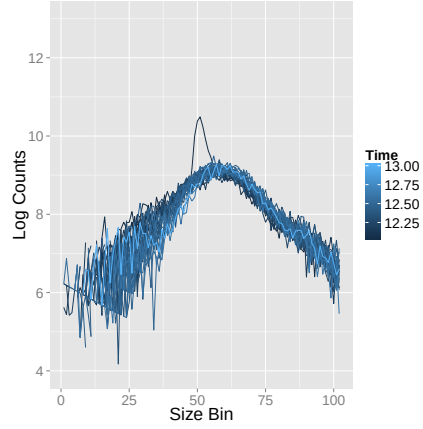
The larger UFP mode for all three Saturdays tends to be located around size bin 60 for both the upwind and downwind locations. Though the height of this mode differs between Saturdays, all show the trend of an increasing or steady larger UFP mode height until early afternoon, then a decreasing height into the evening with some deviation at the end of the day for the closure Saturday at the downwind location. The closure does not seem affect the height of the larger UFP mode as the post-closure Saturday has lower heights over time than the closure Saturday.

At many time points there is not a nucleation mode for the upwind location. For the downwind location there is much more variation in the nucleation mode; it appears and reappears at different hours on the pre-closure Saturday, while it remains through the course of the day on the post-closure Saturday but ranges

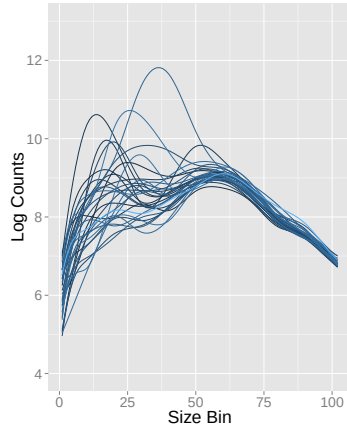
between size bins 20 and 40. Its height is also extremely variable across hours on the pre and post-closure Saturdays. Though a downwind nucleation mode does appear midday during the closure Saturday, its height is greatly reduced compared with the pre and post-closure Saturdays indicating the closure did have a substantial affect on this mode.



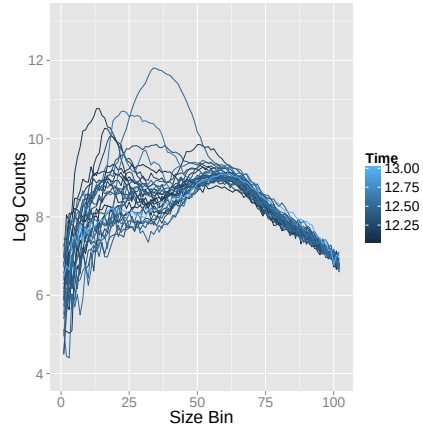
(a) Pre-closure, upwind fitted



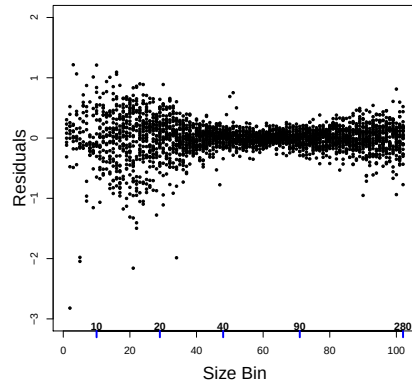
(b) Pre-closure, upwind actual



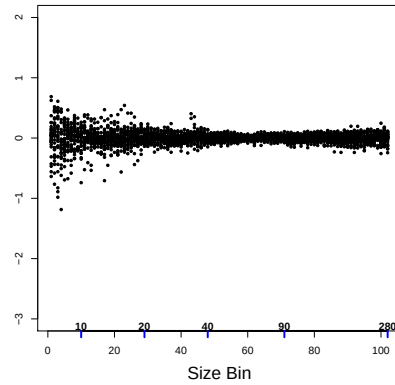
(c) Pre-closure, downwind fitted



(d) Pre-closure, downwind actual

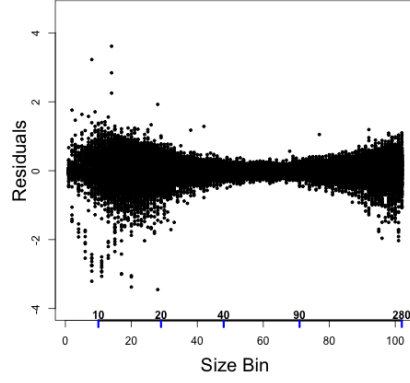


(e) Pre-closure, upwind residuals

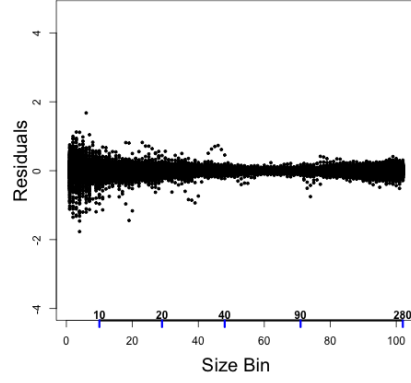


(f) Pre-closure, downwind residuals

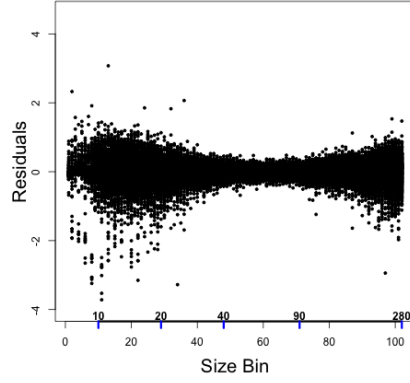
Figure 3.5: Fitted (a)(c) and actual (b)(d) log counts and residuals (e)(f) for post-closure Saturday, July 23th from 12-1pm for both upwind and downwind locations. Curves are plotted progressively lighter over the course of the hour.



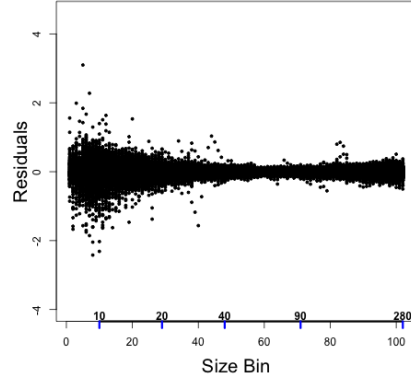
(a) Saturday pre-closure, upwind



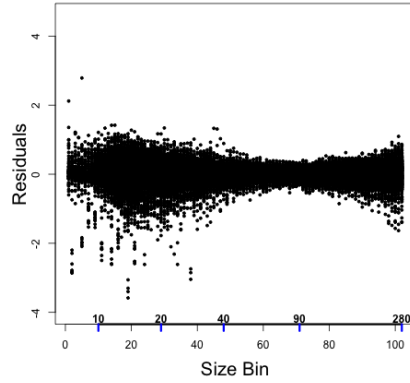
(b) Saturday pre-closure, downwind



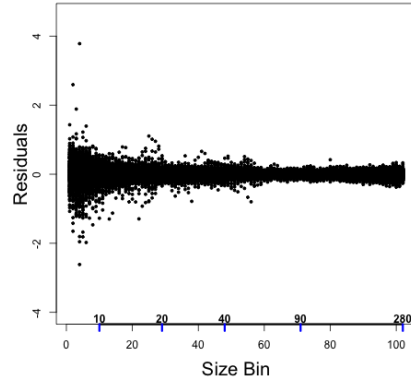
(c) Saturday closure, upwind



(d) Saturday closure, downwind

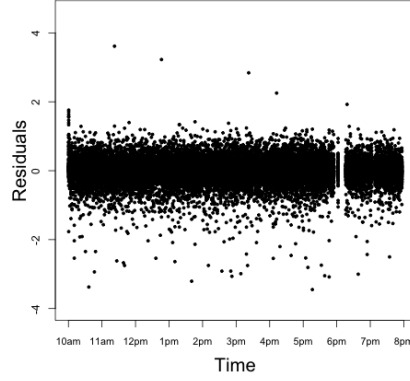


(e) Saturday post-closure, upwind

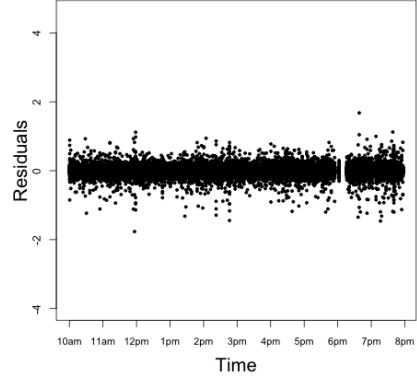


(f) Saturday post-closure, downwind

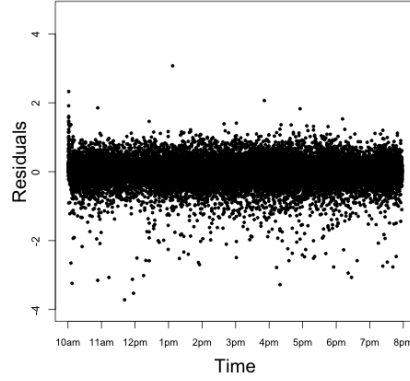
Figure 3.6: Upwind and downwind residuals, $\hat{e}_{izst}$, over size bin for pre-closure (a)(b), closure (c)(d), and post-closure (e)(f) Saturdays. Residual variance is not constant over size bin and is larger for the upwind data than for the downwind data.



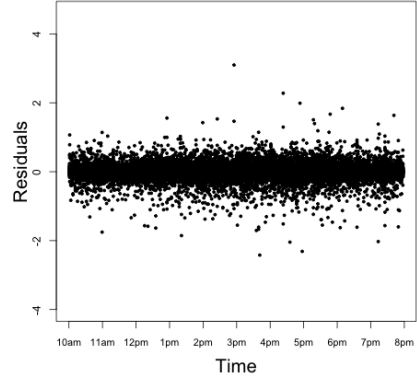
(a) Pre-closure, upwind



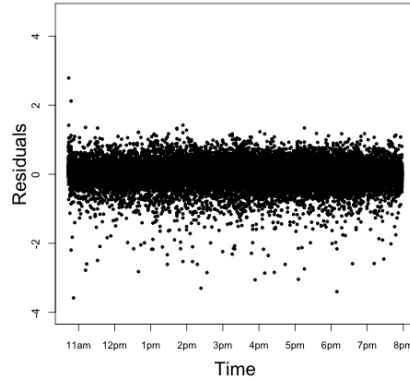
(b) Pre-closure, downwind



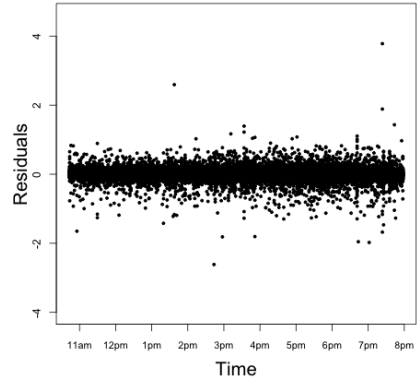
(c) Closure, upwind



(d) Closure, downwind

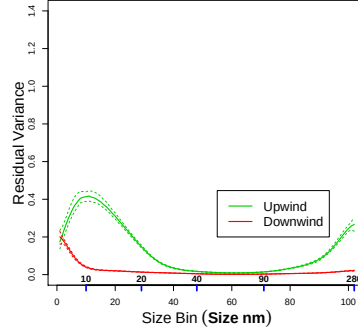


(e) Post-closure, upwind

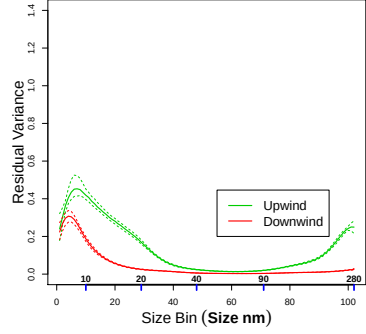


(f) Post-closure, downwind

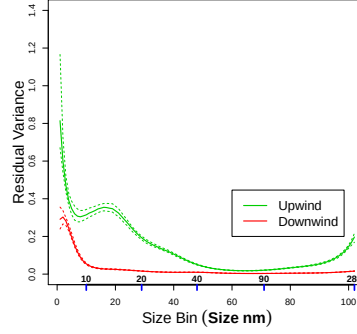
Figure 3.7: Upwind and downwind residuals, $\hat{e}_{izst}$, over time for pre-closure (a)(b), closure (c)(d), and post-closure (e)(f) Saturdays. Residual variance is roughly constant over time on all Saturdays. Residual variance is larger for model fit to upwind data than to downwind data.



(a) Pre-closure

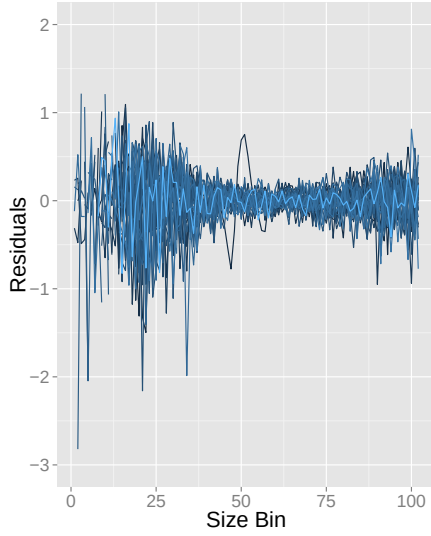


(b) Closure

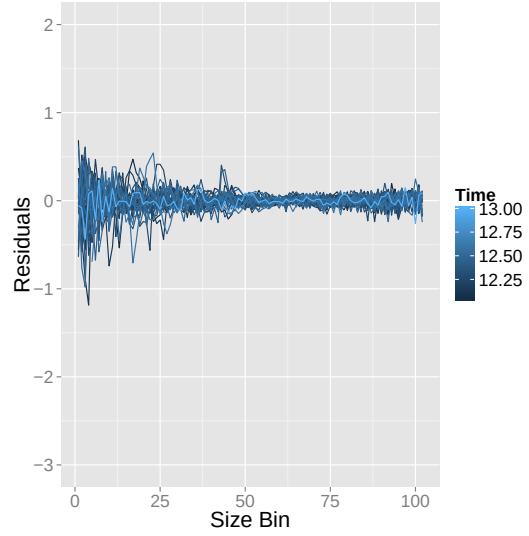


(c) Post-closure

Figure 3.8: Posterior median σ_{zs}^2 estimates for pre-closure (a), closure (b), and post-closure (c) Saturdays. Dotted lines show 95 percent credible intervals. Downwind residual variance estimates are similar by size across day while upwind residual variance estimates differ to a larger extent. Upwind residual variance estimates are consistently larger than downwind residual variance estimates across all days for sizes smaller than 40nm.



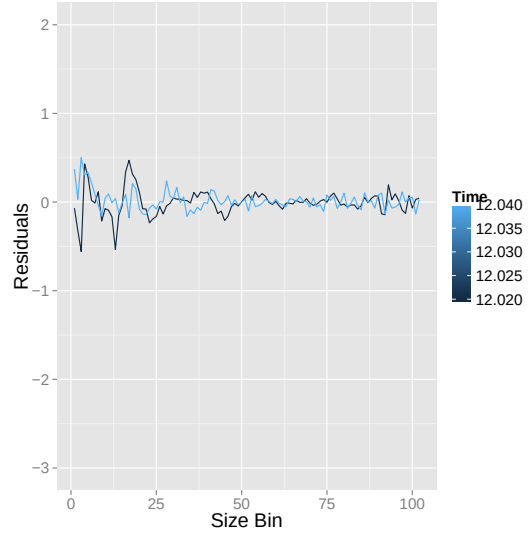
(a) Upwind



(b) Downwind

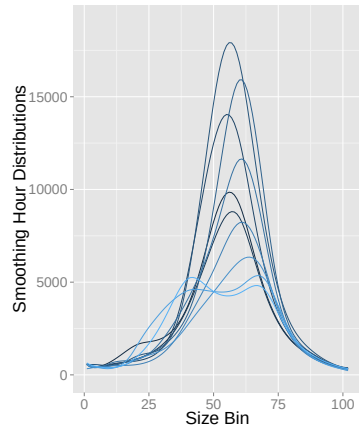


(c) Upwind

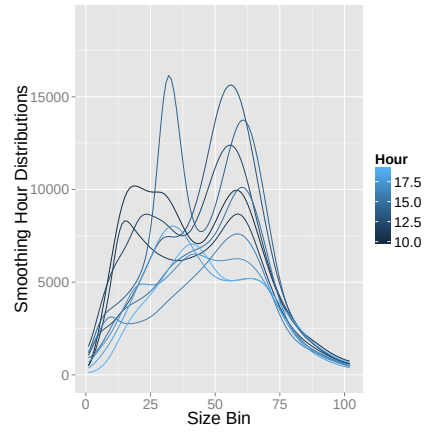


(d) Downwind

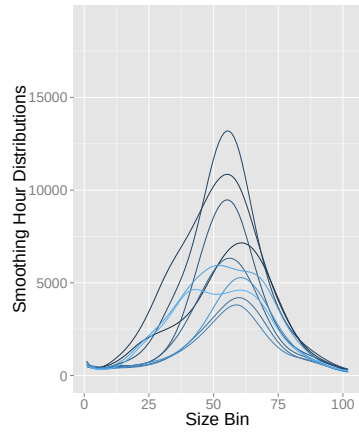
Figure 3.9: Figures (a) and (b) plot upwind and downwind residuals, $\hat{e}_{izst}$, across size bin for Saturday, July 23 from 12-1pm, this time connecting residuals for the same time point with a line across size. Figures (c) and (d) are similar, but with only 2 timepoints plotted to allow better examination of the residual pattern. Jagged lines around zero indicate deviations from a smooth mean over size, and these are much larger for the upwind machine with larger sampling variance.



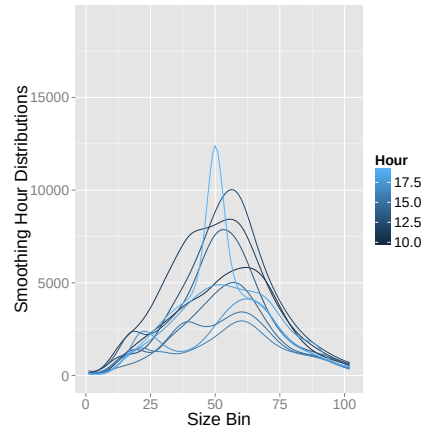
(a) Pre-closure, upwind



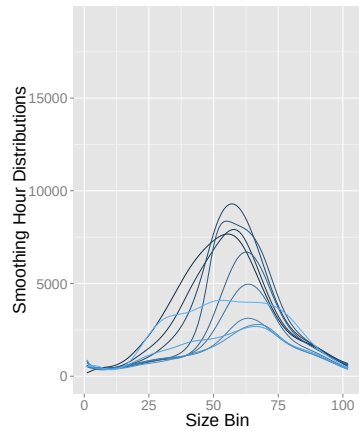
(b) Pre-closure, downwind



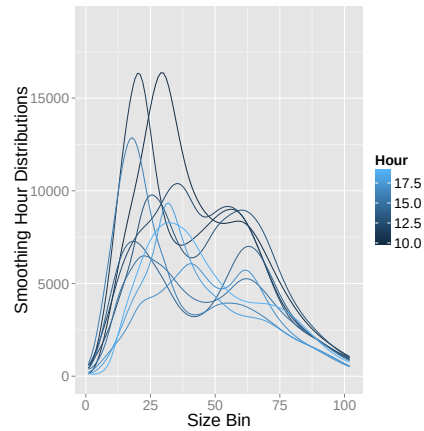
(c) Closure, upwind



(d) Closure, downwind



(e) Post-closure, upwind



(f) Post-closure, downwind

Figure 3.10: Upwind and downwind smoothing hour distributions for pre-closure (a)(b), closure (c)(d), and post-closure (e)(f) Saturdays. Non-closure Saturdays tend to have one mode, while downwind counts tend to have 2 local modes.

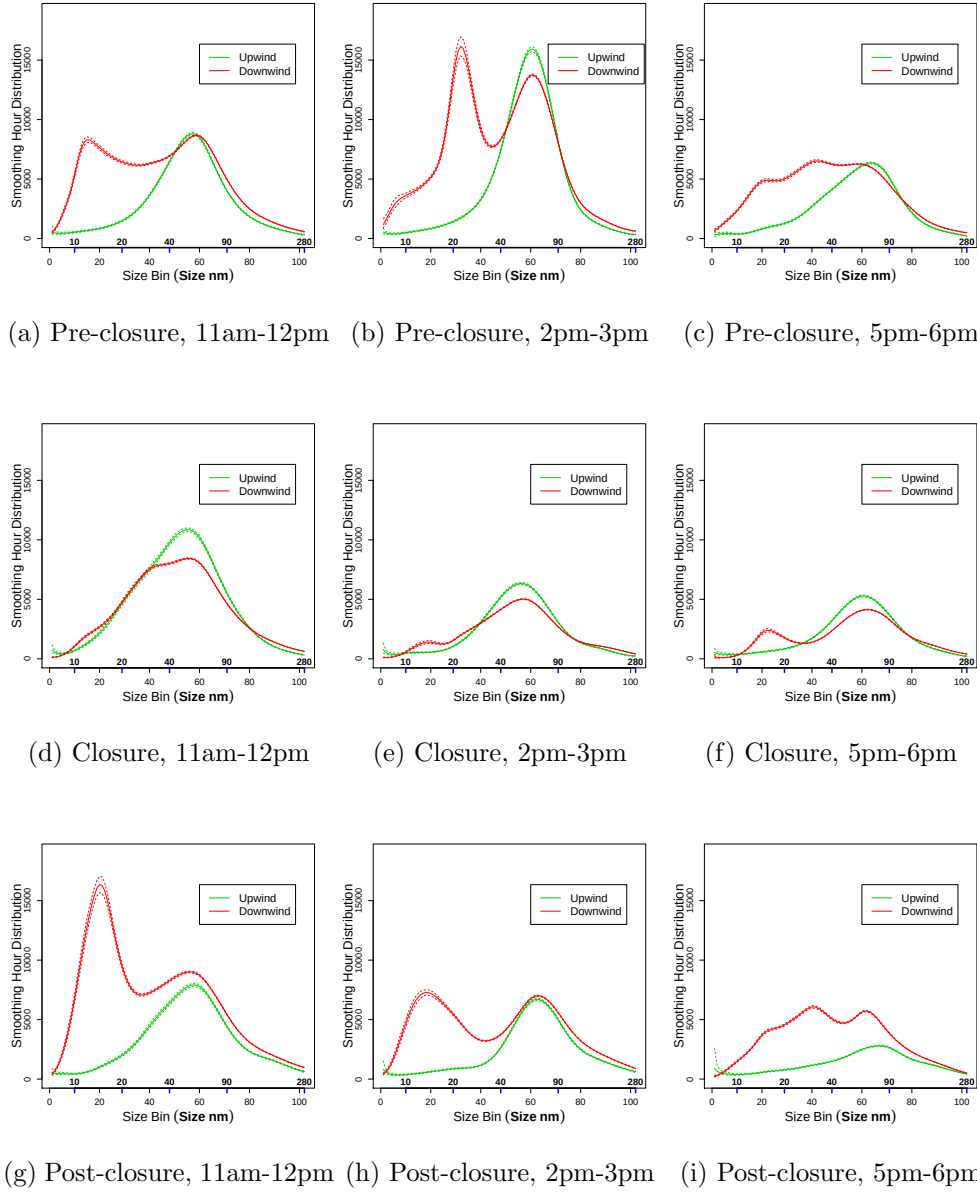
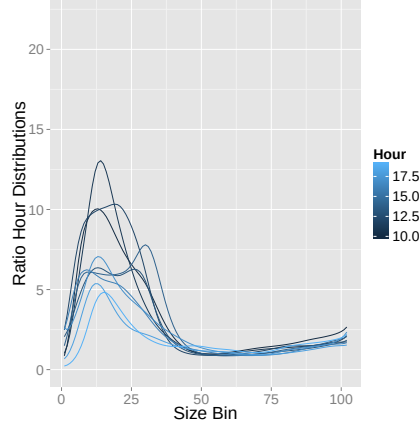
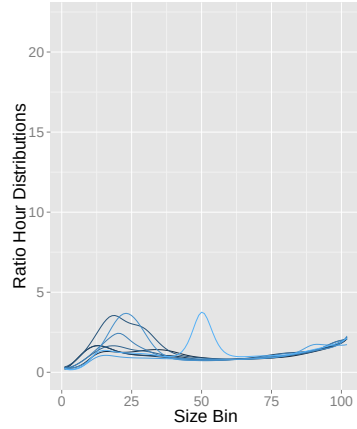


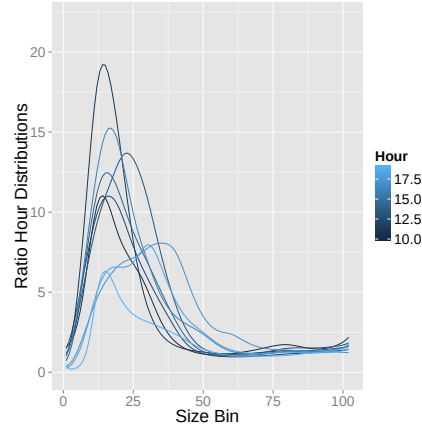
Figure 3.11: Upwind and downwind smoothing hour distributions for 11am-12pm (a)(d)(g), 2pm-3pm (b)(e)(h), and 5pm-6pm (c)(f)(i) for all Saturdays. Ninety-five percent credible intervals plotted with dotted lines. Non-closure downwind smoothing hour distributions at all three selected hours tend to have an additional mode at smaller size bins reflecting contributions from freeway traffic. Closure upwind and downwind smoothing hour distributions look very similar, as little traffic was allowed in the area on that Saturday and the freeway was completely closed. For particle size bins smaller than 5 (and on the closure Saturday smaller than 10), downwind estimates are often lower than upwind estimates.



(a) Pre-closure



(b) Closure



(c) Post-closure

Figure 3.12: Ratio hour distributions as defined by Equation (3.27) over time for pre-closure (a), closure (b), and post-closure (c) Saturdays. These plots show the ratio between the downwind and upwind smoothing hour distributions at different hours, highlighting when downwind counts are higher than upwind counts. For pre and post-closure Saturdays, the ratio of downwind to upwind counts is very high for particles between 10 and 20nm, but that this tends to decrease by the end of each day. The ratios are much lower during the closure Saturday: the ratio was less than 5 during the closure Saturday compared to up to 20 during the non-closure Saturdays.

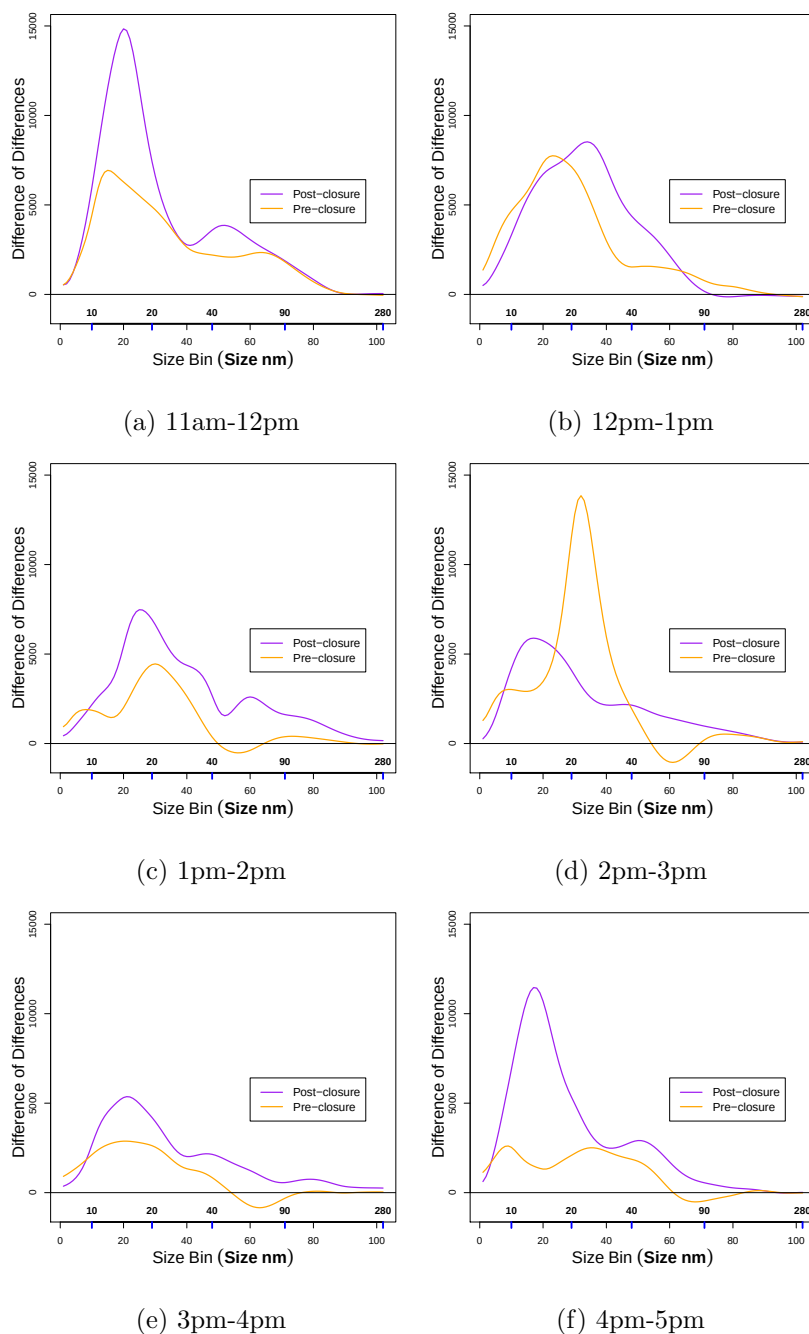
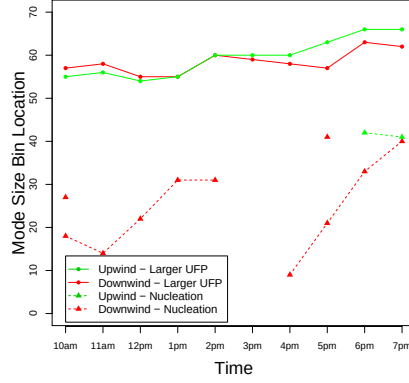
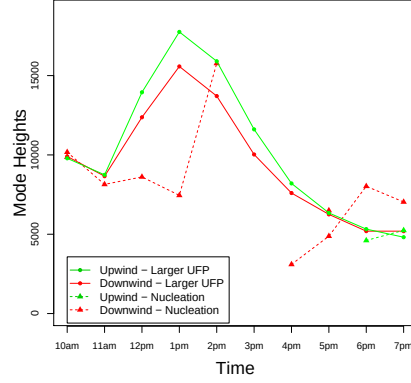


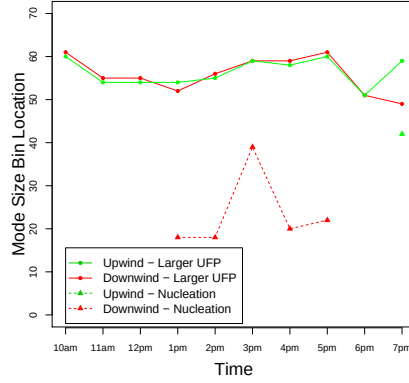
Figure 3.13: Difference of differences for select hours comparing pre- and post-closure Saturdays to the closure period. These plots show that in general the downwind–upwind differences were always higher during non-closure weekend than during the closure weekends. They also show that generally counts were higher for the post-closure weekend compared to the pre-closure weekend. The largest differences are seen in particles smaller than 40nm, reflecting contributions from vehicle emissions to downwind UFP size distributions and possibly meteorological conditions, which can vary greatly by day.



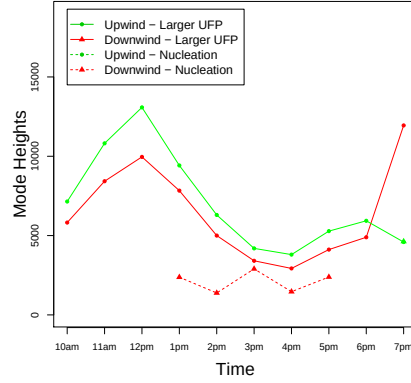
(a) Pre-closure, mode location



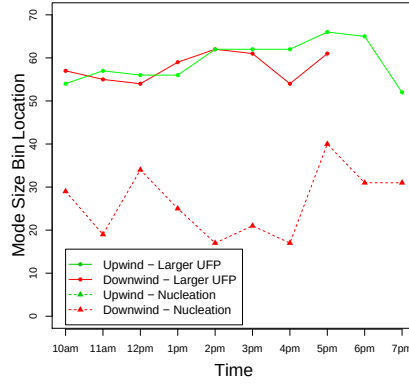
(b) Pre-closure, mode height



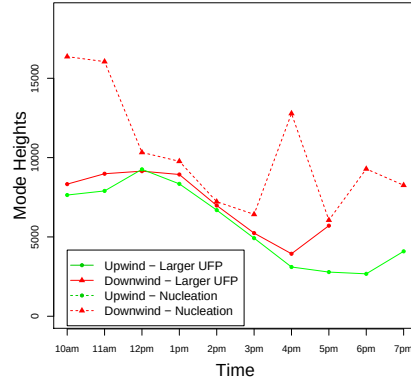
(c) Closure, mode location



(d) Closure, mode height



(e) Post-closure, mode location



(f) Post-closure, mode height

Figure 3.14: Figures (a)(c)(e) plot the size bin locations while Figures (b)(d)(f) plot the heights of nucleation modes (dotted lines with triangles) and larger UFP modes (solid lines with circles) over time for upwind (green) and downwind (red) locations the three Saturdays. Closure and post-closure Saturdays had a maximum of two modes for both the upwind and downwind locations, while on the pre-closure Saturday for the downwind location a third mode appeared (a second nucleation mode) during certain hours.

3.5 Friday and Sunday Results

Results for Fridays and Sundays are examined in this section. It is important to note that the freeway never closed on the closure Friday and the freeway reopened at 12 noon on the closure Sunday. Results here can be used to understand how counts varied by size bin on various Fridays and Sundays, rather than being a comparison of closure and non-closure days. Only pre-closure and closure data are available on these days.

3.5.1 Model Evaluation

Based on the model evaluation using DIC and AR estimates θ_{iz} that was done in Section 3.4.1, all inferences for Fridays and Saturdays are based on the separate independent model unless otherwise noted. Figure 3.15 shows residuals, $\hat{e}_{izst}$, over size bin for pre-closure and closure Fridays (Figures(a)-(d)) and Sundays (Figures (e)-(h)) while Figure 3.16 shows upwind and downwind residuals for these Fridays and Saturdays over time. Residuals demonstrate similiar behavior as observed on Saturdays: residual variance is roughly constant over time, but larger for smaller sizes especially for the upwind data.

Figure 3.17 plots posterior median σ_{zs}^2 estimates for pre-closure and closure Fridays (row 1) and Sundays (row 2). Dotted lines show 95 percent credible intervals. As with Saturdays, downwind residual variance estimates are similiar by size across day while upwind residual variance estimates differ to a larger extent. Upwind residual variance estimates are consistently larger than downwind residual variance estimates across all days for sizes smaller than 40nm.

3.5.2 Smoothing Hour Distributions

For the reasons discussed in Section 3.4.2, we calculate smoothing hour distributions $\mathbf{S}_{izt}$ using medians ξ_{zst} as defined in Equation (3.26). Figure 3.18 plots

smoothing hour distributions over the course of the day for pre-closure (Figures (a)(b)) and closure (Figures (c)(d)) Fridays. Two downwind and one upwind smoothing distributions are not plotted due to extreme outliers caused by vehicles idling right by the SMPS machines, which led to extremely high estimates that cloud inference for that hour. For all Fridays, both upwind and downwind UFP size distributions tend to decrease into the evening, but they do not begin at lower levels as seen on Saturdays. Pre-closure Friday, and to some extent closure Friday, show much more variation throughout the day for both upwind and downwind smoothing hour distributions compared to the other weekend days. Two or more modes are seen during many hours for closure downwind Friday as the freeway was not completely closed.

Figure 3.19 plots smoothing hour distributions over the course of the day for pre-closure (Figures (a)(b)) and closure (Figures (c)(d)) Sundays. There are no upwind estimates for the closure Sunday for size bins less than 10 because almost all observations are missing. Pre-closure Sunday also shows a pattern of both upwind and downwind UFP size distributions decreasing into the evening, though during the closure Sunday this is not the case. Across all sizes UFP counts are at their lowest on Sundays, however two modes are seen during many hours for closure downwind Sunday as the freeway was opened at 12pm.

Figure 3.20 and 3.21 compare upwind and downwind smoothing hour distributions for 11am-12pm, 2pm-3pm, and 5pm-6pm for pre-closure (Figures (a)(b)(c)) and closure (Figures (d)(e)(f)) Fridays and Saturdays, respectively. For all selected hours except for the closure Sunday from 11-12pm (Figure 3.21(d)) when the freeway was still completely closed, there is an additional mode at the downwind location at smaller size bins. The downwind pre-closure Friday has 3 modes during the earlier selected hours.

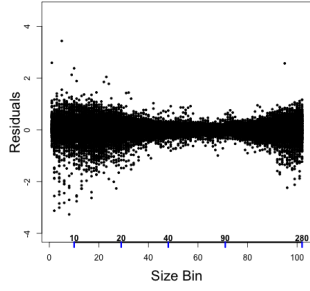
3.5.3 Difference Estimates

Figure 3.22 plots the ratio hour distributions as defined by Equation (3.27) over time for pre-closure and closure Fridays (3.22(a)(b)) and Sundays (3.22(c)(d)). The ratio hour distributions on pre-closure and closure Fridays are similar, indicating the upcoming closure did not affect downwind-upwind UFP size distribution ratio, but the ratio hour distributions are higher for the pre-closure Sunday compared to the closure Sundays. As seen on Saturdays, the ratio hour distributions also tend to decrease by the end of the day on all Fridays and Sundays.

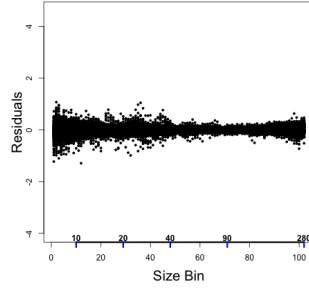
3.5.4 Local Modes

Figures 3.23 plot local modes for the upwind and downwind locations on the pre-closure and closure Fridays. The erratic behavior of both the upwind and downwind UFP size distributions on the pre-closure Friday is reflected in Figures 3.23(a) and (b). Both nucleation and larger UFP modes disappear and reappear from hour to hour at different size bins, and their height changes drastically across hour. Toward the end of the day, the height of each decreases. The closure Friday tends to have more constant nucleation and larger UFP mode locations, which decrease over the course of the day, however these are still more variable than those seen on Saturdays. The nucleation mode does not disappear on the closure Friday, and has levels similar to the pre-closure height until the end of the day.

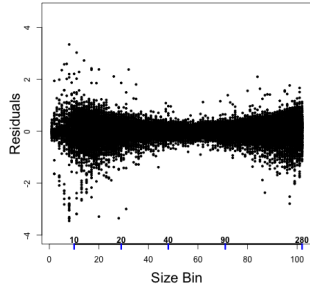
Figures 3.24 plot local modes for the upwind and downwind locations on the pre-closure and closure Sundays. Sunday larger UFP modes tend to resemble those seen on Saturdays, located at around size bin 60 and decreasing slightly in height throughout the day. However, the height of the larger UFP mode increases slightly by the end of the day closure Sunday, possibly an effect of the reopening of the freeway. Nucleation modes are present on both pre-closure and closure Sundays, but the closure Sunday nucleation mode is lower in height.



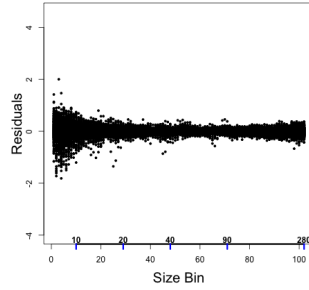
(a) Pre-Closure, upwind



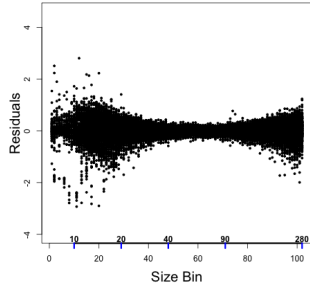
(b) Pre-Closure, downwind



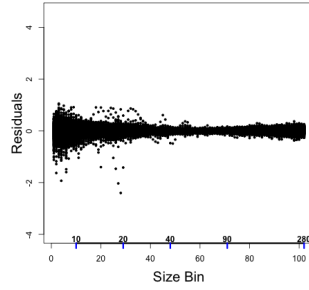
(c) Closure, upwind



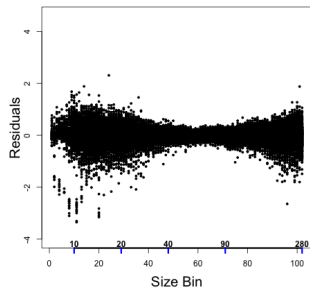
(d) Closure, downwind



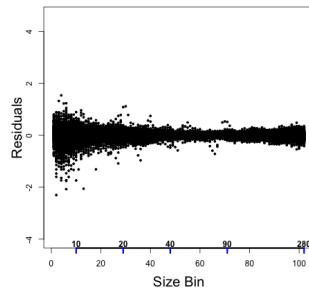
(e) Pre-closure, upwind



(f) Pre-closure, downwind

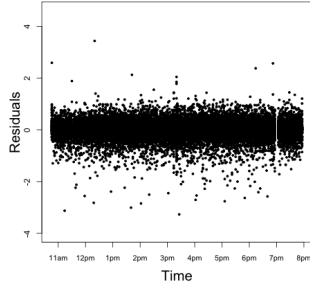


(g) Closure, upwind

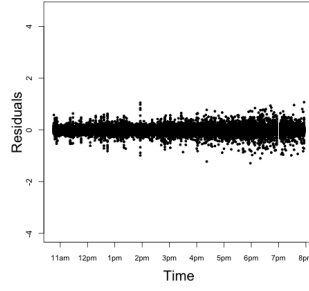


(h) Closure, downwind

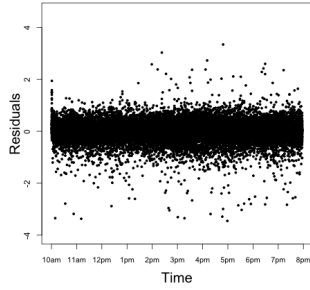
Figure 3.15: Upwind and downwind residuals, $\hat{e}_{izst}$, over size bin for pre-closure and closure Fridays(a)(b)(c)(d) and Sundays(e)(f)(g)(h). Residuals demonstrate similar behavior as observed on Saturdays: they are larger for smaller sizes especially for the upwind data.



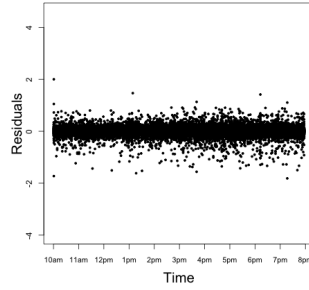
(a) Pre-closure, upwind



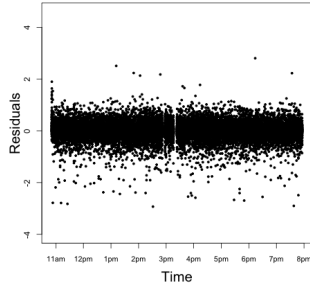
(b) Pre-closure, downwind



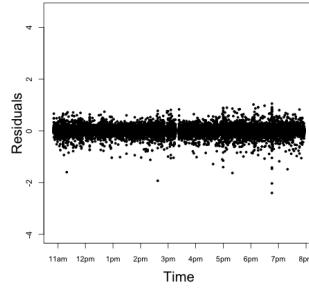
(c) Closure, upwind



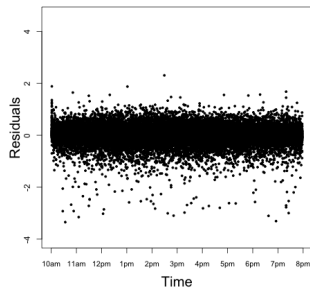
(d) Closure, downwind



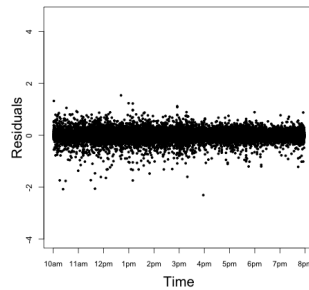
(e) Pre-closure, upwind



(f) Pre-closure, downwind



(g) Closure, upwind



(h) Closure, downwind

Figure 3.16: Upwind and downwind residuals, $\hat{e}_{izst}$, over time for pre-closure and closure Fridays(a)(b)(c)(d) and Sundays(e)(f)(g)(h). As observed on Saturdays, residual variance is roughly constant over time and is larger for model fit to upwind data than to downwind data.

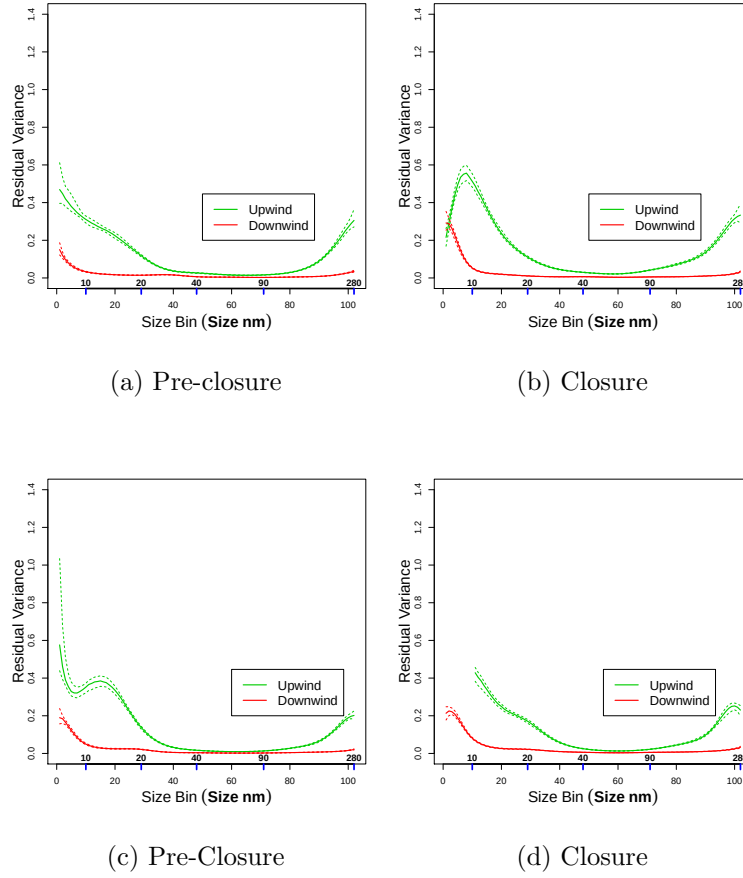


Figure 3.17: Posterior median σ_{zs}^2 estimates for Fridays (row 1) and Sundays (row 2). Dotted lines show 95 percent credible intervals.

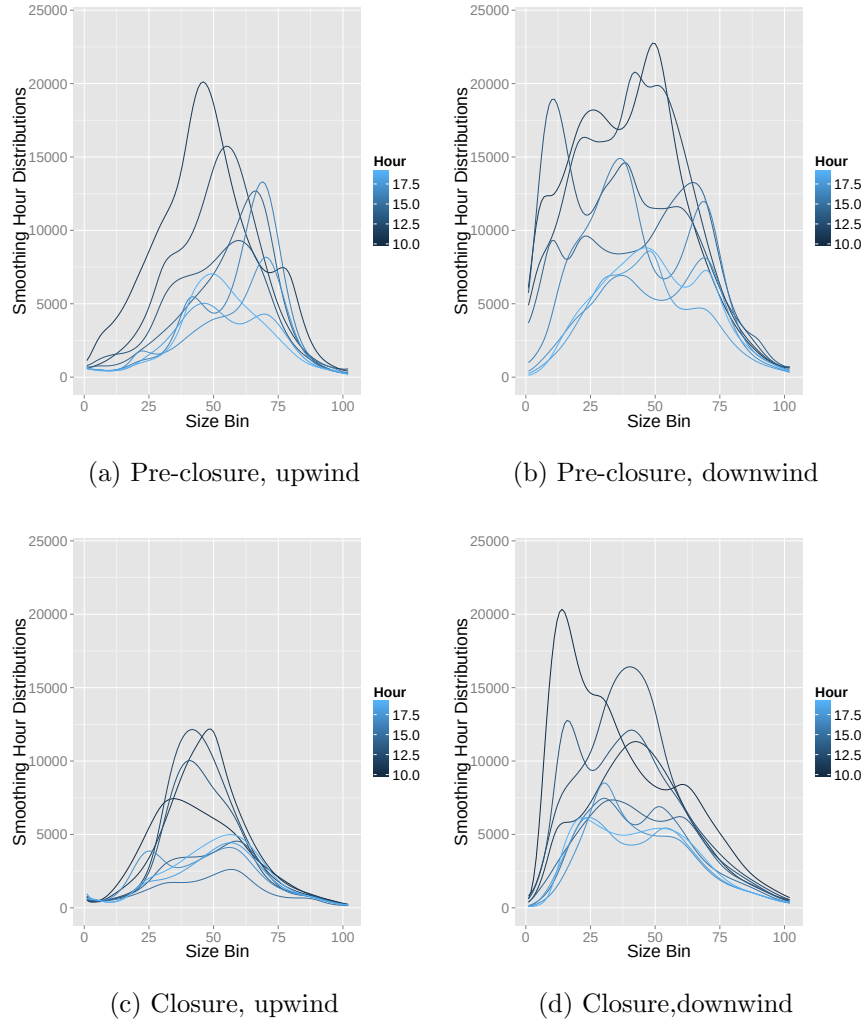
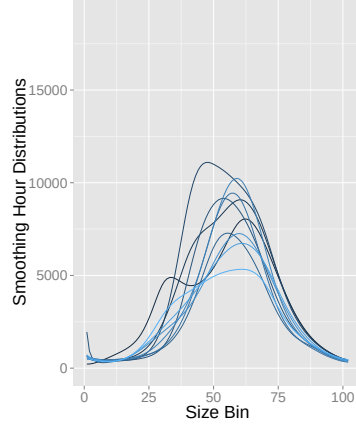
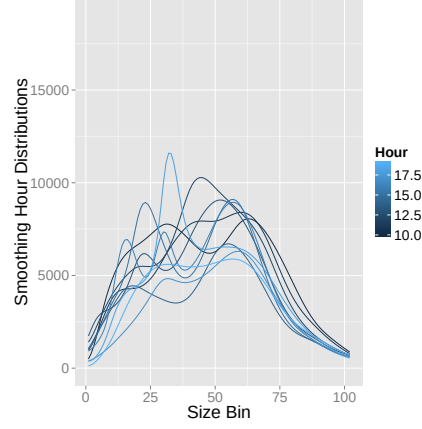


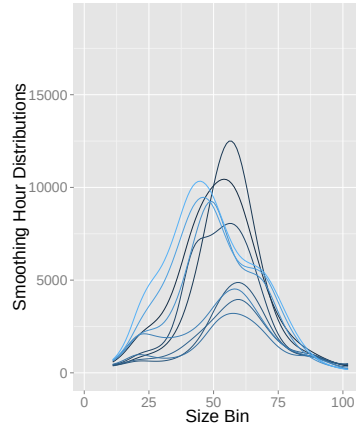
Figure 3.18: Smoothing hour distributions over the course of the day for pre-closure(a)(b) and closure(c)(d) Fridays for both upwind and downwind positions. Plots suggest that for all Fridays, both upwind and downwind UFP size distributions tend to decrease into the evening, but they do not begin at lower levels as seen on Saturdays. Pre-closure Friday, and to some extent closure Friday, show much more variation throughout the day for both upwind and downwind smoothing hour distributions compared to other days. Two or more modes are seen during many hours for closure downwind Friday as the freeway was not completely closed.



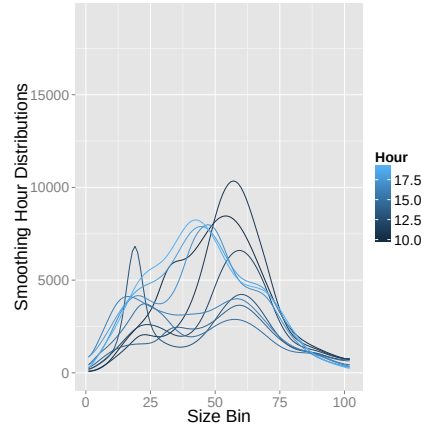
(a) Pre-closure, upwind



(b) Pre-closure, downwind



(c) Closure, upwind



(d) Closure, downwind

Figure 3.19: Smoothing hour distributions over the course of the day for pre-closure (a)(b) and closure(c)(d) Sundays. There are no upwind estimates for the closure Sunday for size bins less than 10 because almost all observations were missing. Pre-closure Sunday also shows pattern of both upwind and downwind UFP size distributions decreasing into the evening, though during the closure Sunday this is not the case. Across all sizes UFP counts are at their lowest on Sundays, however two modes are seen during many hours for closure downwind Sunday as the freeway was opened at 12pm.

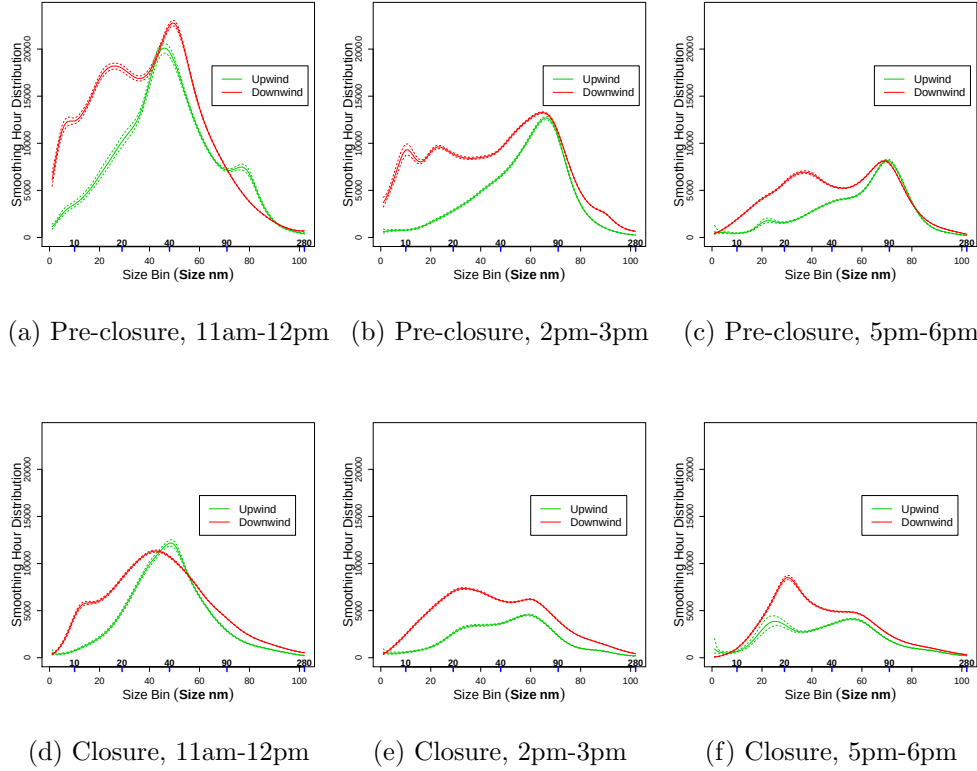


Figure 3.20: Upwind and downwind smoothing hour distributions for 11am-12pm(a)(d), 2pm-3pm(b)(e), and 5pm-6pm(c)(f) on all Fridays. Ninety-five percent credible intervals plotted with dotted lines. For all selected hours there is an additional mode at the downwind location at smaller size bins as the freeway was never completely closed. The downwind pre-closure Friday has 3 modes during the earlier selected hours.

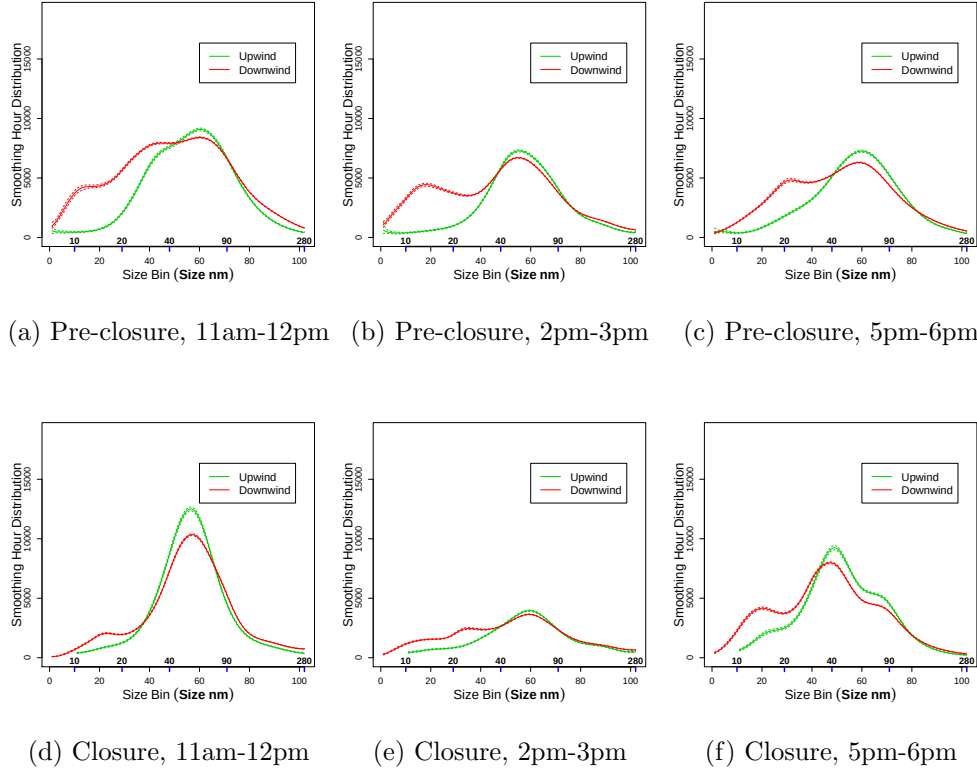


Figure 3.21: Upwind and downwind smoothing hour distributions for 11am-12pm(a)(d), 2pm-3pm(b)(e), and 5pm-6pm(c)(f) on all Sundays. Ninety-five percent credible intervals plotted with dotted lines. For all selected hours except for the closure Sunday from 11-12pm (Figure(d)) when the freeway was still completely closed, there is an additional mode at the downwind location at smaller size bins.

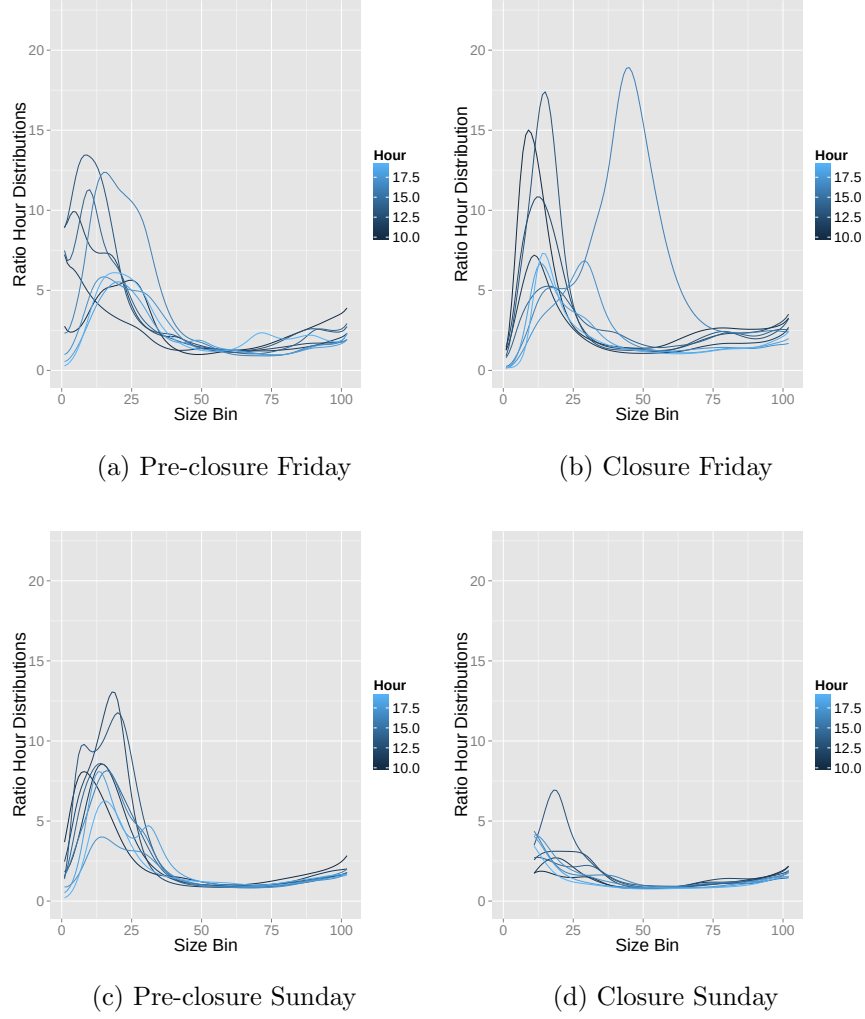
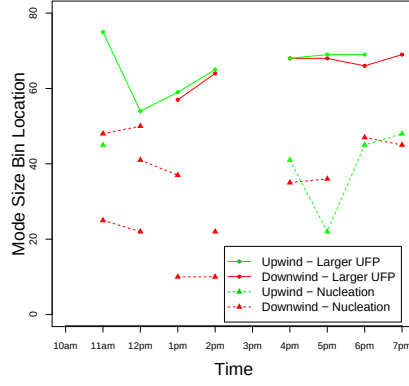
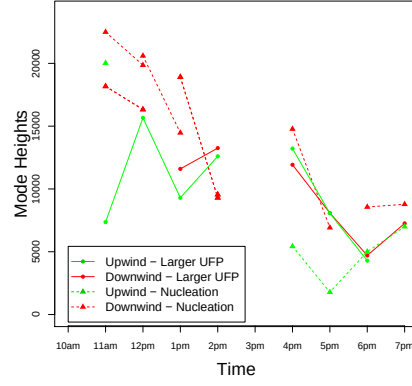


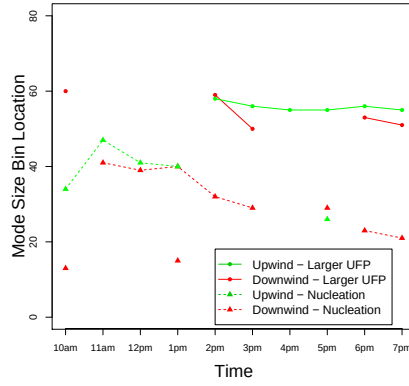
Figure 3.22: Ratio hour distributions of Equation (3.27) over time for pre-closure(a)(c) and closure(b)(d) Fridays and Sundays. The ratio hour distributions on pre-closure and closure Fridays are similar, indicating the upcoming closure did not affect downwind-upwind UFP size distribution ratio, but the ratio hour distributions are higher for the pre-closure Sunday compared to the closure Sundays. As seen on Saturdays, the ratio hour distributions also tend to decrease by the end of the day on all Fridays and Sundays.



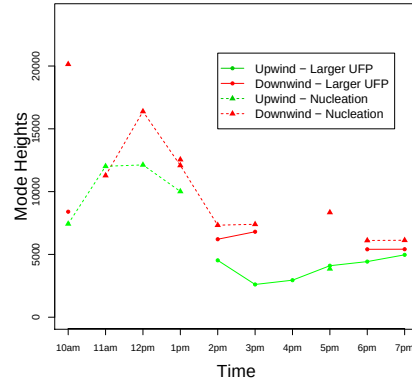
(a) Pre-closure, mode location



(b) Pre-closure, mode height

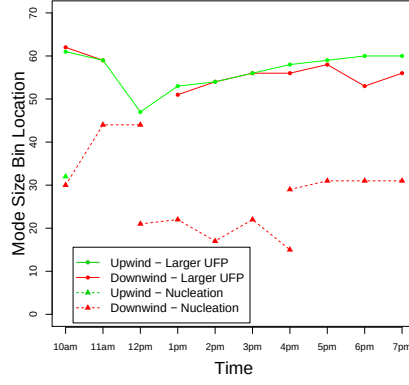


(c) Closure, mode location

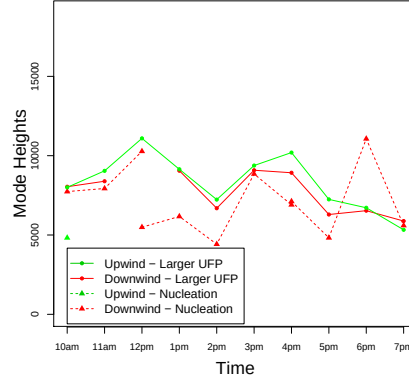


(d) Closure, mode height

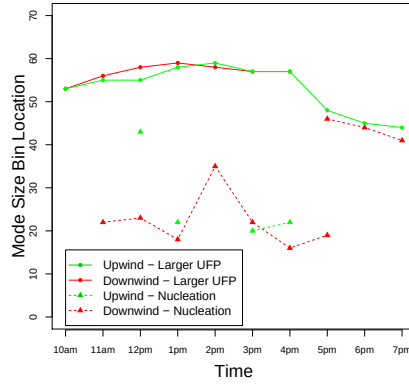
Figure 3.23: Figures(a)(c) plot the size bin locations of nucleation modes (dotted lines with triangles) and larger UFP modes (solid lines with circles) over time for the upwind and downwind locations on the pre-closure and closure Fridays. Figures (b)(d) plot the heights of these nucleation modes and larger UFP modes over time. The erratic behavior of both the upwind and downwind UFP size distributions on the pre-closure Friday is reflected in Figures 3.23(a) and (b). Both nucleation and larger UFP modes disappear and reappear from hour to hour at different size bins, and their height changes drastically across hour. Toward the end of the day, the height of each decreases. The closure Friday tends to have more constant nucleation and larger UFP mode locations, which decrease over the course of the day, however these are still more variable than those seen on Saturdays. The nucleation mode does not disappear on the closure Friday, and has levels similar to the pre-closure height until the end of the day.



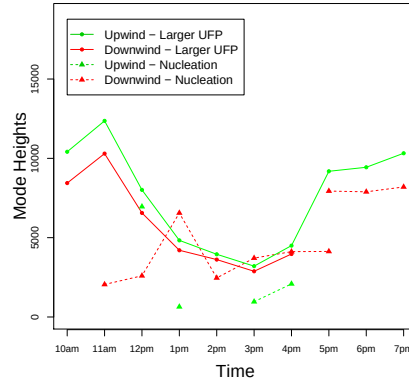
(a) Pre-closure, mode location



(b) Pre-closure, mode height



(c) Closure, mode location



(d) Closure, mode height

Figure 3.24: Figures(a)(c) plot the size bin locations of nucleation modes (dotted lines with triangles) and larger UFP modes (solid lines with circles) for the upwind and downwind locations over time on the pre-closure and closure Sundays. Figures (b)(d) plot the heights of these nucleation modes and larger UFP modes over time. Sunday larger UFP modes tend to resemble those seen on Saturdays, located at around size bin 60 and decreasing slightly in height throughout the day. However, the height of the larger UFP mode increases slightly by the end of the day closure Sunday, possibly an affect of the reopening of the freeway. Nucleation modes are present on both pre-closure and closure Sundays, but the closure Sunday nucleation mode is lower in height.

3.6 Discussion

Our use of functional time-series methods allows inference on UFP size distributions over time for both upwind and downwind locations, not available in Quiros et al. (2013) which provides day long summaries using simple averages for each particle size bin. Simple averages do not allow the computation of other statistical summaries of interest to researchers. Additionally, simple averages ignore missing values. Our methods are flexible and allow researchers to calculate numerous other quantities with uncertainty measures, such as the difference of differences. They also permit inference about actual particle counts not available using modal methods and, unlike simple averages, are able to impute missing values.

One of the useful statistical summaries we provided in this analysis were mode locations and heights over time. Though the larger UFP mode heights varied on different Saturdays, this level does not seem to depend on the freeway closure, nor on the upwind or downwind position. On all Saturdays we see an initial increase, then a decrease in the mode height over the course of the day, possibly reflecting natural patterns in these particle counts affected by changing meteorological conditions over the course of the day. The larger UFP mode stays consistently around size bin 60. Friday showed more variable behavior in the location and height of the larger UFP mode. Sunday showed similar patterns as Saturday. The height and location of the nucleation mode was much more variable across time, particularly for Fridays. This mode was greatly affected by the freeway closure, disappearing for most of the day on the Saturday of complete closure and only reaching a height of less than 3,000 particles maximum, compared to over 15,000 particles on non-closure Saturdays.

The flexibility of these methods allowed us to provide other useful statistical summaries such as multiplicative differences and difference of difference estimates. On non-closure days, the multiplicative difference between downwind and upwind

particle counts was variable earlier in the day, but tended to decrease by later in the day, possibly due to changes in meteorological or traffic conditions. Multiplicative difference plots show the closure had a major effect on the ratio of downwind to upwind counts, as the ratio was less than 5 during the closure Saturday compared to up to 20 during the non-closure periods. The difference of difference plots allow researchers to see on one plot how downwind increases for the non-closure weekends compare to the closure weekend, and how these compare to each other. The freeway closure resulted in lower downwind-upwind UFP size distribution additive differences compared to both the pre- and post-closure weekends, however this difference was higher during the post-closure weekend indicating traffic flow possibly increased during the weekend following the freeway closure.

Another major benefit of these methods are the ease in which model fit can be evaluated, both through the use of DIC and through residual checks allowing visual inspection of model fit across size bin and time. Our methods are easily adapted to different model specifications, such as the addition of variables to further explain changes in counts over time, such as traffic flow or meteorological conditions. Additionally, unlike methods such as a simple average, these methods allow the estimation of standard errors as well and imputation of missing values.

Though our methods allowed for the imputing of missing values, the malfunction in the upwind SMPS machine could possibly lead to biased estimates for the smaller size bins. This is because it is possible that lower actual particle counts were more likely to be given erroneous zero count readings by the SMPS machines. However, in terms of inference about actual particle counts, the effective bias is likely very small, as demonstrated by the predicted hour plots in Figures 3.10, 3.18, and 3.19 where upwind estimates were only slightly larger than downwind estimates for size bins less than 10.

Given UFPs' toxic nature with health effects strongly related to particle size bin, these methods provide information needed for understanding how UFP ex-

posure changes over time and under different traffic conditions near roadways. Closing the freeway caused the nucleation mode, which contains the smallest and most toxic particles, to completely disappear for most of the closure Saturday. When it did appear its height was greatly reduced. From a public health perspective, this implies that policies limiting vehicle traffic can have an immediate impact in reducing the counts of the smallest UFPs near a major roadway. Further studies using datasets with a longitudinal design could help to verify these important initial findings.

CHAPTER 4

Next Steps

The goal of this dissertation was to provide statistical methods to summarize the behavior of UFP size distributions near high emission sources. A major benefit of these methods is the ease in which model fit can be evaluated, both through the use of DIC and through residual checks allowing visual inspection of model fit across size bin and time. Graphical techniques also allow for the evaluation of the fit of random effects. Our methods allow uncertainty in predictions to be quantified and communicated graphically in ways easily interpretable to a non-statistician. We provide statistical summaries of useful quantities currently used by researchers such as mode locations and heights. Our methods are easily adapted to different model specifications, a number of which are our focus for future work.

The following list highlights our ideas for future work:

1. We propose modeling autoregressive error as a function of the following model covariates:
 - Autoregressive (AR) parameter θ will be a function of size bins, so that we have AR θ_s ,
 - For the bus model, AR θ will be modeled separately for engine-on and engine-off states, so that we have AR as θ_{s1} for $t < 0$, and θ_{s2} for $t \geq 0$.
2. For the 405 data, we will introduce covariates to further explain the evolution of UFP counts over time
 - We will consider traffic flow, temperature, wind speed, and humidity.

- We will explore different functional forms, first allowing UFP counts to change at a size bin dependent linear rate over time. We will let these rates vary smoothly by size bin s by modeling them as cubic B-spline functions of s
3. For the 405 data, we will explore smoothing UFP size distribution modes over time using a moving average rather than a discrete hour average. This will allow the visualization of changes in mode behavior continuously over time while still smoothing over short term fluctuations. Statistical summaries will be developed to compare moving averages between different time periods and days.
 4. We will develop experiments and statistical methods to formally evaluate the contribution of SMPS measurement error to residual variance λ^2 which will lead to greater accuracy in back-transformed inferences.

CHAPTER 5

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