EVALUATING THE AIRPACT-4 AIR QUALITY MODEL FOR
WINTERTIME CONDITIONS IN THE YAKIMA VALLEY,
AND REFINING THE TREATMENT OF RESIDENTIAL
WOOD COMBUSTION EMISSIONS

By

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Abstract

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In January 2013, researchers at Washington State University and Central Washington University performed the Yakima Air Wintertime Nitrate Study (YAWNS) in order to obtain a better understanding of the elevated PM$_{2.5}$ and nitrate levels in the Yakima Valley. The YAWNS measurements provided an extensive data base that was used to evaluate the AIRPACT-4 air quality forecast system during stagnant, wintertime conditions. The data show significant differences in the daily pollutant concentration patterns for clear sky stagnant conditions vs. cloudy stagnant conditions, when it appears low clouds promote significant vertical mixing and secondary aerosol production. Results from several different model
configurations are compared to the available observations. These comparisons focus on the ability of the modeling system to capture the very distinct concentration patterns that occur in clear sky vs. cloudy conditions.

The emission inventory for Yakima is then evaluated using source attribution for mobile sources and residential woodstove emissions. Based upon both observed source contributions and model results, residential wood combustion (RWC) is shown to be a prominent source of wintertime PM$_{2.5}$ in the Yakima Valley. However, the numerous uncertainties in compiling wood stove emissions limit the accuracy of wintertime air quality forecasts in the region. The latter portion of this study expands the scope to the entire Pacific Northwest for testing a forecast-mode temperature adjustment to RWC emissions within the AIRPACT-4 model. The temperature-adjustment experiment failed to significantly improve AIRPACT-4 PM$_{2.5}$ forecasting, but highlighted the more fundamental limitation of precise spatial allocation of RWC emissions. Accordingly, a variety of RWC spatial surrogates are tested throughout the AIRPACT domain. Results indicate the best approach to improving RWC emissions processing, and subsequent wintertime PM$_{2.5}$ modeling, is the application of an aggregate spatial surrogate for woodstove emissions. In the Pacific Northwest, a RWC spatial surrogate blending maps of forest coverage and rural population helped ease the tendency to over-predict wintertime PM$_{2.5}$ in many populated urban areas, while the census data for population using primary wood heating, though outdated, continued to be most representative of wood burning emissions in most of the rural and agricultural sectors of the domain.
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CHAPTER ONE

INTRODUCTION

The United States Environmental Protection Agency has long established that consistent, high concentrations of airborne fine particulate matter (PM$_{2.5}$) contribute directly to respiratory and cardiovascular disease. Recent studies even suggest that fine particulates comprise one of the biggest precursors to human disease on a global scale (Lim et al., 2012). Therefore PM$_{2.5}$ is treated as a criteria pollutant by the U.S. EPA and monitored closely under the Clean Air Act, with a primary 24-hour standard of 35 μg/m$^3$ (U.S. EPA, 2013). Furthermore, the EPA’s most recent National Emissions Inventory, NEI 2014, subdivides PM$_{2.5}$ into dozens of chemical species from thousands of sources. The highest PM$_{2.5}$ concentrations are often formed during wintertime stagnation conditions. PM$_{2.5}$ tends to peak when wintertime reductions in solar heating contribute to a shallow planetary boundary layer, which limits vertical mixing; this is particularly an issue in confined urban valleys. This process, in combination with low wind speeds, characterizes typical wintertime stagnation conditions. Thus in urban valleys during stagnant wintertime conditions, mobile and other combustion sources, including residential wood heating, can generate high ground-level PM$_{2.5}$ concentrations.

The Yakima region of Washington State exemplifies this type of urban area located in complex terrain. The population of nearly 250,000 resides in a valley on the eastern edge of the Cascade Mountain range, bordered by large hills on nearly all sides. The Yakima Valley, consisting of about 6,150 square miles, is bordered by the
Cascade Mountains to the west, Wenatchee Mountains to the north, Rattlesnake Hills to the East, and Horse Heaven Hills to the south. In recent years, high PM$_{2.5}$ levels have been measured in Yakima that have approached non-attainment values at times during the winter. Within this context, the Washington State Department of Ecology observed an unusually high fraction of nitrate PM$_{2.5}$ specific to the Yakima Valley. These factors motivated Ecology to fund an extensive measurement campaign in January 2013, through Washington State and Central Washington Universities. The project was named the Yakima Airshed Wintertime Nitrate Study (YAWNS). This campaign emphasized speciated aerosol measurements, with the goal of facilitating a better understanding of the conditions leading to the elevated levels of wintertime PM$_{2.5}$ in Yakima. High-resolution gas-phase and meteorological measurements were also made.

1.1 GOALS AND OBJECTIVES

The overall goal for this work is to evaluate the AIRPACT-4 air quality modeling framework for wintertime stagnant conditions in the Pacific Northwest and to identify aspects where improvements are needed. This comprehensive evaluation was motivated by the availability of extensive high-resolution measurements of pollutants and meteorology for the Yakima Valley via the YAWNS campaign, and the lack of published evaluations for the current AIRPACT-4 configuration. The specific objectives include 1) to evaluate AIRPACT-4 for the YAWNS observational period, including both clear and cloudy stagnation episodes
conditions in the observation period; 2) to assess the role of residential wood combustion (RWC) in the model results in comparison to the observational source attribution analyses; 3) to test new RWC emission algorithms to improve model performance. The end results of this work will include better documentation of the performance of AIRPACT during wintertime conditions, a better understanding of the contribution of RWC emissions to wintertime PM levels, and an assessment of methods to improve the treatment of RWC emissions within the AIRPACT framework.

1.2: LITERATURE REVIEW

The biggest source of PM$_{2.5}$ emissions in wintertime Yakima is biomass burning – which is dominated by residential wood combustion for heating purposes. Thus, it is noteworthy that carbonaceous PM$_{2.5}$ poses a particularly high health risk and also acts as a climate change agent (Highwood and Kinnersly, 2006) due to its tendency to scatter and absorb incoming and outgoing radiation. Accordingly, the EPA’s most recent National Emissions Inventory, NEI 2014, subdivides PM$_{2.5}$ into dozens of chemical species from thousands of sources. Unfortunately, some of the most significant sources of carbonaceous particulates, including RWC, are difficult to quantify. For instance biomass combustion is simply estimated by reporting agencies. In the Pacific Northwest, RWC emissions are reported by the local clean air agencies as estimated county totals in tons per year. These estimates are based on a combination of RWC usage surveys and census data on use of RWC as a primary
source of household heat. State agencies distribute the county totals spatially based on surrogate grids reflecting census data for population and wood stove usage. Temporal allocation is modeled according to the EPA’s temporal profile factors for expected daily human behavior and seasonal climate patterns. Emission factors in pollutant mass emission rate per unit time or per unit of wood burned are taken from EPA compilations for various combustion devices (US EPA AP-42, 1995).

Combustion conditions, such as wood moisture content, strongly influence smoke content, but are difficult to quantify on a large scale. Strushka (1993) determined that poor, or incomplete, wood combustion generates much more total PM, CO, and gaseous volatile organic compounds (VOCs) than more complete combustion does. Furthermore, Bari et al. (2009) characterized the very different organic compound emission fingerprints of hardwood versus softwood burning. Additionally, the RWC emissions inventory fails to address the burning of different wood types than those for which the stoves are certified, and suffers from unreliable testing methods.

As typified by Yakima, RWC is often a major contributor to carbonaceous PM$_{2.5}$ in non-attainment areas. Many studies over the last several decades have recognized the significant effects of RWC on cold-weather particulate levels. An early study in the Portland, OR and Vancouver, WA area attributed approximately 52% of hazardous particles emitted in January to RWC (Cooper, 1980). In the warmer climate of San Jose, CA, RWC contribution to wintertime PM$_{10}$ was still found to be around 42% (Fairley, 1990). In Christchurch, New Zealand, over 90% of wintertime ambient PM was attributed to heating stoves and open fires burning
wood (McGowen et al., 2002). Across eight provinces of Canada, RWC was found to account for 30% of annual PM emissions (Larson and Koenig, 1994).

The circumstances that make RWC emissions so difficult to accurately compile likewise create a barrier to reconciling model results to field studies when RWC is significantly involved. Napelenok et al. (2014), one of many studies to attempt to improve RWC emission inventories, recommended applying a daily temperature adjustment algorithm to modeled RWC emissions. Typically, RWC emissions are assigned temporal profiles on an hourly, daily, and monthly basis to reflect expected human activity and climate, but have not referenced temperatures on a smaller timescale. The Napelenok et al. study observed strong inverse correlations between RWC and daily minimum observed temperature, which can vary greatly in a given month. This particular study generated significant improvements in their CMAQ PM$_{2.5}$ simulation for the Southeastern U.S. region by adjusting the temporal application of the RWC inventory accordingly. It may be inferred that a similarly apt modification to RWC modeling in the Northwest, where wood stoves are much more common, could have an even greater impact.

Ultimately the YAWNS measurement campaign generated a wealth of high-resolution measurements of speciated aerosol, gas-phase pollutants, and meteorological factors. Cumulatively, these measurements shed light on wintertime pollutant production and dispersion in the Yakima Valley, but also pointed to some additional uncertainties regarding secondary aerosol production. The extensive observation data has the secondary advantage of providing a strong backdrop against which to evaluate the current northwest air quality forecasting system,
AIRPACT-4, specifically for complex wintertime stagnant conditions. Given the generally biased nature of RWC emission inventories, a secondary focus of improving the accuracy of northwestern wood stove emission sector of the NEI was adopted.

1.3 FINE PARTICULATE MODELING

In state-of-science air quality models, such as the Community Multiscale Air Quality model (CMAQ) used in AIRPACT-4 and the Comprehensive Air Quality Model with Extensions (CAMx) – the two most widely used photochemical grid models in policy making - the dispersion of gas and particulate pollutants are governed by full continuity equations. These fundamental equations characterize the number flux of particulates as a summation of the rates of atmospheric dispersion, molecular diffusion, emissions of all precursors, deposition, sedimentation, nucleation, coagulation, and washout. Of course, to create rates of emission and atmospheric diffusion, a chemical transport model (CTM) requires emission, meteorology, initial conditions, and boundary conditions along the borders of the domain. The CTM then processes pollutant dispersion as an Eulerian model on a three-dimensional grid fit over mapped land data. Basic outputs include hourly concentrations of aerosol pollutants, wet and dry deposition fluxes, and visibility metrics. In CMAQ, particle size distribution in the model is built from three lognormal sub-distributions, or modes: Aitken mode (less than 0.1 μm), accumulation mode (0.1-2.5 μm), and coarse mode (2.5-10 μm). For each mode, the model predicts the chemical
components of the particulate matter (PM), including those of both primary and secondary origin. From the combination of number concentrations and size distributions, the CTM can calculate and report the mass concentrations of total PM$_{10}$ and PM$_{2.5}$. CAMx, on the other hand, only fits aerosols into size bins of PM$_{2.5}$ and PM$_{10}$. In addition, CMAQ and CAMx differ in treatments of inorganic and organic aerosols, sea salt and wet and dry deposition. However they can be configured to utilize similar options for using emissions processors and meteorology inputs.

Because the simulation of PM formation involves the transport and interactions of primary and secondary pollutants encompassing gas, solid, and aqueous phases, PM concentrations are generally more difficult to predict than gas-phase pollutants (Chen et al., 2007). The Air Quality Model Evaluation International Initiative (AQMEII) mandated comprehensive evaluations of CMAQ and CAMx for North America and Europe. Observations came from 958 U.S. EPA Air Quality System sites, and CMAQ was run with 12-km horizontal grid spacing and 34 vertical layers (13 layers below 1 km). The AQMEII evaluation for CMAQ v.4.7.1, the version used in AIRPACT-4 for this YAWNS evaluation, showed a consistent overestimation of PM$_{2.5}$ in the winter months of North America for 2006 data, with a normalized mean bias (NMB) of 30.4% (Appel et al., 2013). The largest overestimations occurred mostly in the Western U.S. On the other hand, CMAQ underestimated summer PM$_{2.5}$ with a NMB of -4.6%. Appel et al. attribute the overestimations of winter PM$_{2.5}$ mostly to overestimation of unspeciated PM$_{2.5}$ mass, as well as some smaller overestimations of elemental carbon (EC) and organic carbon (OC). The next version of CMAQ, v5.0, addressed this issue by including the speciation of trace
metals, allowing for better comparison of model estimates to observations. However, the next phase of the AQMEII evaluation, for CMAQ v5.0.1, showed that wintertime PM$_{2.5}$ bias in North America actually increased to a NMB of 60% for the same dataset (Hogrefe et al., 2014). The authors related the increased bias to the introduction of a wind blown dust module to the new model version, updates to the stable layer boundary treatment, and revised emissions inventories.

The AIRPACT (Air Indicator Report for Public Awareness and Community Tracking) modeling framework was developed to provide air quality forecasts for the immediate future to people of the Washington, Oregon, and Idaho. AIRPACT collaborators, all members of the NW-AIRQUEST consortium, include the Puget Sound Clean Air Agency, Washington State Department of Ecology, the University of Washington Atmospheric Sciences Department, U.S. EPA Region 10, and the Washington State University Laboratory for Atmospheric Research, Oregon Department of Environmental Quality, and Idaho Department of Environmental Quality. The initial version of AIRPACT used weather forecasts from the fifth-generation Penn State-NCAR Mesoscale Model (MM5) to drive the California Meteorological Model (CALMET)/California Photochemistry Grid Model (CALGRID) Eulerian photochemical modeling suite (Vaughan et al., 2002). Soon thereafter, the ability to provide relevant modeling guidance for the Puget Sound region motivated an expansion of the domain to include all of Oregon and Idaho. The AIRPACT-3 system was updated to an MM5-CMAQ system, with the Sparse Matrix Operator Kernel Emissions (SMOKE) as the emissions processor. A model evaluation for August-November 2004 EPA-AQS data found a 17% NMB in PM$_{2.5}$ forecasting,
compared to a 32% NMB at the more rural observation sites of the IMPROVE network (Chen et al. 2007). However, there was no clear distinct concentration range in which AIRPACT-3 performed better. The current iteration, AIRPACT-4, uses CMAQ v.4.7, SMOKE, and meteorology modeling by the Weather Research and Forecasting model (WRF) v.3.4. Forecasts are calculated on a domain with a 4-km horizontal spacing.

1.4 YAKIMA AIR WINTER NITRATE STUDY OBSERVATIONS

The Yakima Air Wintertime Nitrate Study (YAWNS, VanReken et al., 2014) provided both the motivation for this study and extensive data for model evaluation. YAWNS was a comprehensive air quality measurement campaign motivated by elevated PM$_{2.5}$ observations in the region and a higher nitrate PM$_{2.5}$ fraction than the rest of Washington State. Accordingly, Washington State University deployed its Mobile Atmospheric Chemistry Lab to Yakima from January 5 to 26, 2013, in Yakima to take high-resolution measurements of speciated PM$_{2.5}$ and volatile organic compounds (VOCs), as well as NO$_x$, CO, CO$_2$, and meteorological measurements.

The YAWNS measurement campaign indicated a build-up of high concentrations of both aerosol and trace gas pollutants during clear-sky stagnation conditions that characterized the first half of the month. Starting on January 16$^{th}$, conditions changed to cloudy but remained stagnant, and were accompanied by rapid dilution of primary gas and particulate pollutants. However, secondary pollutants, including nitrate aerosol, remained elevated and exhibited a much
slower decline in concentration over several days. This observed behavior, summarized below in Figure 1.1, was believed to be due primarily to mixing effects associated with clouds. It should be noted that there was a burn ban in effect during the period of rapid dilution. Following the sweeping drops in all primary pollutants, on the 16th, the burn ban was reduced from a Stage 2 to a Stage 1 ban. However, burn bans are expected to result in a more gradual reduction in pollutant levels than what was observed in this episode. Additionally, a burn ban would not solely account for the drastic reductions in gas-phase CO and NOx that were also observed. Based on measurements, the YAWNS report concluded that the rapid dilution in primary pollutants was more closely tied to the simultaneous formation of a cloud layer above Yakima. Meanwhile, the relatively gradual decline of secondary species suggested the presence of secondary chemical production effects associated with the clouds. In all time series plots included in this report, the date and time are shown in Pacific Standard Time (PST).
Figure 1.1: Time series for observed ambient concentrations of both primary (left) and secondary (right) pollutants during both clear and cloudy conditions in Yakima during January, 2013. A persistent cloud cover formed on the 16th, coinciding with a rapid dilution of primary pollutants and much more gradual dilution of secondary species. Date provided by VanReken et al. (2014).

The apparent complexity of the pollutant-meteorology interactions during January 2013 in Yakima called for a closer look at state-of-science air quality model results for this domain. This served the dual purposes of reinforcing the understanding of pollutant formation and transport, and evaluating the performance of the current model structure. While modeled processes cannot be blindly adopted as factual, the model output can be evaluated on the basis of accurate observations. The extensive pollutant and meteorological measurements of the YAWNS campaign provided an opportunity to evaluate the AIRPACT-4 for a
wintertime stagnation scenario with relatively complex terrain and meteorology. Accordingly, the majority of this paper will be devoted to AIRPACT-4 model evaluation.

1.5 AIRPACT MODELING SYSTEM

The AIRPACT modeling framework forecasts air quality for the immediate future in Washington, Idaho, and Oregon. It was developed collaboratively by the U.S. EPA Region 10, the Washington State Department of Ecology, Washington State University and the University of Washington with the consistent goal of providing timely and convenient air quality information and forecasts to the public in the form of both measurements and model results (Vaughan et al., 2002). The most current iteration of the AIRPACT air quality modeling system, “AIRPACT-4,” utilizes the Community Multiscale Air Quality (CMAQ) model v4.7.1 as the chemical production and transport processor. CMAQ is used in combination with the Sparse Matrix Operator Kernel Emissions (SMOKE) processor and the Weather Research and Forecasting Model (WRF) v3.4.1 for meteorology input. Forecasts are calculated on a 4-km gridded domain. For this experiment, modifications were made alternately to the WRF meteorological output and within the SMOKE inventory and emissions processing.

For general AIRPACT-4 evaluation against YAWNS observations, two versions of the model were run with identical emissions inputs but different meteorological results from the Weather Research and Forecasting Model (WRF).
The initial run used the standard WRF forecast configuration with 37 defined vertical layers, including a 39-meter ground layer. The number of vertical layers was collapsed to 21 in the Meteorology-Chemistry Interface Processor (MCIP) before being input to CMAQ. The second AIRPACT-4 run employed WRF runs performed by Zhang et al. (2013), using both finer layer resolution in WRF near the surface and meteorological nudging in order to assess the impact of refined meteorology on AIRPACT-4 accuracy. The bottom layer was replaced by two layers. In the meteorological nudging, both analysis nudging and observational nudging were implemented for the WRF simulations. The WRF model includes the option to apply these adjustments in order to improve prediction accuracy; the accuracy of forecast meteorology fields tends to decrease with time after the initial conditions. Observational nudging requires uploading and converting measurement data to WRF-compatible format, and then applying Newtonian relaxation to forecast terms at individual grid points toward observations at those locations (Otto, 2008). The process of Newtonian relaxation involves adding an empirical term to the prognostic equations, which effectively nudges the solution towards the observations. The scales of impact are based on user-specified radii of influence, time windows, and relaxation time scales. For the YAWNS study, observation data was only available near the surface. Thus, to address terms in the vertical layers, analysis nudging was applied to 3D meteorological variables by upward propagation of nudged model predictions upward using minimization of sum of squares of deviation from observation. The result is a set of dynamic analyses that more accurately characterize local meteorology patterns. The net effect of the
meteorological nudging on air quality model output is discussed in the Results section.

1.6 AIRPACT EMISSIONS INVENTORY

The AIRPACT-4 modeling system also relies on emission inputs, which are processed by SMOKE from reported annual totals into hourly, gridded, CMAQ-compatible format. SMOKE utilizes the EPA's National Emissions Inventories (NEI), which are updated every three years with comprehensive estimates of air emissions of both Criteria and Hazardous air pollutants from all source categories. The categories are assigned Source Classification Codes (SCC) for identification within models. Estimates are provided by State, Local, and Tribal air agencies for sources within their jurisdictions (US EPA, 2008 NEI Version 3, 2013). These values are sorted into county totals, which are identified in models by the counties' Federal Information Processing Standard (FIPS) codes.

For the YAWNS evaluation and source attribution performed in this study, AIRPACT-4 utilized the 2008 NEI, which was the most current version at the starting time of the study. Sources in the NEI are allocated into five different categories: Point, Onroad, Nonroad, Nonpoint, and Event. The Events category consists of wildfires and prescribed burns, and is processed by a separate “Fire” module within SMOKE. The Onroad and Nonroad categories provide estimated emission totals for vehicles on the ground. The Point category consists of facility emissions estimates located at a stationary site, while the Nonpoint category
contains estimates for combinations of smaller sources, such as industrial and residential fuel combustion, best listed as aggregates or “area” sources. In addition, biogenic emissions are estimated on a county basis by month, and processed separately within SMOKE on the basis of gridded land use characteristics.

The AIRPACT-4 modeling system relies directly upon EPA NEI totals for counties within its Northwestern U.S. domain. For the YAWNS case study, the most relevant emissions estimates were those for January in Yakima County, Washington (FIPS code = 53, 77). In contrast to the complicated terrain and weather that characterized this site, the wintertime emissions in Yakima are relatively simple, consisting predominantly of mobile sources and biomass burning. The mobile sources here consist mostly of on-road vehicles, while the biomass burning represents residential wood combustion (RWC) for home heating purposes; a consequence of the typically cold winter weather. The major effects of on-road mobile emissions are steady emissions of NO\textsubscript{x}, CO\textsubscript{2}, CO, O\textsubscript{3}, hydrocarbons, and PM\textsubscript{2.5} along the major roadways, especially during peak commute hours. In AIRPACT-4, mobile emissions are estimated by the Motor Vehicle Emission Simulator (MOVES), developed by the EPA. AIRPACT-4 uses MOVES to create emissions estimates from on-road mobile sources, based on pre-calculated MOVES-based emissions indexed by meteorology data, to generate hourly gridded, CMAQ-ready files of speciated emissions. The source inputs for MOVES include road type and vehicle type. Vehicle type population is a distinct variable, which is used in combination with temperature and relative humidity to calculate start and off-road evaporative emissions. For on-road vehicles, road type and miles traveled are also applied with
the aforementioned variables to estimate speed. Pre-computed factors for combinations of these variables are supplied in look-up tables within the model. Country, state, and county codes must be identified, as for all sources, and furthermore the exact coordinates for major thoroughfares are typically provided.

In the 2011 EPA NEI, the residential wood combustion category contributes over 6% of the total estimated 6.1 million tons of PM$_{2.5}$ emissions, nationally. In the state of Washington, this percentage was nearly 19%. Of course, these numbers reflect the entire year. In the colder wintertime months, particularly in the more rural sectors of the northern states, RWC combustion often accounts for well over half of all particulate emissions. However, accurate totals of nonpoint emissions are inherently difficult to compile accurately. Since emissions from this category are not typically measured directly, they must be estimated.

The EPA’s treatment of residential wood combustion has continued to evolve in successive National Emissions Inventories. The final version of the 2002 NEI updated the RWC spatial surrogates to assign fireplace emissions to “low intensity residential land,” rather than to the previous “residential heating-wood” surrogate of the previous NEI (U.S. EPA Emissions TSD, 2008). This was a reaction to overestimating RWC in more densely populated areas, where fireplaces are not generally used for home heating. However, this over-estimation of urban wood stove emissions continued, and the RWC spatial surrogate continued to evolve through the 2008 NEI. The third version of 2008 NEI updated the RWC spatial surrogate to incorporate the latest residential housing surveys, reallocating outdoor wood boilers and indoor furnaces to have fewer in urban areas, and removing
woodstove emissions altogether in urban centers (2008 NEI v3). In addition, county
level temperature data were applied to account for burn activity as a function of
temperature in all wood burning appliances. Updates are continuing to be made to
the inventory and to the spatial and temporal allocation methods of residential
wood combustion emissions within the 2014 NEI, currently under development.
CHAPTER TWO

RESULTS

2.1 EVALUATION OF METEOROLOGICAL VARIABLES

A comprehensive AIRPACT-4 performance evaluation for YAWNS results began with a comparison of modeled to observed meteorological variables. As part of the measurement campaign, relevant met data was gathered with Vaisala WXT package instruments at one-minute intervals. Planetary boundary layer (PBL) heights were also measured with a Vaisala Ceilometer at 30-minute intervals. The most glaring discrepancy between the meteorological measurements and model results for YAWNS was the failure of WRF to simulate a sky condition change from clear to cloudy skies in Yakima. This transition occurred during the latter half of January 2013. Beginning on the 16th, the condition change corresponded closely with a rapid dilution in all primary pollutant concentrations. Thus, this meteorological model discrepancy had a pronounced negative impact on the accuracy of AIRPACT-4’s air quality forecasts for the parameters of this study.

Recall that the AIRPACT-4 modeling framework was run twice for this study, once for each set of WRF results. The first AIRPACT-4 run, identified as “version 1” in the following figures, used the WRF meteorology forecasts. The second AIRPACT-4 run, identified as “version 2,” employed WRF results for finer surface layer resolution in WRF and meteorology nudging to archived observed meteorology. This second run was performed in order to assess the impact of refined meteorology
input on AIRAPCT-4 forecast accuracy. The refined version of WRF, utilizing nudged output, did only a slightly better job of modeling cloud cover during this period. Even in the nudged meteorological output, the clouds were neither dense enough nor persistent enough to facilitate the relevant cloud effects on pollutant behavior. 

**Figure 2.1** below graphically summarizes the key meteorological differences in cloud cover observations and WRF simulations for Yakima in January 2013.

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**Figure 2.1**: Modeled and observed cloud cover fractions presented as time series.

Plot (a) shows observed and WRFv1-simulated cloud cover. This plot shows the distinct transition from clear to cloudy conditions, which occurs on the 16th, and the failure of WRFv1 to simulate this change. Plot (b) compares the cloud cover from
WRFv1 to that of WRFv2 – the nudged version. While there are some differences, the improvement in cloud modeling between versions is not significant.

The cloud coverage time series suggest that no significant differences between modeled and observed cloud cover fractions emerge until the morning of the 16th, when the cloudy period begins. At that time, the observed cloud cover fraction increases to 1.0, or full, almost continuously for the next week. Meanwhile, the modeled cloud cover fractions generally stay close to zero, or clear, until the second half of this week. It is also during this clear period that some obvious differences in cloud coverage between the two WRF runs emerge. The initial forecast run (WRFv1) actually generates some brief cloud coverage at the beginning of the cloudy period, while WRFv2 does not. On the other hand, WRFv2 forms significant cloud coverage beginning about two days earlier than WRFv1, toward the end of the pronounced cloudy period. WRFv2 also generates some sparse clouds in the middle of the cloudy period, while WRFv1 remains mostly clear. While WRFv2 is an improvement over the original meteorology forecasts, the upcoming pollutant concentration analyses illustrate that even the nudged model meteorology is still not realistic enough to facilitate accurate air quality forecasts during that specific period.

First, a brief comparison of planetary boundary layer (PBL) heights is provided to help illustrate the differences in ventilation between observed and modeled conditions. Figure 2.2, below, shows that, like cloud cover, modeled and observed PBL heights did not significantly diverge until the morning of the 16th. At
that point, observed PBL heights were consistently higher than modeled heights by at least 100m, indicating that actual surface conditions were better ventilated than modeled conditions until after the 22\textsuperscript{nd} when the observed clouds began to dissipate. A comparison of modeled and observed wind speeds is also provided below in Figure 2.3. While there is discrepancy in modeled vs. observed wind throughout the month, it is notable that modeled and observed wind speed during the more “mixed” period of around 1/16 – 1/22 are quite similar. To expand on this point, Figure 2.4 combines wind speeds and PBL heights to create the “ventilation index” within observations and model results. This clearly illustrates the fact that the meteorological observations actually indicate greater ventilation than the model during the cloudy period, when all pollutants were over-predicted. Taken together, this would seem to eliminate wind speed and isolate cloud modeling, and consequently PBL height, as the most influential meteorological model shortcoming for this study.
Figure 2.2: Time series comparison of modeled and observed PBL heights for (a) clear period of YAWNS and (b) cloudy period of YAWNS. Note that ceilometer measurements of PBL heights during the cloudy period were incomplete. Still, it appears that insulation effects of the persistent cloud cover during the latter half of the month limited diurnal variation in PBL heights.
Figure 2.3: Time series of modeled and observed wind speeds for the majority of the YAWNS period. Both modeled wind speed results stay close to observations during the cloudy period, and thus are not likely to cause pollutant concentration biases during this period. A wind rose showing observed directions for distinct wind speed magnitudes for this period is inset.
Figure 2.4: The ventilation index variable is introduced as the product of wind speed and PBL height, displayed as a time series for the cloudy period of YAWNS. The higher observed ventilation indices are reflective of a consistent, deeper average PBL height than simulated in either WRF version. This is a result of persistent cloud cover, which the model failed to simulate.

While ceilometer PBL height measurements were incomplete during the cloudy period, it appears that the surface layer stabilized at a higher elevation than what was modeled by WRF. This is supported by observed temperature and relative humidity time series in Figure 2.5, which both stabilized at mostly constant values during that period with little diurnal variation. Of course, the emergence of a persistent cloud layer is represented by the highest observed relative humidity values of the month, constant around 90%. The lower plot of Figure 2.5 shows that the initial model run, WRFv1, fails to simulate this stabilization and generally under-predicts relative humidity for most of the cloudy period. The nudged WRF model
output, WRFv2, does add some humidity to the cloudy segment, but still less than what is observed. Meanwhile the observed temperature hovered around 270 K. during this cloudy period. This is likely an outcome of the insulator effect of the cloud layer. On the other hand, the modeled temperature, lacking any significant cloud insulation, varied above and below the observed temperatures throughout this period. One of the many important effects of atmospheric moisture is the facilitation of secondary aerosol production, which will be discussed more in the following section.

![Modeled and Observed Temperatures](image)

**Figure 2.5:** Time series of modeled and observed ambient temperature at 2m (a), and corresponding time series of modeled and observed relative humidity at 2m (b).
Together, the results show a failure of both WRF meteorology model runs to generate sufficient water vapor during the second half of January, 2013 in Yakima, which in turn causes a strong divergence between observed and modeled temperature. These discrepancies appear to be an effect of deficient cloud production and, in turn, lead to significant model bias in air quality forecasting during cloudy periods.

2.2 EVALUATION OF AIRPACT-4 POLLUTANT FORECASTING

2.2.1 PRIMARY POLLUTANTS

Beginning in the late morning of January 16th, a rapid dilution of all monitored gas-phase pollutants was observed. This dilution period lasted about one week and corresponded directly with increases in relative humidity and cloud coverage, and a stabilization of temperature and PBL height. Primary particulate species, such as black carbon, also followed this pattern. Together, the measurements suggest that during this “cloudy” period, abrupt increases in ventilation, and wet deposition by cloud droplets, and a lack of tropospheric temperature inversion layers caused all primary pollutant concentrations to quickly drop and to remain low throughout this period. However, this pattern is not simulated by the initial WRFv1-CMAQ run. The failure of WRF to accurately forecast these key condition changes, as shown above, appears to be the primary cause of a significant divergence between observations and AIRPACT-4 model predictions during the cloudy period. The second run utilizing nudged WRF output does a
slightly better job of decreasing aerosol concentrations as conditions change, but as suggested by the discrepancy in cloud cover, the timing does not quite match up. Meanwhile, gas phase pollutants still are not modeled well throughout the period.

The following figures (2.6 – 2.13) illustrate the model evaluation before and during the cloudy period for key individual pollutant species. Each time series compare observed ambient pollutant concentrations from WSU YAWNS measurements to AIRAPCT-4 model results. The “CMAQ V1” designation represents results from the initial AIRAPCT-4 run with WRF meteorology inputs based on the 40m surface layers and forecasted conditions. “CMAQ V2” signals results from the second run with a 20m surface layer and nudged conditions in an attempt to mimic actual conditions more closely.

**Figure 2.6** compares time series of modeled and observed CO and NOx. These plots show that both model runs fail to capture the spikes in CO concentration just before the cloudy period, then over-predict CO during the more ventilated conditions of 1/16 – 1/22. As shown in the lower CO plot, the second run utilizing the refined meteorology does predict slightly lower diel maxima during that period in coordination with slightly improved meteorology assessment. However, as indicated by the cloud cover and ventilation index plots earlier, the improvement is still not great enough to generate statistically accurate results during this period. If there is a positive aspect to the model evaluation for CO, it is that the diel temporal agreement between forecasts and observations appeared to increase with the second model run. This affirms that improving the accuracy of the meteorology
input does indeed have detectable, however weak, positive effects on trace gas modeling.

**Figure 2.6:** Time series of observed ambient carbon monoxide and NOx mixing ratios compared to model results from both forecast and nudged WRF meteorology, throughout the YAWNS period. In both model runs, peak CO and NOx concentrations are under-predicted as they build during clear period in the first half of the month. The model then fails to simulate the rapid dilution of gas-phase pollutants beginning on the 16th.

The evaluation of NOx forecasting in AIRPACT-4 is similar to that of CO. Again, the model misses the concentration spikes during the clear period at the
beginning of the month. During the cloudy period beginning on the 16th, the model begins to agree relatively well with observed magnitudes of NO\textsubscript{x} concentrations. However, this is likely a coincidental result of the general under-prediction of NO\textsubscript{x}. During this more ventilated period, observed concentrations drop drastically, while forecasted concentrations maintain similar patterns of magnitude and diurnal variation. The cloudless model results and observed NO\textsubscript{x} concentrations just happen to converge during that period. Of course, this lack of forecasted dilution in NO\textsubscript{x} is likely due to the same basic problem in maintaining cloud cover in the WRF meteorology simulation. It is again noteworthy that the slight meteorological model improvements in the second model run, which did include an increase in water vapor, induced a drop in peak forecasted NO\textsubscript{x} concentrations during the cloudy period. This highlights an inverse relationship between NO\textsubscript{x} and water vapor in the model that seems to be representative of the in-situ processes. Water vapor is expected to scrub out ambient NO\textsubscript{x} through chemical reactions and physical deposition. Lower NO\textsubscript{x} within combustion emissions has even been observed under the presence of high water vapor mixing ratios (Landman et al. 2006). Of course, the significant under-prediction of NO\textsubscript{x} during the drier clear period of the YAWNS study remains an issue.

At lower temperatures, gas-phase ammonia condenses and reacts to form ammonium nitrate (Curci et al. 2015). Given the high concentration of agricultural land in some areas of the Yakima Valley, an analysis of ammonia gas concentrations for YAWNS is relevant. Figure 2.7 shows time series of modeled and observed ammonia, which despite limited measurements, appear similar to those for other
gas-phase pollutants. Transport from the Lower Yakima Valley, southeast of Yakima, to the more urban area has been a concern, but source attribution in the YAWNS report concluded that emission sources were actually quite different for the two areas. An analysis of wind patterns displayed above in Figure 2.8 also indicates that very little wind came from the Southeast to Yakima during January 2013. Ammonia, and all other ambient pollutants in Yakima, is most likely to be locally generated. Despite the ammonia measurement data being limited mostly to the cloudy period from January 16th to the 24th, Figure 2.7 shows evidence that AIRPACT-4 appeared to drastically under-predict ammonia during the clear conditions before and after the aforementioned cloudy period. Such a discrepancy could limit the model’s ability to produce secondary ammonium nitrate aerosol.

![Figure 2.7: Time series of observed and modeled ammonia concentrations.](image)

Ammonia measurements were limited to the time period displayed here, but they appear to follow the trend of CO and NOx gas-phase concentrations throughout
YAWNS. Likewise, the model fails to predict both peak concentrations during clear periods and the rapid dilution beginning on the 16th.

AIRPACT-4 forecasts of PM$_{2.5}$ also under-predicted peak observed particulate concentrations, though not as badly as the gas-phase pollutants. Alternatively, one of the more interesting aspects of the PM$_{2.5}$ model results, plotted as time series with observations in Figure 2.8, was the great discrepancy between the two model runs. In general, the second CMAQ run, utilizing nudged WRF meteorology output, generated lower PM$_{2.5}$ concentrations throughout the study period. As shown in Figure 2.8, the two model results diverge most conspicuously during the cloudier period, when observed PM$_{2.5}$ concentrations decrease as well. While it is encouraging to see modeled particulate concentrations drop during this period, it is apparent that the modeled dilution phase of PM$_{2.5}$ actually begins and ends sooner than for the observed concentrations. Temporal agreement between observed and modeled values is also very poor during the cloudy period, in contrast with the reasonably accurate diel profile during the earlier clear period. Temporal errors in humidity modeling, which intensify during the cloudy period as shown in the previous section, is likely a major reason for this bias in PM$_{2.5}$ modeling, as for many other pollutant species. Figure 2.9 shows a Q-Q plot pairing ranked PM$_{2.5}$ observations against corresponding AIRPACT-4 model results for both runs during only the clear period, up to the 16th. When compared against the normality line it is clear that the initial model forecasts were more likely to over-predict PM$_{2.5}$. The
WRF nudging tended to decrease AIRPACT-4 PM$_{2.5}$ forecasts significantly, to the point that the model shifted to the conservative side of estimates.

![YAWNS CMAQ Analysis: PM2.5](image)

**Figure 2.8:** Time series of observed surface PM$_{2.5}$ concentrations compared to AIRPACT-4 model results using both forecasted (v1) and nudged (v2) WRF meteorology inputs. Reasonably accurate temporal predictions give way to significant error in both timing and magnitude of PM$_{2.5}$ concentrations during the transition from clear to cloudy conditions. Washington State Department of Ecology data were used for PM$_{2.5}$, as the available WSU AMS measurements represent PM$_{1.0}$.
Figure 2.9: Q-Q plots of Observed PM$_{2.5}$ concentrations vs. AIRPACTv1 (a) and AIRPACTv2 (b). Comparing the distributions to the normality (y=x) line illustrates the shift to a tendency of the model to under-predict with the change to nudged WRF meteorology input. In comparison, the initial AIRPACTv1 PM$_{2.5}$ is relatively normal. The observations here are taken from WA Ecology’s TEOM measurements.

To expand on the YAWNS PM$_{2.5}$ modeling difficulties in AIRPACT-4, Figure 2.10 provides a closer looks at modeled and observed PM$_{2.5}$ concentrations, including both WSU AMS and WA Ecology TEOM measurements, over a shorter time period. The 12th to 16th of January 2013 represents a period of clear conditions and relatively high concentrations of many pollutant species in Yakima. Figure 2.10
shows some sporadic agreement in concentration magnitude, but indicates that AIRPACT-4 is often predicting the peaks and valleys in PM$_{2.5}$ concentrations at opposite times from the observations. The timing of both predicted and observed PM$_{2.5}$ concentration spikes vary between morning and evening. Note that the AMS is actually measuring PM$_{1.0}$, which may explain the discrepancy from the TEOM during the concentration spikes on the 15$^{\text{th}}$. Of course, it can be difficult to pinpoint the cause of such modeling errors. The differences could be rooted in a number of shortcomings including biases in the emissions inventory, WRF meteorological model, and CMAQ chemical transport model.

For the YAWNS scenario, relative humidity has been suspected as a crucial model error affecting secondary PM$_{2.5}$ forecasting. However, throughout the study period, the respective diel patterns of observed and modeled relative humidity correspond quite well. In terms of magnitude, the two model runs tend to over-predict relative humidity during the nighttime hours. It is well established that humidity plays an important role in the formation of PM$_{2.5}$. More specifically, a recent study by Cheng et al. (2015) found that the diel pattern of observed PM$_{2.5}$ in wintertime Beijing directly followed that of relative humidity. Accordingly, the AIRPACT-4 model’s accuracy problems in humidity and cloud forecasting for this specific domain appear to be one of its fundamental limitations in particulate forecasting. For comparison, a closer look at the diel profiles of modeled and observed relative humidity is included in Figure 2.10, for the same shortened period. The strong disagreement between modeled and observed PM$_{2.5}$ using forecast meteorology is likely due in part to the differences in cloud cover between
observations and both model runs. The second version of AIRPACT-4 PM$_{2.5}$ forecasts does appear to smooth out some of the temporal error. However, comparing the **Figure 2.10** (a) and (b) time series, as well as the Q-Q plots of PM$_{2.5}$ evaluation for both model runs, indicate that much of this apparent temporal improvement is largely a result of minimizing the over-prediction errors by decreasing overall PM$_{2.5}$ concentrations in the second (v2) model run. Of course, for much of this period the relative humidity predictions do not actually change much between the forecasted (WRFv1) and nudged (WRFv2) output. This suggests that the significant magnitude shift in modeled PM$_{2.5}$ is likely caused by changes in some other meteorological processes between model runs.
Figure 2.10: Closer comparison of modeled PM$_{2.5}$ with observed PM$_{2.5}$ from the Department of Ecology's TEOM instrument and PM$_{1.0}$ from WSU's AMS from 1/12/13 to 1/16/13 in Yakima, separated by the two model run versions. During this clear period, before observed humidity increased and pollutants rapidly dispersed, the temporal agreement was actually quite strong, although both model runs tended to over-predict relative humidity. This slight difference does not appear to account for the significant error in total PM$_{2.5}$ forecasting.

Evaluation of black carbon (BC) forecasts gave similar results to those of other primary species. Of course, unlike PM$_{2.5}$, black carbon consists entirely of primary particulates. This explains why the AIRPACT-4 forecasts for BC more
closely followed those of the primary gas-phase species; the model consistently under-predicted the highest observed concentration events, and then failed to sufficiently disperse BC during the poorly modeled cloudy period. The nonreactive nature of black carbon provides a clear signal that BC emission sources may be underrepresented in the inventory, thus creating a failure to predict the concentration spikes throughout the month of January. Source attribution has indicated that during the winter in Yakima, residential wood stoves account for over half of BC emissions (VanReken et al., 2014).

Figure 2.11: Times series of observed and AIRPACT-4-predicted black carbon concentrations throughout the YAWNS period in Yakima.
2.2.2 SECONDARY AEROSOL

One of the primary motivating factors for the YAWNS measurement study was the persistent high fraction of nitrate aerosol (NO$_3$) within the elevated wintertime total PM$_{2.5}$ concentrations. Accordingly, the WSU mobile atmospheric chemistry lab included an Aerosol Mass Spectrometer for speciated aerosol measurements. The nitrate and ammonium speciated aerosol predictions, shown in Figures 2.12 and 2.13 respectively, follow the same general pattern as the total PM$_{2.5}$ predictions, displaying a pronounced ventilation period but having it occur a couple days earlier than in the observations. Also, there are still obvious discrepancies in diel variation. Given some of the key discrepancies in modeled and observed meteorological variables, these temporal differences are not surprising. A recent secondary aerosol study by Curci et al (2015) reiterated, “The nitrate formation process is predicted to be largely driven by the relative-humidity vertical profile, which may trigger efficient aqueous nitrate formation when exceeding the ammonium nitrate deliquescence point.” In other words, nitrate aerosols require sufficient atmospheric moisture to form. In this way, inaccuracies in relative humidity forecasting would seem to limit secondary aerosol formation just as it would for total PM$_{2.5}$. Interestingly, previous analysis of relative humidity modeling revealed accurate temporal profiles and a slight tendency of the model to over-predict this term. The forecasted relative humidity term does not appear to be responsible for inaccuracies in AIRPACT-4 PM$_{2.5}$ forecasting.
Observed ANO$_3$ peaks just as the full layer starts to form, then slowly dissipates throughout the cloudy period, while still remaining above concentration levels that characterized most of the first half of the month. The fact that ANO$_3$ remains elevated while all primary pollutants drop provides evidence of secondary aerosol production within the cloud layer. This is further supported by the failure of AIRPACT-4, using the nudged WRF output, to produce sufficient ANO$_3$ during that specific period. The significant differences in ANO$_3$ production between the two model runs are also likely associated closely with the cloud cover variable. While relative humidity values are actually similar between the two WRF versions, a review of cloud cover output in Figure 2.1 shows some key differences in modeled cloud cover. Notably, WRFv1 includes some clouds during the 16$^{th}$ and 17$^{th}$ while WRFv2 does not. This seems to be the reason for higher predicted secondary aerosol concentrations those days by the corresponding AIRPACT-4 run. The apparent correlation between clouds and secondary aerosol would suggest significant production aloft, which will be explored in the upcoming section.
Figure 2.12: Time series of observed ambient aerosol nitrate concentrations alongside AIRPACT-4 results from both model runs segmented roughly into (a) the predominantly clear period and (b) predominantly cloudy period. The sizable differences between the three profiles demonstrate the large sensitivity of ANO₃ formation to changes in meteorology.
Figure 2.13: Observed and modeled ambient ammonium nitrate concentrations for the YAWNS period. Similarly to nitrate results, ANH$_4$ concentrations appear to be strongly connected to the presence of clouds.

The significant error in AIRPACT-4 PM$_{2.5}$, particularly secondary nitrate, forecasting during the YAWNS period does not appear to be rooted in relative humidity (RH) production in WRF. This is contrary to initial expectations, given the large nitrate variances between observations and both model runs, as well the established behavior of nitrate aerosol production to follow RH profiles. However, the modeled and observed relatively ground-level RH values show very little discrepancy. In fact, WRFv2 forecasts slightly higher RH than WRFv1, yet forecasts
consistently lower nitrate aerosol concentrations than WRFv1 during the cloudy period. This suggests that the limitations in AIRPACT-4 nitrate production are rooted elsewhere; likely cloud formation and associated secondary nitrate production. One of the hypotheses of the YAWNS Final Report (VanReken et al. 2014) was that the cloudy period created a shift in nighttime chemistry that enhanced the formation of particulate nitrate by increasing NO$_3$ radical concentrations. AIRPACT-4 evaluation would support this idea, given the drastic differences in cloud cover between the observations and two model runs. It may also be useful to recall the model’s substantial under-prediction of NO$_x$ and ammonia, both emitted precursors to nitrate aerosol formation, during the days leading up to the cloudy period. An under-prediction of these species combined with insufficient clouds in the model appear to be key factors in limiting production of secondary nitrate aerosol in AIRPACT-4 during YAWNS.

2.2.3 VERTICAL PROFILES

When considering ground level concentrations of secondary species such as nitrate aerosol, it can be important to examine the relationship of processes occurring both within and above the PBL. Many studies (e.g. O’Dowd and Smith, 1996) have observed the tendency of aerosols to form layers at or above the top of the mixing layer, especially when clouds are present. Curci et al (2015) recently expanded on this process, finding the residual layer above the PBL to form nitrate aerosol that contributed up to 40% of ground level PM$_{2.5}$ in the morning hours.
While the Curci et al study took place in Summertime Italy, the topography of their Po Valley site is similar to our Yakima Valley measurement site, albeit with a higher population density and more serious air pollution problem. In any case, the large impact of nitrate formation aloft on ground-level PM$_{2.5}$ concentrations observed in recent studies motivates an examination of some pollutant vertical profiles in wintertime Yakima. Vertical measurements of pollutants were not taken in the YAWNS study, but we can inform our understanding by consulting model output for some of these processes.

Vertical profiles of some key species were constructed from the second version of CMAQ results, utilizing the nudged WRF meteorology. As suspected, the most interesting modeled vertical profiles were those of secondary particulates. Past YAWNS analyses have hypothesized that during stagnant conditions, the spikes in observed nitrate during morning hours might be due to nitrate production in the overnight layer that mixes downward as the surface layer deepens in the morning (VanReken et al. 2014). One important discrepancy between the modeled and measured meteorological conditions is that observed clouds were often not replicated in the model, and vice versa. However, it appears that modeled clear days do support the hypothesis of downward particulate mixing. This is likely due in part to the presence of a temperature inversion during the nighttime and morning hours of those clear, stagnant days. As shown below in Figure 2.15, a buildup of ANO$_3$ forms in the early morning hours and mixes downward throughout the day on the 18th. By including the modeled PBL height on the vertical plots, it is apparent that the buildup forms in the early morning and remains right around the PBL through
the morning hours before mixing downward. Meanwhile, a modeled cloudier day, 1/6, did not display significant ANO₃ concentrations in the residual layer, as shown in Figure 2.14. Below this series of plots, Figure 2.16 shows some vertical temperature profiles for 1/06 and 1/18. There is a strong temperature inversion modeled on the 18th, which corresponds to the strongest ANO₃ buildup in the residual layer.

Vertical nitrate profiles were also constructed and examined for the 15th of the month. This day was considered useful for the analysis because it experienced some of the highest nitrate concentrations of the study period. Within the model version used here, it was characterized as cloudy in the morning, shifting to clear, which also added interest. However, the 15th shows a pretty constant vertical nitrate profile below the PBL with no buildup layer. Nitrate profiles on the 15th were similar to those of the 6th, just greater in magnitude. Another conclusion of Curci et al. (2015) was that aerosols should form layers near the top of the mixing layer, especially when stability and clouds increase. Given our combination of modeled vertical profiles, analysis suggests that the model successfully generates secondary aerosol aloft under stable conditions, but is not replicating chemical production in the cloud layers and thus under-predicting all secondary aerosol during cloudy periods, independently of whether the cloud modeling within the model is accurate to begin with.
**Figure 2.14**: Vertical profiles of $\text{ANO}_3$ and water vapor mixing ratio throughout January 6\(^{th}\) during YAWNS. This was modeled as a cloudy day in the nudged WRF (v2) which was used in this forecast. However, no buildup of nitrate aloft is modeled. Modeled PBL heights at each hour are shown as black horizontal lines.
Figure 2.15: Vertical profiles of ANO$_3$ and water vapor mixing ratio throughout January 18$^{th}$ during YAWNS. In WRF v2 meteorology used for this forecast, this was a mostly clear day, yet the model generates a buildup of ANO$_3$ in the morning hours near the PBL, which does correspond to a layer of increased moisture content. Modeled PBL heights are shown as black horizontal lines.
Figure 2.16: Vertical profiles of ambient temperatures modeled by the nudged version of WRF (v2) for 1/6/13 and 1/18/13, to supplement the corresponding nitrate and water vapor profiles. The 6th, which is modeled as being relatively cloudy, shows no temperature inversion, while the 18th, modeled as a clear day, shows a strong inversion near the surface.

Modeled vertical profiles for primary and secondary organic aerosols were also examined, and are shown below in Figure 2.17. No “inverted” concentrations appeared to be modeled for POA. The 11th, displayed below, was the only day of the month that a slight buildup aloft was evident. Of course, this can be explained by the fact that primary aerosols are, by definition, directly emitted near the surface rather than formed chemically. Meanwhile, some slight SOA buildups in the residual layer were modeled mostly on the same days that showed increased aerosol nitrates.
These buildups also appeared to mix downward as morning fell, increasing the surface concentrations. However, total modeled SOA was insignificant compared to POA. One potential cause of this, seemingly common to the modeling of all aerosols within this study, is that the CMAQ model within AIRPACT-4 is failing to simulate aqueous processing within the cloud that could enhance secondary aerosol formation.

Finally, AIRPACT-4 vertical profiles of NO\textsubscript{x} were examined due to its ability to contribute to nitrate production. Figure 2.18 shows vertical profiles of NO\textsubscript{x} on January 6\textsuperscript{th} and 18\textsuperscript{th}. These profiles show no buildup aloft of NO\textsubscript{x} above surface concentrations. On the 6\textsuperscript{th}, which is a relatively cloudy day in the WRFv2 output, NO\textsubscript{x} concentrations are relatively constant vertically up to the PBL. Surface concentrations are also lower on the 6\textsuperscript{th} than the 18\textsuperscript{th}, which is modeled as a clear day.
**Figure 2.17:** Vertical profiles of primary and organic aerosols from AIRPACT-4 v2 on January 11\textsuperscript{th} and 18\textsuperscript{th}, respectively, when some buildups in the residual layer were evident. The 18\textsuperscript{th} was modeled as a mostly clear day with a strong temperature inversion near the surface. However, on cloudy days the model generally failed to produce secondary aerosols.
Figure 2.18: Vertical profiles of NO\textsubscript{x} from version 2 of AIRPACT-4 for YAWNS on (a) the 6\textsuperscript{th} and (b) the 18\textsuperscript{th}. No buildup of NO\textsubscript{x} in the vertical layers is simulated, but the model does show relatively constant concentrations up to the PBL on the cloudier day of the 6\textsuperscript{th}. 
CHAPTER THREE

SOURCE ATTRIBUTION AND EMISSIONS INVENTORY ANALYSIS

Several approaches were taken in attempt to evaluate the Emission Inventory utilized by CMAQ in this study. Emission estimates were provided by the EPA NEI 2008, processed by SMOKE and input to CMAQ along with WRF meteorology to generate air quality forecasts via the AIPRACT-4 modeling system. The emissions evaluation was motivated by concentration modeling biases in various meteorological conditions throughout YAWNS. The major emission source categories in wintertime Yakima are limited to mobile vehicles and residential wood combustion (RWC), which simplified the analysis. However, both categories are known for their difficulty in being compiled accurately.

The first diagnostic experiment in NEI evaluation for YAWNS compared modeled pollutant concentrations to emission rate (moles/s/grid) normalized by the ventilation index of [WS x PBL ht] (m²/s). A relatively linear correlation was expected, as high emissions coupled with low ventilation should theoretically predict high pollutant concentrations. However, very little correlation was observed for CO or NOx in the Yakima 4-km grid cell, as shown below in Figure 3.1. A stronger relationship between the pollutant concentrations and ventilation indices is visible when presented as a time series in Figure 3.2. Given the time series charts, it appears that the reason for the lack of explicit correlation may be simply that the concentration peaks lag behind the high emissions/low ventilation conditions.
Figure 3.1: Comparison of AIRPACT-4 forecasts of (a) CO and (b) NOx concentrations to emission rate normalized by the ventilation index, $U^*PBL$ (m$^2$/s). A relatively linear correlation was expected, but the plots do not show any obvious, significant relationship.

Figure 3.2: Time series plots of modeled concentration vs. [(emissions) / (ventilation index)] for (a) CO and (b) NOx in the YAWNS Yakima 4-km grid cell
show a temporal relationship that was not evident in Figure E1. Peaks in concentrations tend to follow high emissions to ventilation ratios.

The next phase of emissions evaluation was performed by comparing the CO to NOx ratios within both concentrations and emissions throughout the region. All comparisons were made on a molar basis. For these calculations, a molar weight of 46 g/mol was assumed for NOx emissions simply because this was the molar weight assigned to NOx in the MOVES emissions output. As shown below in Figure 3.3, CO/NOx for mixing ratios were estimated by finding the average slope of CO vs. NOx ambient mixing ratios, both for modeled and observed data in Yakima. Only the days 1/05/13 through 1/17/13 were analyzed because all measurement datasets spanned this period and because it does not include the major concentration discrepancy during the cloudy period, which was highly affected by meteorology bias. In the Yakima grid cell, the modeled CO/NOx ratios are much higher than observed with a model ratio of approximately 30 versus 14 for the measured molar CO/NOx. Additional measurements from the Toppenish site to the southeast of Yakima, shown in Figure 3.4, were also examined, and it is apparent the CO/NOx at that location during the same time period is even lower than in the Yakima observations.

This examination followed the procedure used by Wallace et al (2012), where molar CO/NOx ratios were examined within the emissions inventory and observed concentrations during December 2008 and January 2009 in Boise, ID. The total CO/NOx ratio within all emissions in Boise, via the 2008 NEI, was about 22,
which is slightly less than that calculated for Yakima. However, the total observed CO/NOx molar ratio in Boise was 4.6; significantly lower than that observed for Yakima. This suggests that the discrepancy in CO/NOx between the emissions inventory and observations is not necessarily unusual. It also indicates that the Yakima site may be more greatly impacted by RWC and less so by mobile sources, compared to the Boise site, given the much higher ratio of CO/NOx within RWC emissions than in mobile emissions.

Figure 3.3: Observed (a) and modeled (b) CO/NOx concentration ratios in Yakima from 1/05/13 to 1/17/13 on a molar basis. Observed CO/NOx mixing ratios are less than half of those emitted. However, this observed ratio is still much higher than
that determined by Wallace et al. (2012) in wintertime Boise. Together, these discrepancies could indicate a greater influence of wood burning in Yakima than other sites, and/or significant bias in the emissions inventory for this region and period.

**Figure 3.4:** Observed molar CO to NOx mixing ratios at the Toppenish site to the southeast of Yakima. These were about half the values observed in Yakima, and much closer to those observed in Boise by Wallace et al. (2012). Toppenish air quality is affected by a different set of emissions, but still adds context to the discrepancy between modeled and observed air chemistry.

While the precise causes for the differences in ambient CO/NOx between observations and model is not immediately clear, they probably indicate biases in the emissions inventory since both pollutants will be diluted to the same degree by
meteorological conditions. To further investigate this possibility, ratios in the emissions were compared to ambient mixing ratios. Table 3.1, below, displays a summary of CO and NOx emissions for the YAWNS grid cell and as well as the entire Yakima Valley. CO/NOx molar emission ratios are also provided for each location and emission type, to be compared to ambient mixing ratios. In the Yakima Valley wintertime emissions inventory, the major categories are mobile emission sources (MOVES), residential wood combustion (RWC), and some area sources associated with agriculture. The CO/NOx emission ratios for each emission category and site can be compared to the modeled and observed ambient CO/NOx ratios. As the following data will show, mobile emissions typically exhibit a higher CO/NOx ratio than ambient concentrations. Wood smoke is associated with a much higher CO/NOx ratio still. Thus Toppenish, as a community with lower population density and higher agriculture than Yakima, likely is characterized by a lower CO/NOx ratio as a result of a different emissions profile.
Table 3.1: AIRPACT-4 January emissions of CO, NOx, and wood smoke PM$_{2.5}$ attributed to Yakima by the EPA NEI 2008. Mobile emissions generated by MOVES, residential wood combustion (RWC) and total emissions are shown. CO/NOx ratios are included for each emission category, as well as modeled and observed mixing ratios in Yakima during January, 2013.

As shown above, the CO/NOx ratio associated with RWC anywhere is over 100. This is much higher even than the mobile CO/NOx ratio, which is already significantly higher than ambient ratios. This discrepancy between emitted and observed ratios may point to some combination of excessively modeled RWC and
mobile emissions relative to other factors in Yakima, or a failure of the model to
dissipate these species through chemical reactions and deposition mechanisms. Of
course, it is also a possibility that the pollutant ratios within the modeled emissions
are simply incorrect. After all, NOx was grossly under-predicted by AIRPACT-4 for
most periods of this study. One clear pattern is that higher population density
(Yakima) correlates to a higher CO/NOx ratio within ambient concentrations than
for more rural areas (Yakima County as a whole).

To further investigate the impact of RWC on air quality, particularly PM2.5 in
Yakima, simple source attribution methods were performed. A measurement-based
bivariate source attribution study has previously been conducted by Tom Jobson of
the Washington State University YAWNS measurement group. This attribution was
limited to gas-phase species, and used acetonitrile (CH$_3$CN) as a wood smoke tracer
and NOx as a mobile emissions tracer. These species were found to be the strongest
predictors through linear regression. Thus, this experiment assumed the presence
of only these two major sources, and predicted respective contributions to total
concentrations with the following bivariate formula:

\[ X \text{ [ppbv]} = EF_{CH_3CN} \times [CH_3CN] + EF_{NOx} \times [NOx] \]

Where:

- \( X \) = the calculated pollutant concentration
- \( EF_{CH_3CN} \) or \( EF_{NOx} \) = the emission factor of \( X \), relative to acetonitrile or NOx
- \([CH_3CN]\) or \([NOx]\) = mixing ratio time series of acetonitrile or NOx
This bivariate source attribution analysis suggested that over the YAWNS campaign, emissions of CO, benzene, toluene, and black carbon were split pretty evenly between RWC and mobile sources, while formaldehyde and acetaldehyde emissions are dominated by wood smoke. However, Jobson et al. (2014) observed that during the clear period of January, many of the species’ emissions profiles skewed more toward mobile sources.

The ability to deconstruct certain ambient pollutant concentrations into their approximate emission components is one advantage of air quality model results. To compliment Vanderschelden’s measurement-based source attribution, a similar bivariate model was developed from the 2008 NEI used for YAWNS forecast modeling. For this experiment, emission factors for unique source categories were examined within the emissions inventory, then applied to model results or concentrations to estimate source contributions. Differences in attributed concentrations between model results and observations, as well as differences in attributed and NEI emissions, were expected to help identify biases in the emissions inventory. Of course, this method will work best for nonreactive species. Emission factors for the two major sources here, mobile and RWC emissions, were calculated as the molar fraction of particular species emitted within each category, relative to the total modeled emissions of that pollutant. For instance, to establish emission factors PM$_{2.5}$ from RWC and mobile sources:

\[
\begin{align*}
(1) \quad EF_{RWC} &= \frac{[RWC \ PM_{2.5} \ (\text{moles/s})]}{[\text{total NEI CO} \ (\text{moles/s})]} \\
(2) \quad EF_{\text{MOVES}} &= \frac{[\text{MOVES} \ PM_{2.5} \ (\text{moles/s})]}{[\text{total NEI CO} \ (\text{moles/s})]}
\end{align*}
\]
These emission factors were calculated for each hour of each day for the first four days of the month then averaged. Note that modeled emissions were mostly constant throughout the month, so a large sample was not necessary for this calculation. The major sources of most of the pollutants examined in wintertime Yakima were limited to vehicles, and wood smoke, and some area sources. It was previously established in YAWNS that point sources are virtually absent here. The anthropogenic area sources in the Yakima Valley, other than residential wood combustion (RWC), are mostly related to agricultural land use. The modeled emission factors calculated for some locally important pollutants are displayed below as pie charts in Figure 3.5. These factors were then applied to observed and modeled concentrations in an effort to estimate the approximate contributions of specific sources to ambient air quality in wintertime Yakima. The results of this step are displayed graphically below in Figure 3.6. Most species are dominated by either RWC or mobile sources (MOVES), but it is evident that an additional anthropogenic source category of “All Other Anthro” is prominent in NO\textsubscript{X}, NH\textsubscript{3}, and PM\textsubscript{2.5} for this domain. Accordingly, a third emission factor for this category was easily added to the source attribution exercise summarized in Figure 3.6.

\[
(3) \text{EF}_{\text{All,other}} = \frac{[\text{ALL\_OTHER PM}_{2.5}\text{ (moles/s)}]}{[\text{total NEI CO (moles/s)}]}
\]
Figure 3.5: Major source category proportions of NEI 2008 emissions for relevant pollutants to YAWNS during January. These proportions are used for the simple source apportionment model developed here.
Figure 3.6: Time series of graphical application of the modeled emission factors from Figure E5 to the observed YAWNS ambient concentrations of select
pollutants. Each color represents the estimated contribution of a major source type to observed concentrations, relative to the total emissions. A noteworthy assumption of these calculations is the absence of chemical reactions.

The modeled emission factors shown above were compared to the source attribution performed with tracer measurements by Jobson et al. Attribution results for both methods were especially similar for particulates. Mobile and RWC sources dominated PM_{2.5} emissions and therefore attributable concentrations of all primary aerosol species. However, the tracer study attributed significantly more carbon monoxide and nitrous oxides to RWC. This raises the possibility that mis-allocated RWC emissions in Yakima could be partially responsible for the tendency of AIRPACT-4 to under-predict CO and NOx during the winter. In any case, it is clear that residential wood combustion has a large impact on Yakima's wintertime air quality. Given the many difficulties and assumptions in compiling and modeling RWC emissions, there is reasonable motivation to conduct further analysis focused solely on this emission category.

Analysis of the aerosol mass spectrometer results for ambient organic aerosols during the YAWNS campaign, shown below in Figure 3.7, supports the strong influence of biomass burning on both primary and secondary aerosol production in Yakima. During the clear period, part b) of Figure 3.7 indicates that secondary organic aerosol (SOA) accounts for nearly three quarters of the total OA mass. The majority of the SOA consists of biomass burning organic aerosol (BBOA), as shown in Figure 3.7 part c). For these constructions, BBOA and oxidized organic
aerosol (OOA) were used to represent SOA, while the sum of hydrocarbon-like organic aerosols (HOA_traffic and HOA_BBOA) served as the estimate of total POA. The prevalence of BBOA in these measurements represents the strong effects of wood stove emission products on ambient air quality during this period. Furthermore, the time series of primary organic aerosol (POA) and (SOA) shown in Figure 3.7 part a) indicates SOA production continues during the cloudy period at rates similar to those of daytime hours during the clear period. Together, this information confirms the production of SOA at much higher rates than modeled by AIRPACT-4, and the significant impact of wood burning on both primary and secondary aerosols in wintertime Yakima.
Figure 3.7: Aerosol mass spectrometer (AMS) results for organic aerosols as a) a time series including an estimation of all primary and secondary ambient organic aerosols during YAWNS, b) estimations of primary and secondary organic aerosol components of total organic aerosols during only the clear period of YAWNS, and c) a percentage breakdown of all organic aerosol categories observed during the clear period.
CHAPTER FOUR

TREATMENT OF RESIDENTIAL WOOD COMBUSTION EMISSIONS

4.1 RWC EMISSIONS INVENTORY

Elevated PM$_{2.5}$ originally motivated the YAWNS campaign, and the large role of RWC in the generation of this criteria pollutant places it at the forefront of emissions modeling research. RWC emission totals are known to be very difficult to characterize for any region, and this is especially true for cold weather climates. However, analyses of ambient air quality and emission inventories have given evidence that updating wood smoke emissions will improve our understanding of wintertime air quality in the Yakima Valley, and likely the entire Northwestern U.S.

Residential wood combustion emissions in the Northwest are estimated on an annual basis by state agencies and reported at the county level to the EPA. These totals then exist in the national emissions inventories (NEI) as model-ready input. To increase the precision and accuracy of modeling these emissions, the RWC county totals are then allocated to more specific locations within each county using spatial surrogates based on relevant factors such as population density, land type, and census statistics about wood stoves. These factors can be adjusted within the scripting of the emissions model. The temporal allocation of the annual emissions must also be developed on monthly, weekly, and daily scales to reflect the impacts of climate and human behavior. The large number of assumptions inherent to this process, the impossibility of exhaustive surveys, and the unpredictable impacts of
climate on heating needs make this source category an especially difficult one to model with accuracy. According to NEI 2008, Oregon and Washington are the first and second highest wood smoke emitters, respectively. This means that improving the totals and allocations of RWC emissions can potentially have a profoundly positive impact on air quality forecasting in the Northwest.

Figure 4.1, below, shows an example of the current diurnal allocation of RWC emissions applied to daily totals in January in the AIRPACT-4 domain. This temporal profile includes three distinct emission levels and is intended to be most representative of overall residential wood burning patterns throughout a typical winter day. Evening hours are given the largest allocation of RWC emissions, when most residents are expected to be heating their homes for the night. Early morning hours are given the second-highest allocation, and mid-day the lowest, when more residents are expected to be out of their homes. Note that there are also different profiles for weekdays and weekends; with the weekend days assigned slightly more RWC emissions during the day. In AIRPACT-4 these temporal profiles are uniformly assigned to all pollutant species within RWC emissions.
Figure 4.1: Diurnal variation of modeled residential wood combustion emissions of CO, NOx, and PM$_{2.5}$ during January in Yakima. Note the slight differences between the 4$^\text{th}$, a Friday, and the 5$^\text{th}$, a Saturday.

The spatial allocation step weights the RWC emissions toward the correct residential areas within a county and is crucial for informing the model where the potential RWC pollutant hotspots may be. The spatial surrogates combine relevant population, development, census, and/or land use data into an algorithm that is applied to each individual grid cell to determine allocation fractions of their larger county totals. The EPA developed the wood stove spatial surrogate used for the forecasts shown thus far from census data for “population with primary wood heat”
from a year 2000 nationwide survey. One source of error associated with survey sample size is of course the extrapolation of the raw number of residences using woodstoves for primary heat. Another more complicated source of error is the difference in emission composition between various wood burning device types. To demonstrate the scale of these differences, Table 4.1 displays the EPA’s emission factors of some criteria pollutants assigned to each major stove type. This is actually a simplified version, as the preferred RWC process data includes factors for fuel types; notably hardwoods, softwoods, and the forest types from which they most likely came.
Criteria Pollutant Emission Factors for Residential Wood Combustion (tons/d)

<table>
<thead>
<tr>
<th>Device Type</th>
<th>CO (tons/day)</th>
<th>NOx</th>
<th>PM$_{10}$</th>
<th>PM$_{2.5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Woodstoves and Fireplaces, total</td>
<td>80.1</td>
<td>0.9</td>
<td>10.8</td>
<td>10.5</td>
</tr>
<tr>
<td>Fireplaces, general</td>
<td>146.4</td>
<td>2.5</td>
<td>23.0</td>
<td>23.0</td>
</tr>
<tr>
<td>Fireplaces, non-EPA certified</td>
<td>124.8</td>
<td>1.5</td>
<td>16.5</td>
<td>16.5</td>
</tr>
<tr>
<td>Fireplaces, EPA certified; non-catalytic</td>
<td>57.3</td>
<td>0.8</td>
<td>7.9</td>
<td>7.9</td>
</tr>
<tr>
<td>Fireplaces, EPA certified; catalytic</td>
<td>17.7</td>
<td>0.3</td>
<td>3.4</td>
<td>3.4</td>
</tr>
<tr>
<td>Woodstoves, general</td>
<td>416.8</td>
<td>5.3</td>
<td>58.3</td>
<td>57.6</td>
</tr>
<tr>
<td>Woodstoves, catalytic, general</td>
<td>23.0</td>
<td>0.4</td>
<td>4.5</td>
<td>4.5</td>
</tr>
<tr>
<td>Woodstoves, non-catalytic, general</td>
<td>74.7</td>
<td>1.0</td>
<td>10.4</td>
<td>10.4</td>
</tr>
<tr>
<td>Woodstoves, non-catalytic, pellet fired</td>
<td>2.9</td>
<td>0.7</td>
<td>0.6</td>
<td>0.6</td>
</tr>
</tbody>
</table>

Table 4.1: EPA estimations of tons/day emissions of some relevant criteria pollutants associated with different wood burning device types. The “Woodstoves and fireplaces, total” category is assumed when information for specific wood burning device types is not available. Adapted from the EPA NEI 2008.

4.2 RWC TEMPERATURE ADJUSTMENT EXPERIMENT

Residential wood combustion (RWC) sources are largely comprised of households utilizing woodstoves as their primary heat source. In a recent study aimed at improving RWC emissions modeling in the Southeastern U.S., Napelenok et
al. (2014) recognized a strong inverse correlation between minimum observed temperature and RWC tracer. This relationship reflects the need for higher burning rates to heat homes during colder temperatures. Long-term climate data at the state and county levels has been used to inform the seasonal allocations of RWC emission inventories. Archived temperature data is also commonly used to nudge meteorology input while modeling past time periods and pollutant events. However, for real-time air quality forecast models, such as AIRPACT-4, this coupling has yet to be applied on a large scale. Napelenok et al. (2014) concluded that a real-time temperature adjustment to modeled RWC emissions would be one way to improve modeling accuracy for PM$_{2.5}$. However, the inherent complexity in coupling temperature with emissions in a forecast environment kept them from testing this avenue. In the Pacific Northwest, wintertime RWC emissions are more prominent than in the warmer Southeast region and therefore exhibit an even bigger relative impact on air quality. Given the discrepancies encountered in aerosol forecasts for the YAWNS domain, a temperature adjustment algorithm was hypothesized to be one way to improve the RWC emissions inventory and thus wintertime PM$_{2.5}$ forecasts in the AIRPACT-4 domain.

AIRPACT-4 applies seasonal, daily, and hourly temporal adjustments to reflect the impacts of general climate and human activity on woodstove usage, as shown earlier. However the effects of daily and hourly temperature swings have not been accounted for in previous model runs. To investigate whether minimum temperature did indeed appear to affect January RWC emissions in Yakima, daily maximum observed organic aerosol concentrations were plotted against daily
minimum observed temperature, as shown below in Figure 4.2. Recall that POA emissions were dominated by RWC in the inventory, and thus identified as a strong tracer of wood burning in wintertime Yakima. Observations used in Figure 4.2 were taken only from the clear period of January, 2013, when the complicated effects of cloud chemistry appeared to be minimal.

**Figure 4.2:** Plot of daily maximum observed organic aerosols with daily minimum observed temperature in Yakima. The small sample size is limited to clear days in January 2013 to minimize cloud effects, but the data does indicate a negative correlation between particulate concentration and temperature, which may reflect the impact of increased residential woodstove use during colder temperatures.

As anticipated, Figure 4.2, indicates a correlation between minimum daily temperature and daily maximum observed OA concentrations. Because the primary source of organic aerosols in wintertime Yakima is RWC, this relationship provided
further motivation to test a temperature-adjustment algorithm for wood stove emissions in the AIRPACT-4 modeling framework. Until recently, RWC emissions were processed in AIRPACT-4 in the units of tons/day, allocated temporally from the county totals of tons/year in which they are reported. To accommodate a temperature factor, RWC emission needed to be converted to the units of tons per heating-degree-day (tons/HDD). Heating degree-days represent a measure of the amount of heating necessary for a particular day, relative to a designated threshold temperature under which woods stoves are assumed to be used for the purpose of residential heat. For the application of hourly temperature forecasts, RWC emissions were further divided into tons per heating-degree-hour (HDH).

The wood stove forecast temperature adjustment was then applied to the AIRPACT-4 RWC emissions inventory in a series of steps. First, wood smoke emissions, in tons/heating-degree-hour, were flattened to a constant baseline value over the entire year, expressed hourly. Then the adjusted hourly wood smoke emissions in each cell were calculated using the day-of-week and hour-of-day factors. In this case, the monthly factor was dropped in order to avoid redundantly applying a temperature-related adjustment. The final factor applied to the wood smoke emissions was the hourly heating-degree term, using the forecast 2-m hourly temperature from WRF, in degrees Fahrenheit, relative to a maximum of 50 °F, with a minimum of 20 °F. The result was a new RWC emissions profile, unique for each hour of the day. The general algorithm for this process is shown below in Equation (4).
(4) \( \text{Emis}(i,j,1) = \frac{\text{Tons}}{\text{HDH}(i,j,1)} \times \text{HD\_Hour}(i,j) \times \text{DayOfWk\_Fctr} \times \text{Hr\_Fctr} \)

The DayofWk\_Fctr and Hr\_Fctr represent the original weekday or weekend and diurnal profile factors, respectively. The HD\_Hr term is new and calculated hourly as the difference between 50 °F and the current temperature in °F, as follows:

(5) \( \text{HD\_Hour °F} = \max(50 - \text{TEMP2}(i,j)) \)

For the January period studied, the new RWC emissions tended to scale upward with the temperature adjustment, above previous emissions. A comparison of the initial and \( T \)-adjusted emission profiles for wood smoke PM\(_{2.5} \) tracer (WSPM2.5) is shown below in Figure 4.3 for early January, 2013. Below that in Figure 4.4, the resulting PM\(_{2.5} \) concentrations considering both original and \( T \)-adjusted wood smoke emissions are compared against observations for the month of January in Yakima.
Figure 4.3: Comparison of original and temperature-adjusted emissions of the wood smoke tracer WSPM2.5 for 1/05/13 - 1/15/13 in Yakima. For this period, the temperature adjustment algorithm clearly tended to increase RWC emissions.
**Figure 4.4:** AIRPACT-4-modeled PM2.5 concentrations in Yakima derived from both non-adjusted and temperature-adjusted residential wood stove emissions are plotted along with PM2.5 observations. While there are some significant differences in PM2.5 forecasting between the two versions, there does not seem to be an obvious, immediate increase in accuracy between with the temperature-adjustment method.

Comparing the original AIRPACT-4 and woodstove-temperature-adjusted PM2.5 forecasts against the observations does not yield an obvious improvement in overall PM2.5 modeling. There are some significant differences between the two time series shown in **Figure 4.4**, which assures that the woodstove temperature adjustment does indeed exhibit a strong impact on PM2.5 forecasting for this domain.
However, the new time series does not appear to be consistently more or less accurate than the initial one. For a more statistical evaluation, Figure 5.5 shows plots of observed PM$_{2.5}$ against AIRPACT-4 cases for both the original and woodstove-temperature adjustment cases. It is now obvious that the woodstove temperature adjustment does not generate an overall improvement in PM$_{2.5}$ forecasting for January 2013 in Yakima. In fact, the data has a slightly better $r^2$ trend line fit for the original case. This method does not appear to be an effective way to improve wintertime particulate modeling in the Yakima Valley of Washington. However, now that the woodstove temperature adjustment is configured for AIRPACT-4, it will be tested for a variety of sites throughout the AIRPACT-4 domain with the intention of improving understanding of PM$_{2.5}$ forecasting bias in the Northwest.
Figure 4.5: Plots of observed PM$_{2.5}$ (WA Ecology TEOM) vs. AIRPACT-4 v1 PM$_{2.5}$ forecasts for the original case (top) and the woodstove temperature adjustment (T-Adj) case (bottom). The T-Adj case does not lead to an improvement in PM$_{2.5}$ forecasting in Yakima, and in fact the data has a slightly better fit for the original case.

The integration of the temperature adjustment to woodstove emissions in AIRPACT-4, described above, failed to generate an improvement in PM$_{2.5}$ forecasting for the YAWNS domain and time period. However, this method can certainly be applied throughout the AIRPACT-4 domain with the potential to provide insight about PM$_{2.5}$ modeling biases on a larger scale. The next phase of this experiment will
briefly examine the impact the same woodstove temperature adjustment
mechanism at a variety of sites in the Northwest. Additional analysis sites were
selected on several criteria. Only sites designated as U.S. EPA AIRDATA locations
with extensive, available PM$_{2.5}$ measurements over the last few years were
considered. Sites were also selected in attempt to span a variety of locations,
population values, and development levels.

Extractions of PM$_{2.5}$ forecast data before and after the woodstove
temperature adjustment were analyzed against observations for 20 different sites
within the AIRPACT domain. Table 4.2 summarizes these results for 12 of the
analyzed sites in terms of mean fractional bias for the original and temperature-
adjustment cases. Sites are listed in order of descending mean fractional bias from
the temperature adjustment runs. The final column of Table 4.2 presents the
reduction in PM$_{2.5}$ forecasting bias caused by the woodstove temperature
adjustment method. Measurements and model results for the entire months of
January 2013 and 2014 were examined. A negative value here signals an increase in
bias with the application of the adjustment. A brief examination shows that the
effects of this experiment varied greatly across the AIRPACT domain.
Table 4.2: Mean fractional bias for standard AIRPACT-4 and temperature-adjusted wood stove emission runs, at 12 of the 20 selected sites in the domain. These statistics consider the entire months of January in 2013 and 2014, and sites are listed in order of descending mean fractional bias.

Although there appears to be little consistency in the effects of the temperature adjustment throughout the domain, there is at least one evident trend...
that emerges. At the predominantly urban sites, AIRPACT-4 tended to generally over-predict PM$_{2.5}$. In most cases, this bias increased with the application of the temperature adjustment. On the other hand, AIRPACT-4 generally under-predicted PM$_{2.5}$ at the more rural, sparsely populated sites and agricultural communities. In some cases, the temperature adjustment diminished some of the bias, but usually the improvement was not significant. There is concern that some of the more rural sites with high observed PM$_{2.5}$, such as Oakridge, Pinehurst, and Darrington, may be displaying high measurements due to terrain effects or because they are located very close to residences with wood heating. Still, there is strong evidence that the presented woodstove temperature adjustment, by itself, is not an effective method to improve AIRPACT-4 PM$_{2.5}$ forecasting in the Northwestern U.S. However, the experiment has highlighted an apparent fundamental emission bias concerning wintertime PM$_{2.5}$ emissions, and woodstoves in particular. The correlation between urbanization levels and PM$_{2.5}$ bias associated with the woodstove temperature adjustment suggests that manipulating the RWC spatial surrogate, particularly to be weighted less by population density, may be a better way to improve RWC emissions and consequently wintertime PM$_{2.5}$ forecasting in this domain.

4.3 SPATIAL SURROGATES FOR RWC IN AIRPACT

In coordination with the development of the 2014 NEI, a new method of spatial allocation for residential wood combustion emissions is under development.
The spatial allocation algorithms, or spatial surrogates, are created on the basis of specific residential, population land-use, and/or census data. They are intended for use with updated county-level emissions totals. The function of the spatial surrogate is to allocate the total RWC emissions more precisely to areas within each county, in order to more accurately drive the air quality forecast model. These emissions updates are one component of the larger renovation of the entire forecast model, from AIRPACT-4 to AIRPACT-5, which is currently underway.

The RWC temperature-adjustment experiment highlighted emissions biases within the former spatial surrogate for wood stoves in the Pacific Northwest. AIRPACT-4 tended to over-allocate wood smoke emissions to urban areas, resulting in a consistent over-bias of PM$_{2.5}$ forecasts during the winter. Conversely, wintertime PM$_{2.5}$ tended to be under-predicted in more rural and agricultural areas throughout the domain. This trend has now informed the development of the new wood stove spatial surrogate for AIRPACT-5. In fact, multiple versions of the new RWC spatial surrogate were developed for testing purposes, with the goal of identifying the spatial algorithm most representative of wood stove usage throughout the entire AIRPACT domain. The previous RWC spatial surrogate, assigned the identifier 'USA 3001,' represents census data for households with woodstoves as primary heat sources, applied to the population centers in each county. Each prospective version of the new surrogate, assigned 'USA 165,' equally combines two land-use and/or population factors associated with residential wood stove usage. For instance, the first version consists of an equal split of low intensity development and residential wood heating census data to estimate a spatial
distribution of RWC activities throughout the domain. Table 4.3, below, summarizes all versions of the RWC surrogate tested for this experiment.

<table>
<thead>
<tr>
<th>Wood Stove Spatial Surrogate</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>USA 3001 (AIRPACT-4)</td>
<td>Residential Wood Heating Population</td>
</tr>
<tr>
<td>USA 165 version 1</td>
<td>Low Intensity Development + Res. Wood Heating</td>
</tr>
<tr>
<td>USA 165 version 2</td>
<td>Forest Coverage + Rural Population</td>
</tr>
<tr>
<td>USA 165 version 5</td>
<td>Forest Coverage + Low Intensity Development</td>
</tr>
<tr>
<td>USA 527</td>
<td>Single Family Residential</td>
</tr>
<tr>
<td>USA 165 x 527</td>
<td>USA 165v1 + USA 527</td>
</tr>
<tr>
<td>USA 165 x 300</td>
<td>USA 165v1 + Population</td>
</tr>
</tbody>
</table>

**Table 4.3**: List of the test versions of the new residential wood combustion spatial surrogate to be applied to the NEI 2014 emissions in AIRPACT-5. Each surrogate draws on a different combination of land-use and/or population data in an attempt to find the most representative predictors of residential wood stove usage in the Northwest. The ‘3001’ surrogate was used with the previous version of the forecast model, AIRPACT-4.

The different versions of the wood stove surrogate listed above in Table 4.3 were each applied individually to the NEI 2008 tons-per-year RWC emissions and tested by running AIRPACT-4 and comparing resultant, total wintertime PM$_{2.5}$ forecasts at a variety of sites throughout the domain. All other emissions were kept constant for each test. Evaluation sites were chosen from those with available EPA
AIRDATA PM$_{2.5}$ measurements for January of 2013 and 2014. Priority was given to sites with consistently high wintertime PM$_{2.5}$ concentrations. However, some smaller sites with very high PM$_{2.5}$, including Pinehurst, Idaho and Oakridge, Oregon, were discarded due to the inability of the model’s 4km modeled grid cells to accurately resolve the fine terrain effects and small population centers. Ultimately twenty-one sites were selected for analysis, representing a spectrum of population and land characteristics.

For each set of results corresponding to different RWC spatial surrogate versions, hourly PM$_{2.5}$ forecasts were extracted from each site, averaged daily, and evaluated against AIRDATA daily PM$_{2.5}$ observations. Table 4.4, below, summarizes the results of these analyses in terms of mean fractional bias (MFB) and mean fractional error (MFE). The RWC surrogate that performed the best is identified for each site and compared to the performance of the former RWC surrogate, USA 3001. Sites are grouped by their best-performing new RWC surrogate. The sensitivity of total PM$_{2.5}$ forecast to changing RWC surrogates is also characterized from low to high. The low-sensitivity sites did not experience significant changes in PM$_{2.5}$ forecasts when different versions of the RWC spatial surrogate were applied, and thus should not be factored as heavily when ultimately selecting the best RWC surrogate. Finally, the columns are shaded more darkly for sites where the proposed new RWC surrogate appeared to improve overall PM$_{2.5}$ forecasting in January.
Table 4.4: Summary of results from the testing of seven different residential wood combustion spatial surrogates at sites throughout the AIRPACT domain. Sites are grouped by their best-performing RWC surrogate. Mean fractional bias (MFB) and mean fractional error (MFE) of total PM$_{2.5}$ forecasts using each surrogate were calculated from daily average observations in January of 2013 and 2014. Observations were provided by EPA’s AIRDATA monitoring network. The sensitivity of PM$_{2.5}$ forecasts to changing RWC surrogates was also classified from low to high. Different RWC surrogates performed best at different sites, and thus the best approach to spatially allocating RWC emissions in the Northwest may be a blend of surrogates that depends on land type. The more darkly shaded columns signal sites
where the proposed new RWC surrogate tended to improve overall PM$_{2.5}$ forecasting in January.

The RWC surrogate test experiment suggested that a variety of new RWC surrogates performed best for different sites throughout the AIRPACT domain. While this complicates RWC emissions allocation, there does appear to be some useful trends. The second version of the new ‘USA 165’ surrogate (165v2) significantly improved PM$_{2.5}$ forecasts at urban sites including Seattle-Beacon Hill, Portland-Lafayette St, and Spokane. In general, this surrogate, combining the variables of “forest cover” and “rural population,” tended to reduce the tendency of AIRPACT to over-predict wintertime PM$_{2.5}$ in densely populated areas, where primary wood stove usage is not as common as in rural or suburban areas. The first version of the new surrogate ‘165v1,’ combining “low intensity development” and “residential wood heating” census data, seemed to perform best for the next class of semi-urban areas such as Eugene, Bellingham, and Coeur d’Alene (Lancaster). It is also noteworthy that all of these urban or semi-urban sites examined experienced an improvement in AIRPACT-4 PM$_{2.5}$ bias in January given the new RWC surrogates.

A host of sites generally characterized as more rural, or in many cases agricultural, were best characterized with the ‘USA 165x300’ RWC surrogate – a combination of low intensity development, RWC census data, and total population. In these cases, the population term tended to increase RWC emissions in the population centers of these counties, where AIRPACT-4 has typically under-predicted wintertime PM$_{2.5}$. However, the former RWC surrogate, ‘USA 3001,’ still
generated less PM$_{2.5}$ bias for most of these sites, with the exception of Kennewick – notably the most populated of this group. In other words, the census data for population with primary wood heat still appears to represent the more rural areas relatively well.

Yakima, fittingly, was an outlier among the sites examined. The ‘USA 527’ surrogate, representing singly family residences, was the only new version tested that did not significantly under-allocate RWC emissions to Yakima. However, the old ‘USA 3001’ surrogate for RWC census data performed very comparably. Like many of the sites in Eastern Washington and Eastern Oregon, the low proximity to forests renders the forest-coverage surrogates ineffective here. It is apparent that proximity to forests does not dictate wood burning habits. On the other hand, the forestry term does help to spread RWC emissions out to the suburbs around the more urban sites like Seattle and Portland along the I-5 corridor. Considering the diversity in factors determining RWC emissions across the AIRPACT domain, the most representative spatial surrogate for residential wood stoves will likely need to be a composite of at least a couple different algorithms specific to distinct regions.
CHAPTER FIVE

CONCLUSIONS AND RECOMMENDATIONS

5.1 SUMMARY

Evaluation of the AIRPACT-4 air quality forecasting system against the YAWNS measurements (VanReken et al., 2014) highlighted some significant model discrepancies in the wintertime Yakima Valley of Washington State. Most notably, the model failed to simulate the pollutant concentration changes corresponding to an observed meteorological shift from clear to cloudy skies. Persistent temperature inversions near the surface led to a buildup of high pollutant concentrations by the end of the extended clear period. As the clouds moved in, all primary gas and particulate species rapidly diluted from relatively high to very low measured concentrations as mixing height likely increased to the top of the cloud layer. AIRPACT-4 did not forecast this drop in primary pollutant concentrations. The fundamental limitation in this case appears to be a failure of the meteorological model, WRF version 3.4.1, to generate and maintain cloud cover, which caused over-prediction of primary pollutants during cloudy periods in wintertime Yakima.

Meanwhile, a more complicated pattern emerged for secondary aerosol modeling in Yakima. Measurements of nitrate aerosol (ANO₃), which initially motivated the YAWNS measurement campaign, remained elevated through the cloudy period. AIRPACT-4 also struggled to simulate this secondary production, which may be largely tied to insufficient moisture within the model. Interestingly, a second model run utilizing nudged meteorological conditions generated even less
ANO₃ near the surface, despite marginal improvements in cloud production. A comparison of vertical profiles shows that on clear days, AIRPACT-4 simulates a buildup of ANO₃ in the overnight residual layer and consequent down-mixing in the morning hours, as expected. However the model shows no secondary aerosol production aloft on modeled cloudy days. It is suspected that, on site, emergent cloud layers enhance secondary aerosol production through hydrolysis and partitioning of gas-phase nitric acid. For the 4km Yakima Valley grid cell, AIRPACT-4 did not simulate a correspondence between cloud and secondary aerosol production, which appears to be another significant shortcoming for wintertime particulate modeling.

In further efforts to investigate AIRPACT-4’s more subtle pollutant concentration biases given satisfactory meteorological agreement, simple source attribution was performed on both model results and YAWNS measurements. A previous attribution study, using NOₓ as a mobile source tracer and acetonitrile to trace residential wood combustion, confirmed that these two sources contribute the vast majority of criteria pollutants to the region. For comparison, a similar bivariate source attribution experiment was performed using the modeled emissions and concentrations, and generally agreed with the partitioning of the emissions inventory. However a comparison of modeled-to-observed ambient CO/NOₓ ratios revealed significant differences within the Yakima grid cell. Ambient CO/NOₓ was much higher in the model than observed, which was believed to represent potential bias in the emissions inventory and motivated further emissions analysis. Additionally, aerosol mass spectrometer measurements indicated that biomass
burning likely accounted for well over half of primary and secondary organic aerosol mass during YAWNS.

The next phase of the wintertime AIRPACT-4 evaluation narrowed the focus of the emissions analysis to residential wood combustion (RWC), which is generally recognized as one of the more biased inventories due to numerous uncertainties in the compilation process. Source attribution had indicated that RWC is responsible for over half of wintertime PM$_{2.5}$ in the Yakima Valley, which is often near attainment levels for PM$_{2.5}$ during the winter. In an attempt to improve PM$_{2.5}$ modeling and further investigate the impact of RWC emissions on air quality in the Pacific Northwest, an experimental temperature adjustment mechanism was applied to RWC emissions in AIRPACT-4 forecast mode. In AIRPACT-4, RWC emissions are assigned temporal profiles to reflect diurnal, weekly, and monthly burning activity. At this point the scope was also expanded to the full AIRPACT domain, covering the entirety of Washington, Oregon, and Idaho. This experiment was motivated by higher observed PM$_{2.5}$ trends in colder weather, and similar proposals from some recent studies including Napelenok et al. (2014). It was proven that wintertime PM$_{2.5}$ forecasts in the Pacific Northwest are indeed very sensitive to changes in RWC emissions. However, the wood stove temperature adjustment algorithm, as outlined earlier, failed to generate any consistent improvement in PM$_{2.5}$ forecasting agreement between the model and observations. Instead, the experimental analysis highlighted the more fundamental limitation of RWC spatial allocation within states and counties.
The final phase of this study investigated the impacts of alternate RWC spatial allocation techniques on total PM$_{2.5}$ forecasts throughout the Pacific Northwest. In this region, modeled area sources such as RWC emissions are reported as county totals, then divided and placed more precisely within counties using spatial surrogate allocations. In all iterations of AIRPACT up to this point, RWC emissions were allocated spatially using a single spatial surrogate algorithm for the entire domain, based on population centers and census data for wood stove usage. Examining the relationships between RWC emissions and total PM$_{2.5}$ forecasts showed consistent biases related to land categories of each site examined. For instance, the emissions model has tended to over-allocate RWC emissions to urban areas and under-allocate RWC emissions to more rural and agricultural grid cells, leading to consistent wintertime PM$_{2.5}$ wintertime forecast biases in the Pacific Northwest. Accordingly, several alternate spatial surrogates for RWC emissions were developed and tested at sites throughout the domain. Applying new methods of spatial allocation to RWC emissions yielded significant improvements in wintertime PM$_{2.5}$ forecasts for certain regions, but different sites responded best to distinct spatial surrogate algorithms. The next step toward improving RWC emissions, and consequently wintertime PM$_{2.5}$ forecasting, in the Pacific Northwest may be a more complex, composite spatial surrogate for RWC emissions, as outlined in the next section.
5.2 RECOMMENDATIONS

The AIRPACT-4 evaluation for the YAWNS measurement campaign in Yakima showed that the meteorological model, WRF v 3.4.1, failed to generate and maintain sufficient cloud cover on many days when clouds were observed. This greatly limited the accuracy of all air quality forecasts for the duration of a prolonged change from clear to cloudy conditions. It could be useful to investigate whether a correlation of cloud modeling error to PM$_{2.5}$ modeling error has occurred in other intermountain basins during the winter, when ventilation is low. If so, the implantation of an updated nudging technique has the potential to improve the influential variable of cloud cover. One such example is given by Biazar et al. (2007), who significantly improved the evolution and partitioning of particulate matter in a CMAQ case study covering the continental U.S. by importing satellite-observed cloud characteristics into WRF. Of course, integrating such nudging methods into AIRPACT in forecast mode is not really feasible. One option might be a nowcast framework, where nudging with observed clouds could be applied on a frequent basis and used as a starting place for one or more forecast hours into the future until the next nowcast update.

The extensive examination of residential wood combustion (RWC) emissions treatment in AIRPACT-4 found a temperature-adjustment of wood stove emissions to be a largely ineffective way to improve PM$_{2.5}$ forecasting in the Pacific Northwest. The experiment found a much more fundamental limitation of accuracy in the spatial allocation methods for RWC emissions within counties. A variety of new
RWC spatial surrogates were tested for their impact on overall PM$_{2.5}$ forecasting accuracy in January. The experiment concluded that different sites in the AIRPACT domain are best characterized by different RWC spatial allocation methods. Sites with similar land-use and population levels tended to be best represented by the same RWC surrogate.

Overall, the spatial surrogate testing suggests that a composite approach to allocating RWC emissions throughout the domain, where distinct regions utilize unique surrogates, will likely be the best way to improve the accuracy of RWC spatial allocation in the next version of AIRPACT. Of the six new versions of RWC surrogates tested throughout the domain, the surrogate representing an even split of “forest cover” and “rural population” provided the biggest improvement in winter PM$_{2.5}$ forecasts for urban sites. The surrogate combining “low intensity development” with census data for “population with primary wood heating” performed slightly better for semi-urban areas. Meanwhile, combining the latter surrogate evenly with a “population” term performed significantly better for winter PM$_{2.5}$ forecasts in rural and agricultural regions. However, for these sites, the new surrogates did not typically improve upon the previous RWC surrogate, which is simply the “population with primary wood heat” method. In fact, an update of the 15-year-old wood stove census data for the Pacific Northwest may prove to be the best way to improve RWC modeling throughout the region. In the absence of updated census data, the primary suggestion for improving RWC emissions treatment in AIRPACT-5 is to consider the adoption of the new spatial surrogates described here for the larger population centers of the Pacific Northwest.
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