**\*(PNWIS)\* Evaluating the AIRPACT-4 Air Quality Modeling Framework for Wintertime Conditions in the Yakima Valley**

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Abstract (PNWIS)

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 PM2.5 levels in the region.

High levels of aerosol nitrate in particular characterize wintertime air quality in the Yakima Valley. The measurements during YAWNS also generated an extensive data set that could be used to help 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, where it appears low clouds promote significant vertical mixing. In this paper, 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 different concentration patterns that occur in clear sky vs. cloudy conditions.

This work also includes evaluation of the model in terms of source attribution between mobile and residential woodstove sources. Observations suggest that elevated PM2.5 levels in Yakima can be attributed to a mixture of contributions from these sources. In this paper, we evaluate the modeling results to determine whether the observed source contributions are matched by the model results.

**Introduction**

The United States Environmental Protection Agency has long established that consistent, high concentrations of airborne fine particulate matter (PM2.5) 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 PM2.5 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/m3 (U.S. EPA, 2013b). Furthermore, the EPA’s most recent National Emissions Inventory (NEI 2014) subdivides PM2.5 into dozens of chemical species from thousands of sources. The highest PM2.5 concentrations are often formed during wintertime stagnation conditions. PM2.5 tends to peak when wintertime reductions in solar heating contribute to a lower 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 PM2.5 concentrations.

The Yakima region of Washington State exemplifies this type of complex terrain. The population base 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. In recent years, high PM2.5 levels have been measured in Yakima that have approached non-attainment values at times during the winter. Within this trend, the Washington State Department of Ecology observed an irregularly high fraction of nitrate PM2.5 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 PM2.5 in Yakima. High-resolution gas-phase and meteorological measurements were also made.

**Goal and Objectives**

The overall goal for this work is to evaluate the AIRPACT-4 air quality modeling framework for wintertime stagnant conditions in the Northwestern U.S. and to identify areas 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 YAWNS campaign, and the lack of published evaluations for the current AIRPACT configuration (Brian, is this true? i.e. I had trouble finding recent AIRPACT papers). The specific objectives include 1) to evaluate AIRPACT-4 for the YAWNS observational period, including clear and cloudy stagnation episodes in the observation period; 2) to assess the role of residential wood combustion 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.

**Literature Review**

The biggest source of PM2.5 emissions in wintertime Yakima is biomass burning – which is dominated by residential wood combustion for heating purposes. Thus, it is significant to note that carbonaceous PM2.5 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 2013) subdivides PM2.5 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, residential wood combustion (RWC) emissions are reported by the local clean air agencies as estimated county totals in tons per year. These estimations are based on a combination of RWC usage surveys and census data on use of RWC as a primary source of household heat. The county totals are then distributed spatially by state agencies 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. In addition, combustion conditions, such as wood moisture content, strongly influence smoke content but are difficult to quantify on a large scale. Strushka (1993)determined that bad, 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. Additional RWC emissions inventory lapses include the burning of different wood types than those for which the stoves are certified, and unreliable testing methods.

As typified by Yakima, RWC, is often a major contributor to carbonaceous PM2.5 in non-attainment areas. Many studies throughout 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 irrespirable particles emitted in January to RWC (Cooper, 1980). In the warmer climate of San Jose, CA, RWC contribution to wintertime PM10 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. Previously, 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 PM2.5 forecasting for the Southeastern U.S. region by adjusting the temporal application of the RWC inventory accordingly. It is implied 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, but also pointed to some additional ambiguities 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, 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.

Perhaps a paragraph or two on PM2.5 modeling (how is PM2.5 treated in state-of-science models, CMAQ and CAMx case studies, info on typical performance levels, etc) followed by a paragraph or two describing the AIRPACT system with reference to past AIRPACT papers.

In state-of-science air quality models, such as the Community Mulitscale Air Quality model (CMAQ) and the Comprehensive Air Quality Model with Extentions (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 diffusion, molecular diffusion, emissions of all precursors, deposition, sedimentation, nucleation, washout, and coagulation. Of course, to create rates of emission and atmospheric diffusion, the Chemical Transport Model (CTM) requires input from an emissions processor, meteorological model, 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 gridded map fit over GIS 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 PM10 and PM2.5. CAMx, on the other hand, only fits aerosols into size bins of PM2.5 and PM10. In addition, CMAQ and CAMx have different modules for inorganic and organic aerosols, sea salt and wet and dry deposition. However they are configured to utilize the same options of emissions processors and meteorology models.

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. 2008). The Air Quality Model Evaluation International Initiative (AQMEII) has 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 for this YAWNS evaluation, showed a consistent overestimation of PM2.5 in the winter months of North America for 2006 data, with a normalized mean bias (NMB) of 30.4% (Appel et al., 2012). The largest overestimations occurred mostly in the Western U.S. On the other hand, CMAQ underestimated summer PM2.5 with a NMB of -4.6%. The authors attribute the overestimations of winter PM2.5 mostly to overestimation of unspeciated PM2.5 mass, as well as some smaller overestimations of elemental carbon (EC) and organic carbon (OC). The next version of CMAQ addressed this issue by including the speciation of trace metals, allowing for better comparison of model estimates to observations. However, the next iteration of the AQMEII evaluation, for CMAQ v5.0.1, showed that wintertime PM2.5 bias in North America actually increased to a NMB of 59.8% 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 consist of the Puget Sound Clean Air Agency, Washington State Department of Ecology, University of Washington Atmospheric Sciences Department, U.S. EPA Region 10 (Seattle), 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 Sate-NCAR Mesoscale Model (MM5) to drive the California Meteorological Model (CALMET)/California Photochemistry Grid Model (CALGRID) Eulerian photochemical modeling suite (Vaughan et al., 2004). It’s ability to provide applicable modeling feedback for the Puget Sound region motivated an expansion of the domain to include all of Oregon and Idaho. The next version, AIRPACT-3, was also 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 generated a 17% NMB in PM2.5 forecasting, compared to a 32% NMB at the more rural observation sites of the IMPROVE network (Chen et al. 2008). 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 at 4-km horizontal grid cell spacing.

**YAWNS Observations**

The Yakima Air Wintertime Nitrate Study (YAWNS, VanReken et al., 2014) provided both the motivation for this study and framework for model evaluation. YAWNS began as a comprehensive air quality measurement campaign motivated by elevated PM2.5 observations in the region and a higher nitrate PM2.5 fraction than the rest of Washington State. Accordingly, Washington State University deployed its Mobile Atmospheric Chemistry Lab from January 5th to 26th, 2013, in Yakima to take high-resolution measurements of speciated PM2.5 and volatile organic compounds (VOCs), as well as NOx, CO, CO2, and meteorological measurements.

Key results from 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 16th, conditions changed to cloudy but bu 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 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. However burn bans usually result in a more gradual reduction in pollutant levels than what was observed in this episode. 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 a the presence of secondary chemical production effects associated with the clouds.

(concentration pics? – give credit)

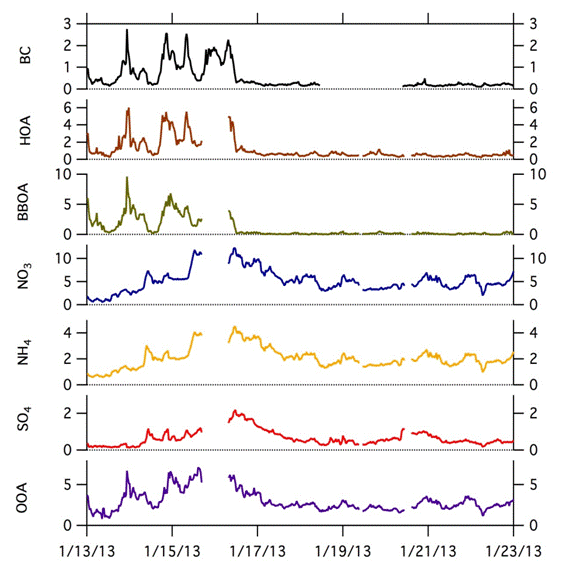
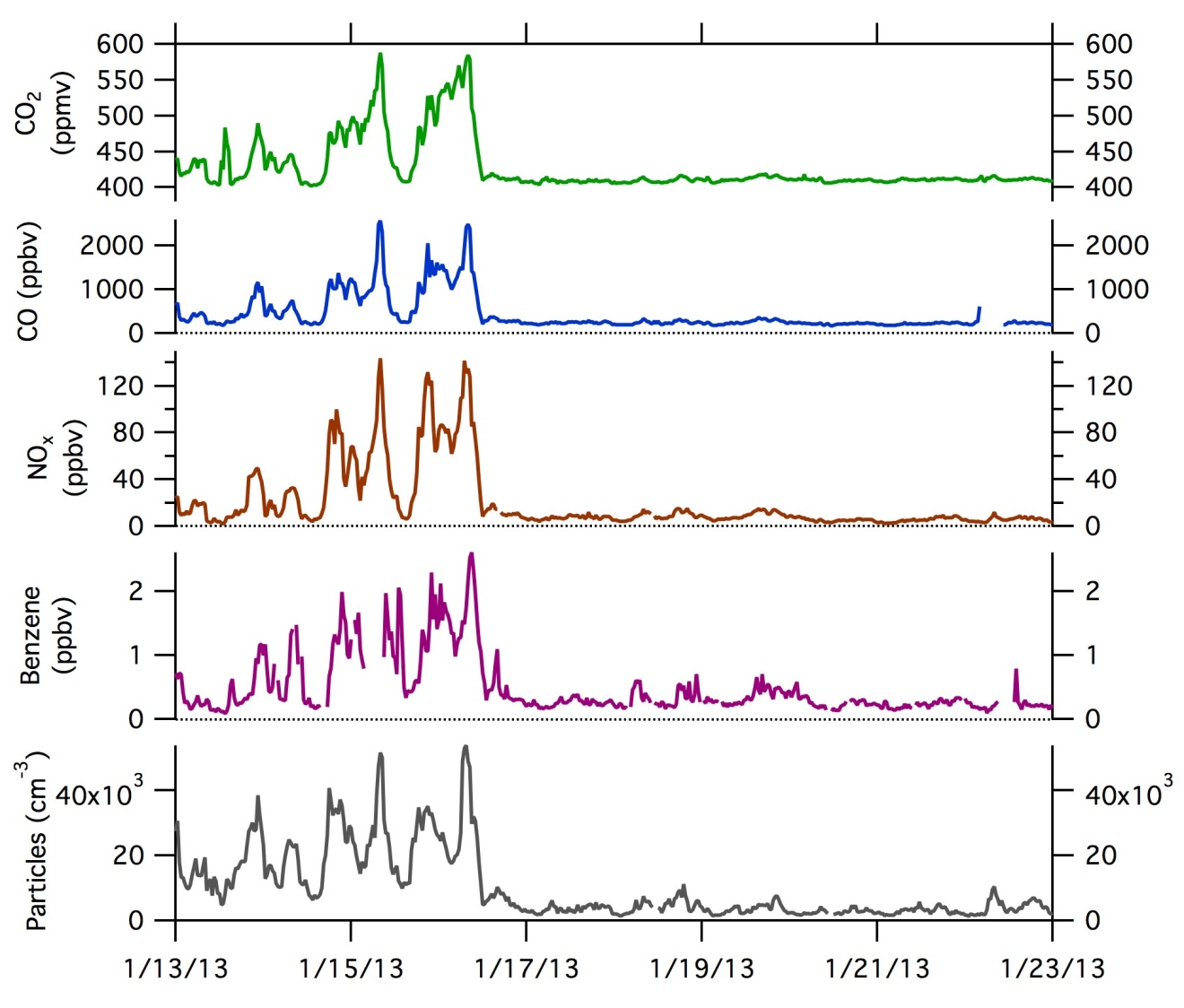


Figure xx: time series for ambient concentrations of both primary (left) and secondary (right) pollutants during both clear and cloudy conditions in Yakima during January, 2013. A persistent cloud cover formed on the 16th, coinciding with a rapid dilution of primary pollutants and much more gradual dilution of secondary species. (YAWNS report?)

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, more importantly, evaluating the performance of the current model structure. While modeled processes cannot blindly be 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 Northwest’s premier air quality forecasting framework, Air Indicator Report for Public Awareness and Community Tracking (AIRPACT) for a wintertime stagnation scenario with relatively complex terrain and meteorology. Accordingly, the majority of this paper will be devoted to AIRPACT model evaluation.

**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 continuous goal of providing convenient air quality information and forecasts to the public in the form of both model results and measurements (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) for meteorology input. Forecasts are calculated at 4-km horizontal grid spacing. The outer AIRPACT modeling domain is below in Figure xx. For this experiment, modifications were made alternately to the WRF meteorological output and within the SMOKE inventory and emissions processing.

(map of model domains)

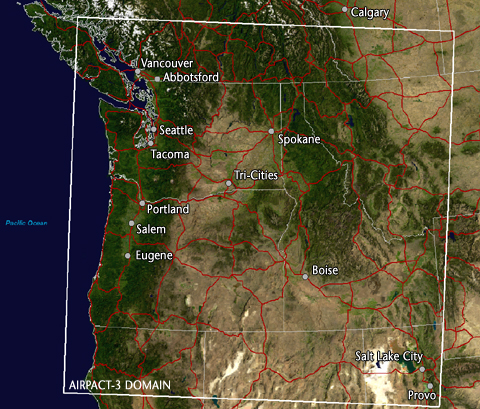


Figure xx: Outer domain of the AIRPACT-4 air quality forecasting framework for the Pacific Northwest.

For general AIRPACT-4 evaluation against YAWNS observations, two versions of the model were run with identical emissions inputs but different meteorological precision from the Weather Research and Forecasting Model (WRF). The initial run used the standard WRF forecast configuration with 38 defined vertical layers, including a 39-meter ground layer. The second AIRPACT-4 run that was analyzed for this study employed finer layer resolution in WRF and meteorology “nudging,” in order to assess the impact of refined meteorology input on AIRAPCT-4 accuracy. An extra vertical layer was added, and the bottom layer was trimmed to 21m. In addition, Rui Zhang (of…?) performed both analysis nudging and observational nudging on the WRF forecasts. The WRF model includes the option to generate these adjustments in order to improve air quality prediction down the line, because the accuracy of forecast meteorology fields tends to decrease with time after the initial conditions. Observation nudging describes the process of 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 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 at near the surface. Thus, analysis nudging was applied to meteorological variables above the PBL by continuing to translate nudged model predictions upward.(methods? need to check this more). The result is a set of dynamic analyses that more accurately characterize local meteorology patterns. The net effect of the meteorology nudging on air quality model output is discussed in the Results section.

Give details on the emission inventory (perhaps show some totals by source type)

The AIRPACT-4 modeling system also relies on emission inputs, which are processed by SMOKE from annual reported 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 NEI 2008). 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 residential heating, 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. Some emissions totals…”EMISIONS TSD…” in Docs?

General discussion of the major sources included / Mobile Sources

Details on MOVES and how it was implemented

The AIRPACT-4 modeling system is directly impacted by 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 streamlined, consisting primarily 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 NOx, CO2, CO, O3, hydrocarbons, and PM2.5 along the major roadways, especially during peak commute hours. In AIRPACT, mobile emissions are estimated by the Motor Vehicle Emission Simulator (MOVES), developed by the EPA. The overall function of MOVES is to create emissions estimates from on-road mobile sources based on pre-calculated MOVES-based emissions and 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 road coordinates for major thoroughfares are typically provided.

Residential Wood Combustion Sources ----- SEE “EMISSIONS TSD” in Documents And “RWC paper!” Maybe this should be a separate, second section after the initial YAWNS evaluation

Details on RWC algorithm, temporal profiles, etc –Again, make this its own section at the end??

**Results, Part 1: Evaluation of Meteorological Variables**

The most glaring discrepancy between the meteorological measurements and model results for YAWNS was the failure of WRF to simulate the condition change from clear to cloudy skies in Yakima during the latter half of January, 2013. This condition change, beginning on the 16th, 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. The refined version of WRF, utilizing nudged output, did only a slightly better job of adding cloud cover to this period. Even in the nudged conditions, the clouds were dense or persistent enough to significantly increase pollutant forecast accuracy. Figure x below graphically summarizes the key meteorological differences in cloud cover observations and WRF simulations for Yakima in January 2013.

Figure x: Graphical comparisons of observed and modeled could cover over Yakima during YAWNS. Note that the observations are reported in eighths of sky coverage.

\*\*Fix scale on top plot

The cloud coverage figures suggest that no truly significant differences between modeled and observed cloud fractions emerge until around the morning of the 16th, when the cloudy period begins. At that time, the observed cloud fraction becomes almost exclusively 8/8 (or 1.0) for the next week. Meanwhile, the modeled cloud fractions generally stay close to zero, until the second half of this week when the refined MCIP output increases its fraction toward 1.0. While this is an improvement over the original meteorology forecasts, the upcoming pollutant concentration analyses illustrate that the modeled meteorology is still not realistic enough to facilitate accurate air quality forecasts during that period.

First, a brief comparison of planetary boundary layer (PBL) heights is provided to help illustrate the differences in ventilation between actual and modeled conditions. Figure x, 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 mixed than in the model until after the 22nd when clouds began to dissipate again. A comparison of modeled and observed wind speeds is also provided below in Figure x. While there is plenty of discrepancy 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. This would seem to isolate cloud modeling, and consequently PBL height, as the most important shortcoming of the model for this study.

Figure x: Time series comparison of modeled and observed PBL heights for YAWNS

Figure x: Time series comparison of modeled and observed wind speeds for YAWNS

Figure x: Comparison of CMAQ v1 and v2 ventilation indices extracted from the WRF-MCIP output. The ventilation index was calculated here as the product of wind speed (m/s) and PBL height (m).

**Evaluation of Pollutant Concentrations**

As a result of increased PBL height with greater boundary layer insulation, most observed pollutant species show an abrupt drop in concentration as mixing increases in the second half of January. However, this pattern is not replicated by the initial CMAQ run. The second run with 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.

In the plots below, observed ambient pollutant concentrations from WSU YAWNS measurements are compared to CMAQ-WRF model results for several pertinent species. In each evaluation, “CMAQ V1” represents the initial WRF-SMOKE-CMAQ run with WRF meteorology inputs based on the 40m surface layer and forecasted conditions. The “CMAQ V2” data represent the second run with a 20m surface layer and nudged conditions in an attempt to mimic actual conditions more closely.

Figure x: Observed ambient carbon monoxide mixing ratios compared to model results from both WRF meteorology inputs.

Figure x shows how each model run fails to capture the spikes in CO concentration just before the cloudy period, and then over-predicts CO during the more ventilated conditions of 1/16 – 1/22. As shown in the lower CO plot, the second run does predict slightly lower diurnal 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, it is that the diurnal agreement between model and observations appeared to increase with the second model run.

The evaluation of CMAQ-WRF-predicted NOx is similar to that of CO. Again, the model does not replicate the highest observed concentrations, and there is still not enough of a concentration decline during the more ventilated period. Of course, this appears to be due to the same basic problem of maintaining cloud cover in the WRF meteorology simulation.

Figure x: Observed ambient nitrogen oxides mixing ratios compared to model results from both WRF meteorology inputs.

The most significant discrepancies between the forecasted WRF-CMAQ and nudged WRF-CMAQ runs occurred for the PM2.5 predictions. As shown in the lower plot of Figure x, the refined WRF-CMAQ results diverge most conspicuously from the original model run during the more ventilated period. While it is encouraging to see modeled particulate concentration drop during this period, it is apparent that the drop actually begins and ends sooner than in the observed concentrations. Why? Note that Department of Ecology data were used for PM2.5, as the available WSU AMS measurements represent PM1.0.

Figure x: Observed ambient fine particulate concentrations compared to model results from both forecasted (v1) and nudged (v2) WRF meteorology inputs.

Below, Figure x provides a closer looks at modeled and observed PM2.5 concentrations, including both WSU AMS and Ecology TEOM measurements, over a shorter time period. The 12th to 16th represents a period of relatively high pollutant concentrations in Yakima. The plot shows some sporadic agreement in concentration magnitude, but indicates that CMAQ is often predicting the peaks and valleys at opposite times from the observations. WHY? The timing of both predicted and observed PM2.5 concentration spikes vary between morning and evening. Note that the WSU AMS is actually measuring PM1.0, which may explain the discrepancy from the TEOM during the concentration spikes on the 15th.

Figure x: Closer comparison of modeled PM2.5 with observed PM2.5 from the Department of Ecology’s TEOM instrument and PM1.0 from WSU’s AMS from 1/12/13 to 1/16/13 in Yakima.

The nitrate and ammonium speciated aerosol predictions, shown in Figures x and x, follow the same general pattern as the total PM2.5 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 diurnal variation. WHY? In the bottom figure of this series, modeled black carbon (BC), expected to be a strong tracer for wood smoke, is also compared to total observed PM2.5. One interesting aspect of the modeled BC time series is that even the second model run does not indicate any period of decreased concentrations, as is the case in other modeled aerosols. This might suggest a relatively constant production and presence of wood smoke particulates throughout even the more ventilated conditions.

Figure x: Observed ambient aerosol nitrate concentrations compared to model results from both WRF meteorology inputs.

Figure x: Observed ambient ammonium nitrate concentrations compared to model results from both WRF meteorology inputs.

Figure x: Modeled YAWNS black carbon (above), compared to total observed PM2.5 (below)

In addition, the modeled divisions and primary and secondary organic aerosols were examined. In the plot below, Figure x, the refined version of CMAQ results for POA and SOA are compared to WSU’s AMS measurements of total organic aerosols. Clearly, CMAQ is underestimating total OA. The relative correspondence during the period of 1/16 to 1/22 appears to be a coincidental result of the model’s failure to capture the more ventilated conditions during that time. Furthermore, CMAQ is allocating a negligible amount of organics as SOA. This apparent under-prediction of SOA may help account for the under-prediction of total OA.

Figure x: Observed AMS total organic aerosols and CMAQ modeled primary and secondary organic aerosols.

A comparison of modeled and observed wind speeds, shown above in Figure x, does not appear to reveal the reason for the discrepancy in the “dispersed” period between modeled and observed aerosols. There are not significant differences in wind magnitudes during the cloudy period when concentrations drop. In fact, the highest wind speeds are both observed and modeled several days before this period. Likewise, the examination of observed and modeled PBL heights does not offer much insight. The highest PBL heights roughly accompany the highest wind speeds. Even during the period of decreased pollutant concentrations, the observed PBL heights are consistently higher than the modeled heights despite the over-prediction of surface pollutant concentrations. This gap in explanation suggests that there are additional, unseen processes at work, which are affecting the pollutant concentrations observed near the surface.

Accordingly, vertical profiles of some key species were constructed from the second version of CMAQ results. This is one area where the model can provide some useful data that is not available in the measurements. The most interesting modeled vertical profiles were those of particulates. Past YAWNS analyses have hypothesized that during stagnant conditions, the observed increases in particulate nitrate during morning hours may be due to nitrate production in the layer overnight that mixes downward as the surface layer deepens in the morning (VanReken et al. 2014). One important discrepancy between the modeled and observed meteorological conditions is that cloudy and clear days often did not correspond. However, it appears that *modeled* clear days do support the hypothesis of downward particulate mixing. This is likely due to the presence of an inversion during the nighttime and morning hours of those clear, stagnant days. As shown below in Figures x and x, a buildup of ANO3 forms in the early morning hours and mixes downward throughout the day. By comparing to the attached time series of modeled PBL height, it is apparent that the buildup forms and remains just above the PBL before mixing downward. Meanwhile, a *modeled* cloudier day, 1/6, did not display significant ANO3 concentrations in the residual layer. At the bottom of this series of plots, Figure x shows some vertical temperature profiles for 1/11 and 1/17. There is a clear, strong modeled temperature inversion on the 17th, which corresponds to the strongest ANO3 buildup in the residual layer. On the other hand, a much weaker inversion, which quickly dissipates, exists over night on the 11th, when only slight ANO3 buildup in the residual layer was predicted.

Modeled vertical profiles for primary and organic aerosols were also examined, below in Figure x. No inverted concentrations appeared to be modeled for POA. However, some slight SOA buildups in the residual layer were formed mostly on the same days as for aerosol nitrates. These buildups also appeared to mix downward as morning fell, increasing the surface concentrations. However, we may recall that modeled SOA was insignificant, compared to POA. One postulation of the initial YAWNS study was that aqueous processing within the cloud might be enhancing secondary organic aerosol (SOA) formation. If this is indeed the case, it may help explain the lack of SOA in the model, if it is not simulating this process.

Finally, some modeled vertical profiles of carbon monoxide are also included below, in Figure x. These profiles suggest that CO mixing ratios decrease rapidly with height at all times.

Vertical Profiles



Figure x: Modeled vertical profiles of aerosol nitrate on 1/6/13 and 1/11/13. In the model, 1/6 represented a relatively cloudy day with higher ANO3 while 1/11 was clearer with lower concentrations. The plot of modeled PBL is included at the bottom for reference.



Figure x: Vertical profile development of nitrate aerosol over the course of 1/17-1/18, which was a near-cloudless day in the model with only moderate ANO3 concentrations.



Figure x: Vertical profiles of primary and organic aerosols when some buildups in the residual layer were evident.



Figure x: Vertical profiles of CO on selected days. As expected, CO levels drop drastically with height.



Figure x: Ambient temperature profiles for 1/11 and 1/17, showing a strong inversion during 17, one of the days with the most dramatic ANO3 buildup in the residual layer.

**Source Attribution and Analysis of Emissions Inventory**

Several approaches were taken in attempt to evaluate the Emission Inventory utilized by CMAQ in this study. Emission estimates were provided by AIRPACT (2002?), processed by SMOKE and input to CMAQ along with WRF meteorology to generate air quality forecasts.

First, modeled pollutant concentrations were compared with a ratio of emissions (moles/s) to a 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 concentration. Unfortunately, very little correlation was observed for CO or NOx in the single Yakima 4km grid cell, as shown below in Figure x. However, a stronger relationship between the pollutant concentrations and ventilation indices is visible when presented as a time series (Figure x). Given the time series charts, it appears that the reason for the lack of explicit correlation may be simply that the concentration peak lag behind the high emissions/low ventilation conditions.

Figure x: Comparison of modeled CO and NOx concentrations to a factor of (emission rate) / (ventilation factor = U\*PBL). The plots do not suggest any significant relationship

\*fix this graph Figure x: Time series plots of modeled concentration vs. [emissions / ventilation index] for CO and NOx in the YAWNS Yakima 4km grid cell

The next phase of emissions evaluation was performed by comparing the ratios of CO to NOx in both concentrations and emissions throughout the region. All comparisons were made on a molar basis. As shown below in Figure x, CO/NOx concentration mixing ratios were calculated by finding the average slope of CO vs. NOx ambient mixing ratios, both for modeled and observed data. Only the days 1/05/13 through 1/17/13 were analyzed because all datasets spanned this period and because it does not include the major concentration discrepancy during the cloudy period. In the Yakima grid cell, it is immediately obvious that the modeled CO/NOx ratios are much higher than observed; around 30 versus 14, respectively. Using measurements from the Toppenish site to the southeast of Yakima, shown in Figure x, it is apparent the CO/NOx at that location during the same time period is even significantly less than in Yakima observations.

Figure x: Observed and modeled CO/NOx concentration ratios in Yakima from 1/05/13 to 1/17/13

While the cause for the differences is not immediately clear, one possible explanation is a set of inaccuracies in the emissions inventory used for the model. To further investigate this possibility, ratios in the emissions were compared to ambient mixing ratios. Table x, below, displays a summary of CO and NOx emissions for the 4km YAWNS grid cell and entire and 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 MOVES mobile emission sources, residential wood combustion (RWC), and area sources associated mostly with agriculture.

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. One of the goals of the remaining portion of this study is how big of a role residential wood smoke plays in driving these different concentration profiles.

**Modeled Wood Smoke Emissions**

(post model ratios for different stove types)

Table x, below summarizes the modeled CO and NOx emissions from both residential wood combustion (RWC) and MOVES in the Yakima Valley and the individual 4km YAWNS grid cell. Additionally, wood smoke fine particulates (WSPM2.5) emissions are included. The CO/NOx emission ratios for each emission category and site can be compared to the modeled and observed ambient CO/NOx ratios. The CO/NOx ratio associated with RWC *anywhere* is strikingly high, at over 100. As previously stated, this is much higher even than the MOVES CO/NOx ratio, which is already significantly higher than ambient ratios. This discrepancy may point to some combination of excessively modeled RWC and MOVEs emissions relative to other factors, or … (failure of model to dissipate these with chemical mechanisms?). Of course, it is also a possibility that the pollutant ratios within the modeled emissions are simply off kilter. After all, NOx was grossly under-predicted by CMAQ for most periods of this study.

Note that expanding the focus from the Yakima grid cell to the entire County decreases the CO/NOx concentrations in both the model and observations, which is clearly a result of less traffic and population density. Clearly, higher population appears to correlate with higher CO ratios in both the model and measurements.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | **RWC CO moles/day Emission** | **RWC NOX mol/day (NO2) Emission** | **WS PM2.5 kg/day Emission** | **RWC CO/Nox molar Emission** |
| **YAWNS grid cell** | 1.96E+04 | 1.93E+02 | 7.97E+01 | 101.95 |
| **Yakima County** | 2.69E+05 | 2.64E+03 | 1.09E+03 | 102.02 |
| **WA state** | 9.95E+06 | 9.22E+04 | 3.96E+04 | 107.99 |
|  |  |  |  |  |
|  | **MOVES Emis. CO avg mol/day, Jan 1-17** | **MOVES Emis. Nox avg mol/day, Jan 1-17** | **MOVES Emis. PMfine avg kg/day, Jan 1-17** | **MOVES Emissions Avg. CO/Nox, Jan 1-17** |
| **YAWNS grid cell** | 3.39E+05 | 6.01E+03 | 1.49E+00 | 40.1 |
| **Yakima County** | 1.60E+06 | 6.37E+04 |  | 25.1 |
|  |  |  |  |  |
|  | **Total CMAQ Emis. CO avg mol/day, Jan 1-17** | **Total CMAQ Emis. NOx avg mol/day, Jan 1-17** | **Total Emis. PMfine avg kg/day, Jan 1-17** | **Total CMAQ Emissions Avg. CO/Nox, Jan 1-17** |
| **YAWNS grid cell** | 3.97E+05 | 1.01E+04 | 4.71E+01 | 35.8 |
| **Yakima County** | 1.92E+06 | 8.20E+04 |  | 22.1 |
|  |  |  |  |  |
|  | **CMAQ Jan. mixing ratio CO/Nox** | **Observed Jan. CO/Nox** |  |  |
| **YAWNS grid cell** | 30.28 | 14.05 |  |  |
| **Yakima County** | 21.45 |  |  |  |

Table x. AIRPACT January emissions of CO, NOx, and fine PM attributed to Yakima by NEI (year?) through MOVES, residential wood combustion (RWC) and total AIRPACT emissions. CO/NOx ratios are included for each emission category, as well as modeled and observed mixing ratios in Yakima during January, 2013.



Figure x: Diurnal variation of modeled residential wood combustion emssions of CO, NOx, and PM2.5 during January in Yakima.

Figure x: Time series of diurnal CO/NOx ratios in the modeled emissions. Ratios appear to peak each day during morning rush hour periods as a result of increased vehicle emissions

Compare magnitude of molar emissions from RWC to those from MOVES – does this suggest RWC is significant? \*Note that WA is the 2nd highest wood smoke emissions state, close behind Oregon, of the ones listed.

TO DO: How does Yakima County compare? This might help with perspective – 9th highest rwc emissions in WA. (roughly equal to population rank) 39 counties in WA

Bivariate model analysis (Move this up? –see PNWIS presentation)

Another advantage of model results is the ability to deconstruct certain ambient pollutant concentrations into their approximate emission components by examining the emission factors within the modeled emissions inventory. Then, these relative contributions can be compared to observations. In this case, a previous source attribution study was performed on the YAWNS measurement data by applying a simple bivariate model (cite)

(show Grahams bivariate model)

For the modeled emissions inventory, RWC and mobile emission factors were calculated as the molar fraction of particular pollutant emissions within each category relative the total modeled emissions of that pollutant. For instance, to establish carbon monoxide wood combustion and mobile emission factors:

EFRWC = [RWC CO(moles/s)] / [total AIRPACT CO(moles/s)]

EFMOVES = [MOVES CO(moles/s)] / [total AIRPACT CO(moles/s)]

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 number of days was not necessary for this calculation. For the most of the pollutants examined, in January in Yakima, the major sources were limited to vehicles, area sources, and wood smoke. It has been previously established in YAWNS that point sources in the area are virtually absent. It was also clear that biogenic sources of these species were extremely limited. Thus, the remaining anthropogenic sources, after mobile and residential wood combustion (RWC), are assumed to be mostly area sources, such as agriculture land use. The modeled emission factors calculated for a handful of pertinent pollutants are displayed below as pie charts in Figure x. Then, these factors were applied to observed 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 x. Note that these comparisons neglect chemical mechanisms.

The modeled emission factors can be compared to a similar source attribution method performed for this study using a simple bivariate model with vehicle and wood smoke tracer species (VanderSchelden/Jobson). This was basically done by Graham! …



Figure x: Major pollutant proportions of AIRPACT emissions for YAWNS during the period of stagnant conditions. This inventory is based on the 2008 NEI





Figure x: Time series of a graphical application of the modeled emission factors from Figure x to the observed YAWNS ambient concentrations of select pollutants. Each color represents the respective, estimated, contribution of a major source type to observed concentrations.

**Temperature Correlation**

Napelenok et al. (2014) recognized a strong inverse correlation between minimum observed temperature and RWC tracer concentration in the Southeastern U.S. Of course, this coupling does not exist in the emissions inventory, and thus their study concluded that a temperature adjustment to modeled RWC emissions was one way to improve modeling accuracy for PM2.5. However, the inherent complexity in coupling temperature with emissions in a forecast environment kept them from testing this avenue. In the Northwest, wintertime RWC are certainly more prominent than in the warmer Southeast region and therefore exhibit an even bigger 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 PM2.5 forecasts in the AIRPACT-4 domain.

In the current NEI, RWC emissions are allocated according to expected monthly and hourly usage patterns given wood stove census data for a particular region. Monthly factors are intended to reflect typical seasonal temperature effects. However, The effects of daily and hourly temperature swings have not been accounted for in previous model runs. The typical diurnal emission patterns of our wood smoke tracer, WSPM2.5, for January, are shown below in Figure x. Distinct temporal emission patterns are modeled for weekends and holidays, but otherwise the magnitudes do not change within a given month. To investigate whether minimum temperature did indeed appear to affect January RWC emissions in Yakima, I plotted daily maximum observed organic aerosol concentrations with daily minimum observed temperature, as shown below in Figure x.



Figure x: Plot of daily maximum observed organic aerosols with daily minimum observed temperature. A limited sample size is applied, but the data does indicate a negative correlation between particulate concentration and temperature.

As anticipated, Figure x, above, suggests 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 out a temperature-adjustment algorithm for wood stove emissions in the AIRPACT4 modeling framework.

Accordingly, a wood stove temperature adjustment algorithm was applied to the modeled emissions inventory in a series of steps. First, wood smoke emissions, in tons/heating-degree-hour, were reduced to a flat baseline value over each day, 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 avoided redundantly using a temperature adjustment. The final factor applied to the wood smoke emissions was of course the hourly heating-degree term, using the forecasted 2-m hourly temperature from the MCIP output file in degrees F, relative to a maximum of 50 degrees F, with a minimum of 20 degrees F. The result was a new RWC emissions profile, generally scaled upward and unique for each hour of the day. A comparison of the initial and T-adjusted emission profiles for WSPM2.5 is shown below in Figure x. Below that, the resulting PM2.5 concentrations considering both original and T-adjusted wood smoke emissions are compared against observations for the month of January in Yakima.

Figure x: Comparison of original and temperature-adjusted emissions of the wood smoke tracer WSPM2.5 for 1/05/13 - 1/15/13 in Yakima.



Figure x: AIRPACT4-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.

Temperature Adjustment Conclusions/concerns:

-Not much of an improvement for Yakima

-At some sites, particularly larger ones, the positive bias for PM2.5 forecasts increased drastically with T-adj emissions.

PM2.5 time series for other sites? OR Mean bias/Mean FE table for our sites?

-there appear to be more fundamental PM2.5 emission biases affecting the accuracy of AIRPACT4 forecasting at both urban and rural sites. A population density adjustment may be a more effective way to increase PM2.5 forecasting accuracy.

-alternative surrogates for wood smoke emissions (Farren) – do these help? -Underway

YAWNS modeling Conclusions…?

* CMAQ has trouble modeling and maintaining cloud cover, which seems to have big negative effect on accuracy during cloudy periods
* Nudged results, 20m instead of 40m surface layer improved accuracy, but not significantly
* Aerosol concentrations changed the most with refined WRF output, although period of enhanced ventilation still did not match observations. This seems to support the idea that clouds are having big effects on aerosol chemistry, particularly nitrates.
* Vertical profiles – model shows higher nitrate aerosol concentrations in early morning residual layer than at surface. This occurred mostly during clear stagnation conditions *in the model*.
* Very limited SOA in the model, but appears to be some formation in residual layer. Is the model failing to simulate SOA production? Maybe this accounts for under-prediction of total organic aerosols?
* Modeled ambient CO/NOx ratios are much higher than observed in the Yakima Valley, January.
* It seems likely that the emissions inventory has some inherent inaccuracies. Could we be over-predicting WS and/or vehicle emissions? Both would seem to drive up the CO/NOx ratio.
* Is the CO/NOx within modeled emissions just too high? Recall that NOx is VERY under-predicted for this study period.

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