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Research Article

Wood-Burning Device Changeout: Modeling the Impact on $PM_{2.5}$ Concentrations in a Remote Subarctic Urban Nonattainment Area

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The effects of exchanging noncertified with certified wood-burning devices on the 24h-average $PM_{2.5}$ concentrations in the nonattainment area of Fairbanks, Alaska, in a cold season (October to March) were investigated using the Weather Research and Forecasting model inline coupled with a chemistry package. Even changing out only 2930 uncertified woodstoves and 90 outdoor wood boilers reduced the 24 h-average $PM_{2.5}$ concentrations on average by 0.6 $\mu g.m^{-3}$ (6%) and avoided seven out of 55 simulated exceedance days during this half-a-year. The highest reductions on any exceedance day ranged between 1.7 and 2.8 $\mu g.m^{-3}$. The relative response factors obtained were consistently relatively low (~0.95) for all $PM_{2.5}$ species and all months. Sensitivity studies suggest that the assessment of the benefits of a wood-burning device changeout program in avoiding exceedances heavily relies on the accuracy of the estimates on how many wood-burning devices exist that can be exchanged.

1. Introduction

In 2006, the Environmental Protection Agency (EPA) has tightened the 24 h National Ambient Air Quality Standards (NAAQS) to 35 $\mu g.m^{-3}$ for fine particulate matter having diameters equal to or less than 2.5 μm ($PM_{2.5}$). From October to March the $PM_{2.5}$ data collected in prior years indicated that $PM_{2.5}$ concentrations exceeded the NAAQS frequently at the official monitoring site in Fairbanks [1]—a remote urban area in the subarctic of Alaska. Therefore, Fairbanks was designated a $PM_{2.5}$ nonattainment area in 2009.

In Fairbanks, wood-burning devices are major contributors to the $PM_{2.5}$ emissions in residential areas [2]. An estimated 9240 wood-burning devices exist in Fairbanks, of which 7980 devices are woodstoves [2]. Due to the increasing price of heating fuel, many Fairbankisan households added wood-burning devices or shifted to a higher percentage of heating with wood as is evident from the threefold increase of wood-cutting permits from 2007 to 2009 (Conner, pers. com. 2010).

The emissions from wood-burning devices vary with fuel type, fuel moisture, burning practice, and control techniques of the devices [3]. In general, EPA-certified woodstoves emit up to 87% less $PM_{2.5}$ than uncertified ones [3]. EPA [4] estimated that 10 million woodstoves are being used in the United States, about 80% of which are uncertified devices. Exchanging uncertified woodstoves with certified ones has been a successful tool to mitigate $PM_{2.5}$ concentrations in many places [5].

The effects of woodstove changeout programs on reducing ambient $PM_{2.5}$ concentrations have been evaluated mainly based on observations. For example, the $PM_{2.5}$ sampling campaign related to the changeout of 1200 uncertified woodstoves in Libby, Montana, showed that 24 h-average $PM_{2.5}$ concentrations decreased by 20% during the changeout period [6]. Indoor $PM_{2.5}$ concentration measured in 16 homes prior and after the woodstove changeout in a Rocky Mountain valley community [7] indicated reduction of average and maximum $PM_{2.5}$ concentrations of 71% and 76%, respectively. A similar study performed in 15 homes in British Columbia, Canada, found no consistent relationship between the indoor $PM_{2.5}$ reductions and the woodstove changeout [8].

Of the 8610 inserts and woodstoves in Fairbanks, about 2930 devices are uncertified ones [2]. An assessment of the benefits of a wood-burning device changeout for any high latitude urban community based on observational studies in midlatitudes is difficult. Fairbanks' subarctic meteorological conditions differ strongly from those in the mid-latitude places where wood-burning device changeout programs have been applied successfully to mitigate air pollution. In Fairbanks, the often stagnant air and strong radiative cooling during the long nights lead to low temperatures and strong inversions. Inversions exist on 78–97 days between October and March and often last for more than ten consecutive days. The 1971–2000 monthly mean temperatures in October, November, December, January, February, and March were −9, −18, −22, −23, −18, and −14°C, respectively. Such extremely low temperatures result in high heating demands. The calm winds (0.5–2.5 m on monthly average between October and March) and inversions mean low mixing of the polluted air with the unpolluted environment.

Whereas the observational approach applied in mid-latitudes requires an extensive measurement campaign over the changeout program lifetime, numerical modeling can provide a quick and low-cost assessment of the benefits of a wood-burning device changeout program. Furthermore, modeling permits assessment of the potential benefits of a changeout program prior to its implementation/completion and hence permits implementation of additional measures in case the changeout program alone may not be sufficient enough to achieve compliance.

To this aspect, the Weather Research and Forecasting model inline coupled with a chemistry model commonly known as WRF/Chem [9, 10] has been widely used to investigate pollution sensitivity to changes in emissions. For example, WRF/Chem served to investigate the effects of changing emission of nitrogen oxides (NO_x) from power plants on ozone concentrations in the eastern United States [11]. The simulations elucidated complex relationships between ozone concentrations and NO_x emission strength, the proximity of other NO_x sources, the availability of volatile organic carbon (VOC), and sunlight. WRF/Chem simulations to study the impacts of urban expansion on the formation of secondary organic aerosol over the Pearl River Delta, China, showed that urban expansion can alter the meteorological conditions and therefore induce increases of secondary organic aerosol between 3 and 9% [12]. WRF/Chem investigations showed that the emission changes between 1990 and 2000 in the North Pacific region caused the increasing trends of sulfate aerosols observed at coastal Alaska sites [13]. These simulations also showed that at coastal sites in southern Alaska, sulfate aerosol was not governed by the local emission changes but by the increased ship emissions and Canadian emissions.

Among many efforts in seeking effective pollution controls to comply with the NAAQS, Fairbanks started conducting a "woodstove replacement" program. Given that Fairbanks' 2008 design value is 44.7 $\mu g.m^{-3}$, any emission-control strategy requires a relative response factor (RRF) lower than 0.78 to reach compliance with the NAAQS. In this study, we used WRF/Chem with its modifications for Alaska [14, 15] to assess the benefits of exchanging uncertified with certified wood-burning devices on the $PM_{2.5}$ concentrations at breathing level in the Fairbanks nonattainment area.

Table 1: Parameterizations used in this study.

Process	Scheme and reference
Cloud microphysics	Six water-class cloud microphysical scheme [16]
Subgrid-scale convection	Further developed 3D version of the Grell-Dévényi cumulus-ensemble scheme [17]
Radiation	Goddard shortwave radiation scheme [18], Radiative Transfer Model for long-wave radiation [19], radiative feedback from aerosols [20]
Atmospheric boundary layer and sublayer processes	[21]
Land-surface processes	Modified Rapid Update Cycle land-surface model [22]
Gas-phase chemistry	[23]
Photolysis frequencies	[24]
Aerosol physics, chemistry and dynamics	Modal Aerosol Dynamics Model for Europe [25] and Secondary ORGanic Aerosol Model [26]
Dry deposition	[27] with the modifications by [14]
Biogenic emissions	calculated inline depending on meteorological conditions [28]

2. Experimental Design

2.1. Simulations. Simulations were performed for October 1, 2008 0000 UTC, to April 2, 2009 0000 UTC, with the Alaska modified WRF/Chem in forecast mode. The physical and chemicals schemes selected for the simulations are listed in Table 1 and were described in detail in [15].

The model domain encompasses most of Interior Alaska centered over the Fairbanks nonattainment with 4 km horizontal grid-increment from the surface to 100 hPa with 28 stretched vertical layers (Figure 1). The top of the first layer (breathing level) is at 8 m height. The initial conditions for the meteorological fields and meteorological lateral boundary conditions were downscaled from the $1^\circ \times 1^\circ$, 6 h-resolution National Centers for Environmental Prediction global final analyses. The chemical fields were initialized with vertical profiles of Alaska typical background concentrations. Since Fairbanks is the only major emission source and urban area within 578 km radius and observational and modeling studies showed hardly any advection of pollutants [13, 15], Alaska background concentrations served as lateral boundary conditions.

We performed simulations without (REF) and with "woodstove replacement" (WSR). In WSR, the numbers of wood-burning devices to be changed out were based on [2]. These authors estimated, there are in total 9240 wood-burning devices of which 2930 and 90 are uncertified woodstoves and outdoor wood boilers, respectively. Since an

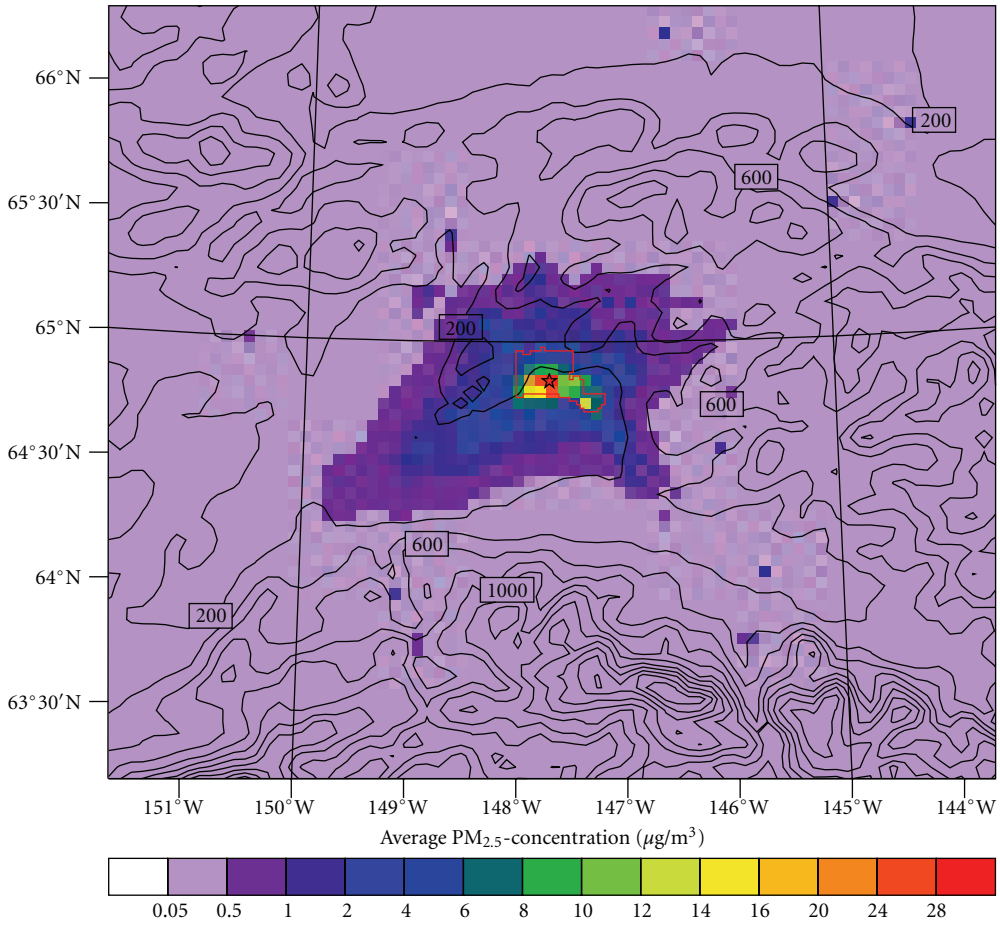

FIGURE 1: Average $PM_{2.5}$ concentration in the domain of interest in October to March as obtained in REF with terrain contours overlain. The star and red polygon indicate the grid cell holding the official monitoring site and the outline of the nonattainment area.

earlier study [29] estimated that there exist 13829 wood-burning devices of which 5042 and 1500 are uncertified woodstoves and outdoor wood boilers, respectively, we performed a sensitive simulation (WSS1) assuming a changeout based on these numbers. A second sensitivity simulation (WSS2) was based on unpublished data by Carlson and collaborators (2009; pers. comm.) that marginally differed in the numbers of total wood-burning devices (9241) and uncertified woodstoves (2934) from the numbers published in [2] and used in WSR, but did not consider pellet stoves (0 versus 370 devices). The sensitivity studies were run for 14 days to assess the sensitivity to the number of wood-burning devices (WSS1) and type of devices (WSS2).

2.2. Emission Inventories. We developed the annual anthropogenic emission inventory based on the National Emission Inventory (NEI) of 2008 available by October 2010. As no point-source emissions were available at that time, we used point-source emission data from facility operators (if provided) and assumed a 1.5%/y increase from the previous NEI otherwise. For some industrial/commercial/institutional sectors that were not available in the NEI2008, we assumed they remained as in the NEI2005 as there was just marginal change in these sectors over 2005–2008. Emission estimates for residential wood combustion were obtained from [29]. The annual emissions for 2009 were assessed with a 1.5% increase from the 2008 base-year.

We considered changes in emission of $PM_{2.5}$, particulate matter having diameters equal to or less than 10 μm (PM_{10}), sulfur dioxides (SO_2), carbon monoxide (CO), carbon dioxides (CO_2), ammonia (NH_3), methane (CH_4), and VOC per wood-burning device exchanged. We calculated the emission of the ith species from wood-burning devices in WSR as follows:

$$E_{\mathrm{WSR},i} = E_{\mathrm{REF},i} + N_{\mathrm{exch}} E_{\mathrm{cert},i} - \sum N_j E_{j,i}, \tag{1}$$

where $N_{\mathrm{exch}} = \sum N_j$ and $E_{\mathrm{cert},i}$ are the number of certified wood-burning devices installed and their emission rates for the ith species; N_j and E_j are the numbers of noncertified wood-burning devices of type j and their emission rates for the ith species per device j; $E_{\mathrm{REF},i}$ and $E_{\mathrm{WSR},i}$ are the total emission rates of the ith species from wood-burning devices in REF and WSR, respectively. The emission rates from wood-burning devices for all species were derived from [29, 30]. Analogously, we calculated the emissions for the assumed changeout of WSS1 and WSS2 with the

corresponding numbers N_{exch} and N_j for each sensitive study. The emissions of all other sectors than wood-burning remained the same in WSR, WSS1, and WSS2 as they were in REF.

This annual emission data was allocated in space and time based on source-specific activity data (land use, population density, traffic counts, point-source coordinates, hour, day of the week, month, etc.) (e.g., Figure 2). In addition, temperature was considered for emissions from traffic, residential, and commercial heating and power generation leading to higher (lower) emissions for daily mean temperatures below (above) the monthly mean temperature [15].

2.3. Analysis Methods. We analyzed the simulations over an area of 80 × 70 grid points (Figure 1) from October 1 0000 Alaska Standard Time (AST) to April 1 0000 AST (which is UTC+8 h) as the 24 h-average is to be evaluated with respect to AST. We determined the differences of $PM_{2.5}$ and its components in REF in comparison with WSR, WSS1 and WSS2. The $PM_{2.5}$ concentration differences (REF-WSR, REF-WSS1, REF-WSS2) were tested for their significance at the 95% confidence level by using a t-test with the null hypothesis that $PM_{2.5}$ concentrations in REF and in each of WSR, WSS1, and WSS2 do not differ.

We evaluated the benefit of the wood-burning device changeout by examining how many "exceedances" and "exceedance days" were avoided. In doing so, we considered 24 h-average $PM_{2.5}$ concentrations at any grid-cell greater than the NAAQS on any day as an "exceedance" and any day that had at least one "exceedance" anywhere as an "exceedance day".

We calculated the relative response factors in response to the emission changes YYY by dividing the concentrations in YYY by those of REF (YYY/REF) where YYY stands for WSR, WSS1, and WSS2, respectively. The RRFs were calculated for total $PM_{2.5}$ and its major components namely sulfates (SO_4), nitrates (NO_3), ammonium (NH_4), organic carbon (OC), elemental carbon (EC), and other primary inorganic particulate matter (others). The RRFs were calculated for all grid cells in the nonattainment area including the grid cell that holds the official monitoring site to assess the effects of the wood-burning device changeout over the nonattainment area.

3. Result

3.1. Model Performance. The evaluation of the baseline simulation (REF) [15] can be summarized as follows. WRF/Chem overestimated temperatures measured at 3, 11 and 22 m at the meteorological tower in downtown Fairbanks by 0.6 K, 0.7 K, and 1.1 K, respectively. It overestimated wind speeds measured at 11 m (22 m) by 1.15 m.s^{-1} (2.39 m.s^{-1}) and overestimated relative humidity by 16%. It well captured the temporal evolution of the meteorological quantities observed at the 23 meteorological surface stations in the domain. In the domain, the overall biases of temperature, dew point temperature, relative humidity, sea-level pressure, wind speed and direction over October to March were 1.3 K, 2.1 K, 5%, −1.9 hPa, 1.55 m.s^{-1}, and 4°, respectively. WRF/Chem slightly overestimated the 24 h-average $PM_{2.5}$ concentration on polluted days ($PM_{2.5}$ concentration >35 μg.m^{-3}) but failed to capture the extremes to their full extent. The occurrence frequency was acceptably captured for $PM_{2.5}$ concentrations between 15 and 50 μg.m^{-3}. WRF/Chem simulated 52 exceedances at the grid cell holding the monitoring site where only 26 exceedances were observed.

The failure to capture the $PM_{2.5}$ maxima (minima) to their full extent on extremely polluted (clean) days does not affect the number of simulated exceedance days and exceedances. During these events, $PM_{2.5}$ concentrations namely were much higher (lower) than the 35 μg.m^{-3} threshold for exceedances. Thus, we can use the REF and WSR simulations to assess the impact of a wood-burning device changeout on the $PM_{2.5}$ concentration in the nonattainment area.

3.2. Emission Reduction. On annual average, $PM_{2.5}$ emissions from residential heating devices made up about 21% of the total $PM_{2.5}$ emissions from all source categories. Wood-burning devices contributed 66.6, 1.4, 14.7, 59.9, 96.5 and 95.8% of the emitted $PM_{2.5}$, SO_2, NO_x, NH_3, VOC, and CO from residential heating.

On average over the nonattainment area, $PM_{2.5}$ emissions in October, November, December, January, February, and March were 941.7, 632.9, 632.5, 799.8, 680.5, and 661.0 g.km^{-2} h^{-1}, respectively. Temperatures were appreciably below the 1971–2000 30-year average in October and above in November, December, January, and February. Consequently, $PM_{2.5}$ emissions were higher in October and lower in November, December, and January than on average.

Over October to March, WSR reduced the total $PM_{2.5}$ emissions by 3.7% compared to REF. The monthly average $PM_{2.5}$ emission reductions were 4.0, 3.2, 2.7, 3.0, 3.9, and 5.6% in October, November, December, January, February, and March, respectively. The magnitude of emission reductions differed among pollutants. On average over the nonattainment area, SO_2 emission reductions were 19.5, 8.16, 9.1, 11.7, 11.0, and 15.8% in October to March, respectively. The respective NO_x (VOC) emission reductions were 16.0 (20.3), 5.5 (8.1), 6.8 (6.6), 8.9 (10.7), 7.3 (11.0), and 11.4 (11.2)%, respectively.

3.3. Reference Simulation. The diurnal courses of $PM_{2.5}$ concentrations were similar in REF and WSR, that is, changes in emissions from wood burning do not affect the general diurnal course of $PM_{2.5}$ concentration. The diurnal course of $PM_{2.5}$ concentration rather reflects the temporal variation of the emissions from all sources. The diurnal course of hourly $PM_{2.5}$ concentrations on days having 24 h-average $PM_{2.5}$ concentrations less than 25 μg.m^{-3} showed a peak at 1000 AST followed by a slightly stronger peak at 1900 AST. On days having 24 h-average $PM_{2.5}$ concentration greater than 25 μg.m^{-3}, the second peak often dominated the first one and had its maximum between 1500 to 1700 AST. Typically, the hourly $PM_{2.5}$ concentrations sharply increased

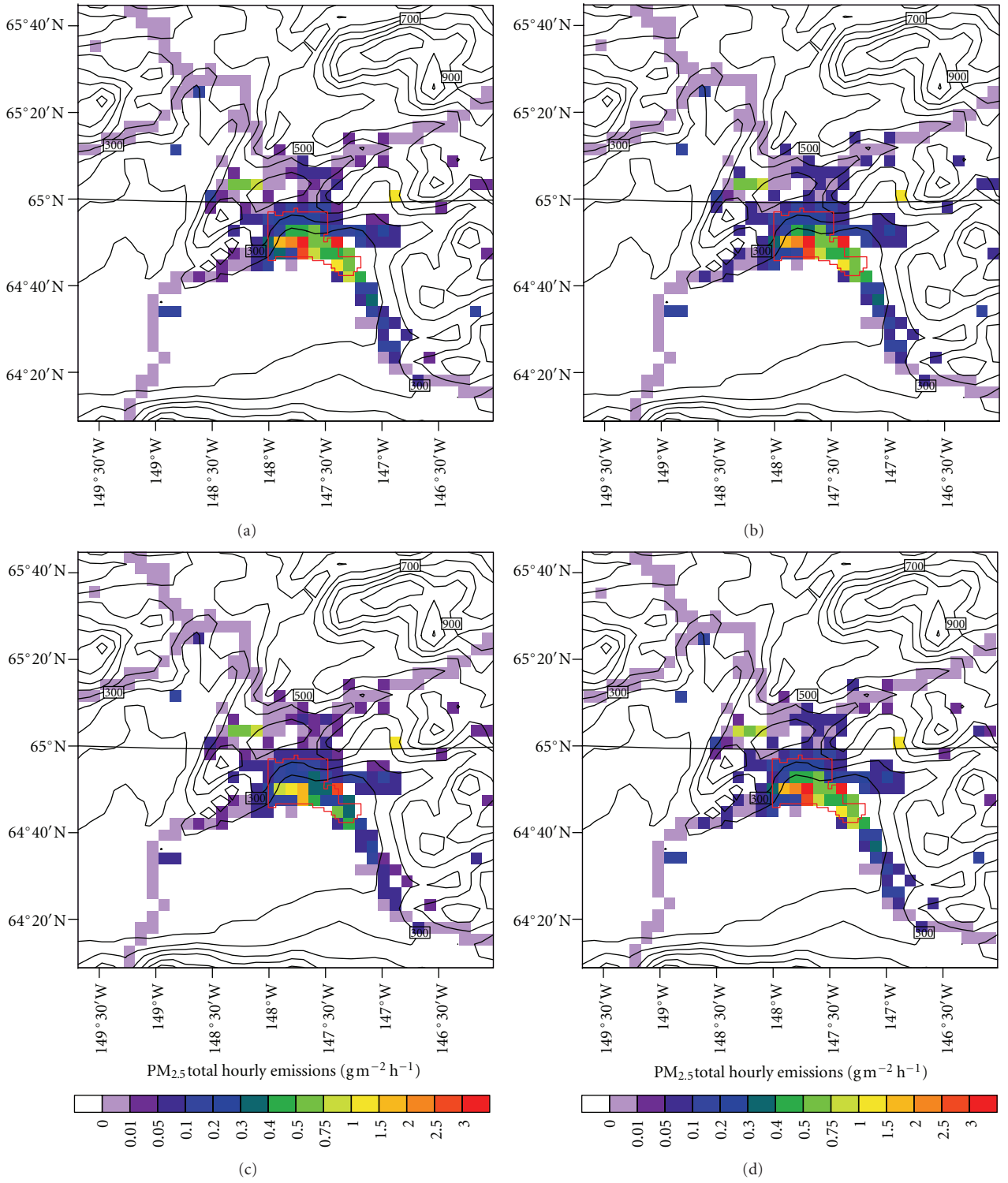

FIGURE 2: Zoom-in on $PM_{2.5}$ emissions in (a) REF, (b) WSR, (c) WSS1, and (d) WSS2 on average over October to March for REF and WSR and October 01–14, 2008, for WSS1 and WSS2.

after 600 AST and quickly decreased after reaching the second peak. From October to March, nighttime (2200–0600 AST) hourly $PM_{2.5}$ concentrations were typically lower and fluctuated less ($\mu = 15.7\,\mu g.m^{-3}$, $\sigma = 9.9\,\mu g.m^{-3}$) than during the remaining hours of the day ($\mu = 37.2\,\mu g.m^{-3}$, $\sigma = 22.0\,\mu g.m^{-3}$).

Over the nonattainment area, REF monthly average $PM_{2.5}$ concentrations were 12.9, 11.0, 9.2, 11.0, 9.8, and

5.7 $\mu g.m^{-3}$ in October, November, December, January, February, and March, respectively. In the nonattainment area, $PM_{2.5}$ concentrations were governed by the emission strength and meteorological conditions. At the grid cell holding the monitoring site, the correlations of 24 h-average $PM_{2.5}$ concentration with 2 m air temperature (T), 10 m wind speed (v), atmospheric boundary layer height (ABL-height), downward shortwave radiation, relative humidity, and sea level pressure were −0.404, −0.626, −0.613, −0.298, 0.043, and −0.001, respectively (all significant at the 95% confidence level). Here, the 24 h-average $PM_{2.5}$ concentrations were strongly driven by emission strength ($R = 0.668$, significant). The average compositions of the 24 h-average $PM_{2.5}$ concentration in all grid cells in the nonattainment area were 21.3–25.0, 0.6–0.8, <0.1, 8.9–9.3, 45.4–47.7, 19.8–20.7% SO_4, NO_3, NH_4, EC, OC, and others, respectively. This finding indicates no notable differences in local $PM_{2.5}$ composition in the nonattainment area.

The on-average high $PM_{2.5}$ emissions (188.3 $g.km^{-2}\,h^{-1}$) and relative low wind speeds (1.9 $m.s^{-1}$) over the nonattainment area in October led to the highest monthly average $PM_{2.5}$ concentrations of October to March. On monthly average, wind speed and ABL-height were lowest (0.9 $m.s^{-1}$ and 122.7 m at the grid cell holding the monitoring site, respectively) in November, which explains the high monthly average $PM_{2.5}$ concentrations despite of the on-monthly-average second lowest $PM_{2.5}$ emissions of October to March. In March, the on-average relatively high wind speed and ABL height (2.6 $m.s^{-1}$ and 567.2 m at the grid-cell of the monitoring site) provided good dilution and transported polluted air out of the nonattainment area, which yielded low $PM_{2.5}$ concentration over the nonattainment area.

In REF, all maximum 24 h-average $PM_{2.5}$ concentrations obtained on any day during October to March occurred in the nonattainment area. Of the 182 days, the highest 24 h-average $PM_{2.5}$ concentrations occurred at the grid-cell holding the monitoring site and/or the grid cells adjacent to it to the south and west (these three grid cells are called site group hereafter) on 86, 64, and 32 days, respectively. This fact is due to relative strong $PM_{2.5}$ emissions in these grid cells in comparison with other grid cells in the nonattainment area. The site group $PM_{2.5}$ emissions made up 34.3% of the total emissions in the nonattainment area that encompasses 31 grid cells.

In REF, 55 exceedance days and 131 exceedances were simulated during October to March, of which 52 exceedances occurred at the grid cell of the monitoring site. The number of exceedance days (exceedances) in October, November, January, February, and March was 20 (57), 10 (13), 5 (13), 15 (37), 5 (11), and 0 (0), respectively. All exceedances typically occurred in the site group. The highest and lowest 24 h-average $PM_{2.5}$ concentrations on any exceedance day were 72.2 and 35.1 $\mu g.m^{-3}$ and occurred on October 27, 2008, and January 4, 2009, respectively.

Exceedances typically occurred when at least any two of the following conditions coexisted: strong emission rate (>3600 $g.km^{-2}\,h^{-1}$), low wind speed ($v < 1$ $m.s^{-1}$), low temperature (<-20°C) and low ABL height (<20 m). These four critical conditions occurred on 23.1, 15.4, 20.3 and 20.3% of the 182 days. Days with high exceedances (>60 $\mu g.m^{-3}$) occurred when all four above mentioned critical conditions occurred concurrently. No exceedances occurred on days with wind speeds greater than 2 $m.s^{-1}$ and ABL-heights greater than 100 m. On days with wind speeds greater than 1 $m.s^{-1}$ and ABL heights greater than 100 m anywhere in the nonattainment area but not at the site group, exceedances were simulated at the grid cell of the monitoring site and/or its adjacent grid cells while the 24 h-average $PM_{2.5}$ concentrations at the other grid cells in the nonattainment area remained low (<15 $\mu g.m^3$). Large concentration gradients always existed between the grid cells of the site group and the other grid cells in the nonattainment area.

On days with calm wind (<0.5 $m.s^{-1}$), high 24 h-average $PM_{2.5}$ concentrations and often exceedances occurred in the nonattainment area and its surrounding area (Figure 3(a)). During October to March, no exceedance occurred when the prevalent northeast wind or the occasional northwest wind advected clean and relatively warm air into the nonattainment area and flushed the polluted air toward the southwest or southeast (Figure 3(b)). Exceedances typically occurred when (1) in the nonattainment area, weak northeast winds were not able to remove the cold and stable air mass (Figure 3(c)), (2) in the nonattainment area, wind came from different directions and hindered the transport of polluted air out of the nonattainment area (Figure 3(d)), (3) northeast or southwest winds transported polluted air out of the nonattainment area that then was advected back into the nonattainment area as aged polluted air (Figure 3(e)), and (4) southeast winds advected polluted air from the community of North Pole (2226 inhabitants, located in the nonattainment area 22 km southeast of downtown Fairbanks) towards the grid-cell of the monitoring site and slowly drained toward the southwest.

3.4. Wood-Burning Device Changeout. On all except eight days, the highest 24 h-average $PM_{2.5}$ concentrations occurred at the same grid cells in WSR and REF. On those eight days, the 24 h-average $PM_{2.5}$ concentration maxima in WSR, however, still occurred within the site group like in REF. The slight shifts in position of the local maxima were due to marginal (in the order of measurement accuracy) changes in meteorological conditions due to indirect and direct feedback between the aerosol concentrations and radiation.

In WSR, the monthly average $PM_{2.5}$ concentrations in the nonattainment area were 12.2, 10.3, 8.6, 10.3, 9.2, and 5.3 $\mu g.m^{-3}$ in October, November, December, January, February, and March, respectively. The values led to monthly average $PM_{2.5}$ differences (REF-WSR) of 0.7, 0.7, 0.6, 0.7, 0.6, and 0.3 $\mu g.m^{-3}$ for October to March, respectively. The $PM_{2.5}$ differences were higher in months with on-average relatively higher than relatively lower $PM_{2.5}$ concentration.

The highest 24 h-average $PM_{2.5}$ difference obtained anywhere in the domain was 5.7 $\mu g.m^{-3}$ (October 27 2008). The highest (2.1 $\mu g.m^{-3}$) and the second highest (2.0 $\mu g.m^{-3}$) 24 h-average $PM_{2.5}$ differences over the nonattainment area were obtained for October 27 2008 and January 1 2009,

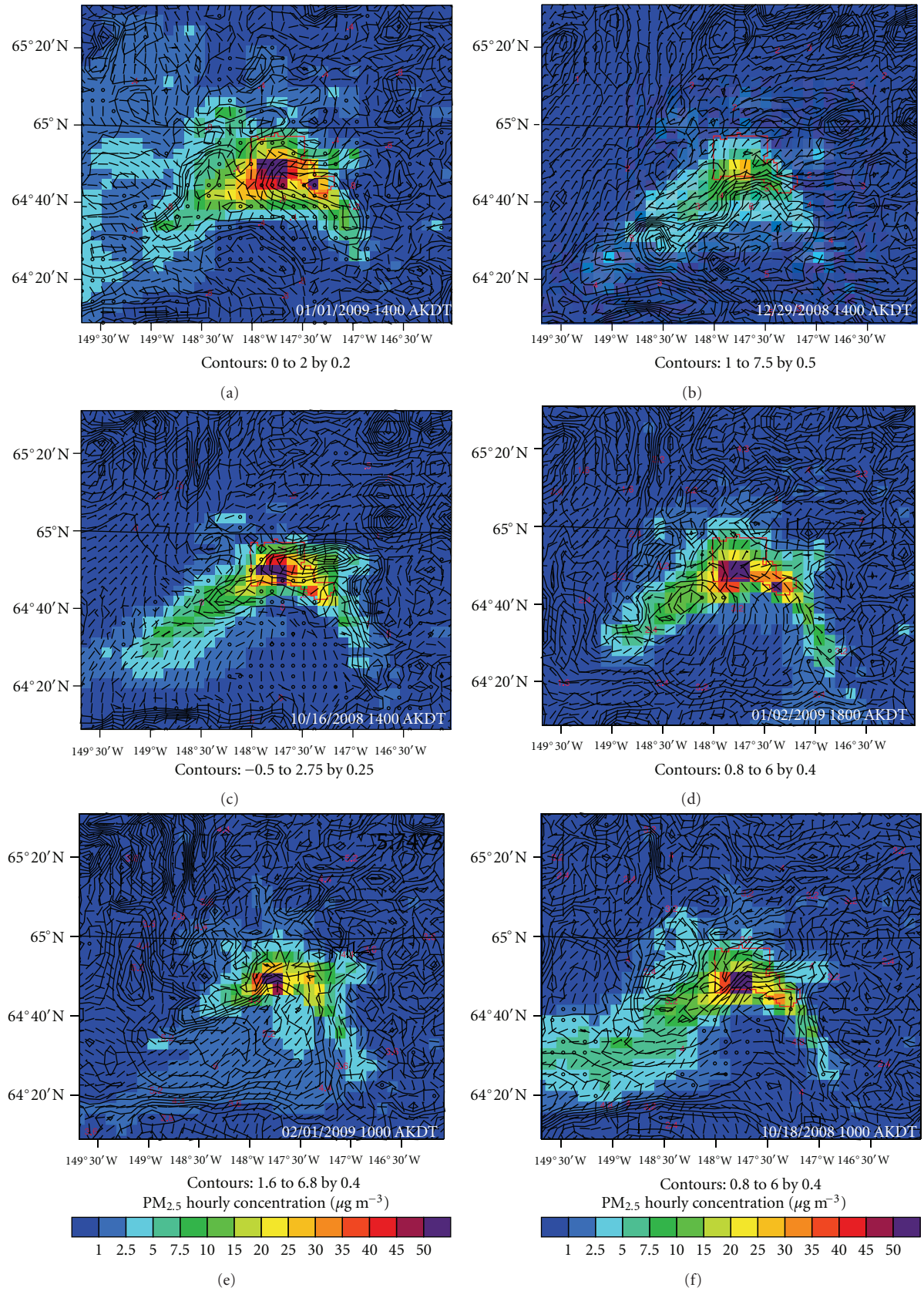

Figure 3: Zoom-in on typical wind circulation patterns at breathing level associated with high and low $PM_{2.5}$ concentrations in the nonattainment area in October to March. The contour lines represent the potential temperature gradient ($\Delta\theta/\Delta z$) (K.100 m^{-1}) between the surface and 150 m above the ground; the red polygon indicates the nonattainment area. The community of North Pole is located in the lower right region of the nonattainment area.

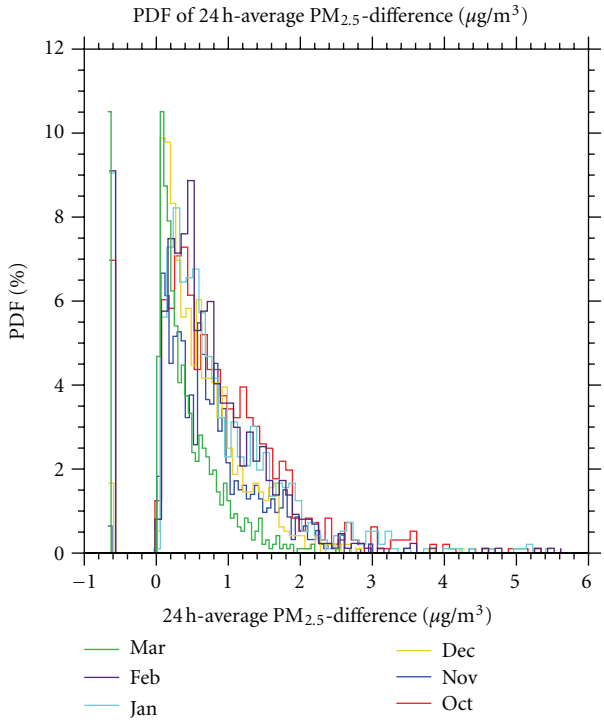

FIGURE 4: Population distribution of 24 h-average $PM_{2.5}$ difference in the nonattainment area as obtained for WSR in each month. The occurrences of all 24 h-average $PM_{2.5}$ differences <0.0 $\mu g.m^{-3}$ were summed up and their distribution is shown on the left most of the x-axis.

respectively. On average over the nonattainment area and October to March, the $PM_{2.5}$ difference was 0.6 $\mu g.m^{-3}$. This value equals to 8% (6%) of the highest (average) $PM_{2.5}$ concentration reductions over the nonattainment area.

In the nonattainment area over October to March, about 45% and 33% of the 24 h-average $PM_{2.5}$ differences fell between 0.5–1 $\mu g.m^{-3}$ and 0–0.5 $\mu g.m^{-3}$, respectively. However, for the nonattainment area the frequency distribution of the 24 h-average $PM_{2.5}$ differences varied strongly among months (Figure 4). High 24 h-average $PM_{2.5}$ differences (>3 $\mu g.m^{-3}$) only occurred 3, 2.4, and 1.2% of the time in October, January and February, respectively. In November, December, and March, more than 75% of the 24 h-average $PM_{2.5}$ differences ranged between 0 and 1 $\mu g.m^{-3}$. In October, more than 40% of the 24 h-average $PM_{2.5}$ differences in the nonattainment area exceeded 1 $\mu g.m^{-3}$.

On the nine days when the maximum 24 h-average $PM_{2.5}$ concentrations exceeded 60 $\mu g.m^{-3}$, the average 24 h-average $PM_{2.5}$ difference in the nonattainment area was 1.5–2.1 $\mu g.m^{-3}$ and the maximum 24 h-average $PM_{2.5}$ difference in the nonattainment area was 3.4–5.7 $\mu g.m^{-3}$. On these days, 60–87% (16–32%) of all grid-cells in the nonattainment area experienced 24 h-average $PM_{2.5}$ differences greater than 1 $\mu g.m^{-3}$ (2 $\mu g.m^{-3}$). On the 46 days when the maximum 24 h-average $PM_{2.5}$ concentrations ranged between 35 $\mu g.m^{-3}$ and 60 $\mu g.m^{-3}$, the average 24 h-average $PM_{2.5}$ differences were 0.7–1.5 $\mu g.m^{-3}$ and the maximum 24 h-average $PM_{2.5}$ differences were 1.9–4.0 $\mu g.m^{-3}$. About 52% of the 24 h-average $PM_{2.5}$ differences were less than 1.0 $\mu g.m^{-3}$ and 8% of all grid-cells in the nonattainment area had 24 h-average $PM_{2.5}$ differences greater than 2 $\mu g.m^{-3}$. On days with maximum 24 h-average $PM_{2.5}$ concentration lower than 35 $\mu g.m^{-3}$, the 24 h-average $PM_{2.5}$ differences were about 0.5 $\mu g.m^{-3}$ on average, and 77% of them were less than 1.0 $\mu g.m^{-3}$. On these days, only 1% of the 24 h-average $PM_{2.5}$ differences exceeded 2 $\mu g.m^{-3}$ and typically occurred in the site group.

On 111 out of the 182 days, the maximum 24 h-average $PM_{2.5}$ difference occurred within the site group. The maximum 24 h-average $PM_{2.5}$ differences typically occurred in the site group on days with calm winds (v < 0.5 $m.s^{-1}$) or on days with winds (v > 2 $m.s^{-1}$) and uniform wind direction over the nonattainment area. When the maximum difference occurred at another place in the nonattainment area, winds ranged between 0.7 and 1.2 $m.s^{-1}$ from various directions and advected pollutants from relatively strong polluted areas within the nonattainment area.

In the nonattainment area at grid-cells with strong $PM_{2.5}$ emissions (>1400 $g.km^{-2}\,h^{-1}$), the 24 h-average $PM_{2.5}$ differences strongly depended on the $PM_{2.5}$ emission reduction (R = 0.617 to 0.894, significant). At grid-cells with low $PM_{2.5}$ emissions (≤1400 $g.km^{-2}\,h^{-1}$), the 24 h-average $PM_{2.5}$ difference was less sensitive to the $PM_{2.5}$ emission reduction (R = 0.161 to 0.556) than at those with high emission rates. Instead, the meteorological conditions gained importance for the magnitude of the concentration reduction.

$PM_{2.5}$ speciation in REF hardly differed from that in WSR (<0.1%). The low changes in the partitioning among SO_4, NO_3, and other $PM_{2.5}$ species was partly due to the low emission reductions, the low availability of NH_3 and low shortwave radiation in Fairbanks during October to March.

In WSR, 1 (8), 3 (5), 2 (3), 1 (8), 0 (0), and 0 (0) exceedance days (exceedances) were avoided in October, November, December, January, February, and March, respectively, as compared to REF. Out of them eight exceedances were avoided at the grid cell holding the monitoring site. On all exceedance-days except February 8, 2009, the locations of exceedances were identical in WSR and REF. On February 8, 2009, more grid-cells experienced exceedances in WSR than REF (three versus two grid-cells) due to the close to 35 $\mu g.m^{-3}$ concentrations and slight changes in meteorological conditions due to radiation-aerosol feedbacks.

At exceedance locations, about 18.3, 9.9, 42.0, 22.1, 10.7, and 6.1% of the 24 h-average $PM_{2.5}$ differences varied between <2, 2-3, 3-4, 4-5, and >5 $\mu g.m^{-3}$, and the maximum 24 h-average $PM_{2.5}$ difference obtained on any exceedance-day was 5.7 $\mu g.m^{-3}$ (October 27, 2008). The maximum 24 h-average $PM_{2.5}$ differences on any avoided exceedance-days were between 1.7 and 2.8 $\mu g.m^{-3}$. This finding means the changeout of wood-burning devices avoided exceedance-days only on days with 24 h-average $PM_{2.5}$ concentrations slightly above 35 $\mu g.m^{-3}$.

At the grid-cell of the monitoring site the RRFs of 24 h-average $PM_{2.5}$ concentrations were 0.951, 0.950, 0.952, 0.956, 0.941, and 0.940 in October, November, December, January, February, and March, respectively. At this grid-cell,

the daily RRFs of 24 h-average $PM_{2.5}$ concentration were 0.938, 0.949, and 0.965 at the 50th, 75th, and 90th percentile, respectively. These findings suggest that the RRFs of total $PM_{2.5}$ concentrations at the grid-cell of the monitoring site were relatively consistent throughout October to March. The overall RRFs for NO_3 were 0.835, 0.893, 0.913, 0.868, 1.035, and 0.873 in October to March, and 0.866, 0.897 and 0.960 at the 50th, 75th, and 90th percentile, respectively. The RRF of NO_3 greater than 1 may be an artifact related to the very low NO_3 concentrations (<1 $\mu g.m^{-3}$). At low concentrations, the RRF becomes highly sensitive to even small concentration changes. The RRFs of NH_4 were relative consistent (~1) throughout October to March.

Similar RRFs as obtained for the grid-cell of the monitoring site were also obtained for the other grid-cells of the site group. At the other grid-cells in the nonattainment area, the RRFs of all $PM_{2.5}$ species were slightly decreased (increased) as compared to that of the grid-cell with the monitoring site when those grid-cells were located in the upwind (downwind) of the site group. For all species, the RRFs obtained at these other grid-cells in the nonattainment area varied about ±0.1 of the RRFs obtained at the grid-cell of the monitoring site. The grid-cells with the lowest RRFs, that is, lowest reduction, were typically located along the boundary of the nonattainment area and in the upwind of grid-cells with high pollution. The grid-cells along the boundary of the nonattainment area namely experienced frequently clean air advection from outside the nonattainment area. Therefore, the emission reductions related to the changeout of wood-burning devices hardly affected them. The grid-cells with the highest RRFs typically occurred inside the nonattainment area and had low 24 h-average $PM_{2.5}$ concentrations (<4 $\mu g.m^{-3}$) because the RRF tends to be more sensitive to low than to high $PM_{2.5}$ concentrations.

The benefits of the changeout of wood-burning devices on the 24 h-average $PM_{2.5}$ concentrations drastically decreased outside and downwind of the nonattainment area. At radii of 4 km, 8 km, 12 km, and 16 km downwind of the nonattainment area, the 24 h-average $PM_{2.5}$ differences were about 27.5, 13.1, 7.3, and 4.6% of the 24 h-average $PM_{2.5}$ differences obtained on average over the nonattainment area. A *t*-test showed that the 24 h-average $PM_{2.5}$ differences were significant nowhere in the domain except within the nonattainment area and some adjacent grid-cells (Figure 5).

3.5. Sensitivity Studies. WSS1 represents a large emission reduction (Figure 2) due to the high number of wood-burning devices being changed out. On average over the nonattainment area and the 14 days, the total $PM_{2.5}$ emission was 39.8% less in WSS1 than in REF for the same time. WSS2 examined the impact of pellet-stove replacement. Over the 14-day period, WSR and WSS2 yielded total $PM_{2.5}$ emission reductions of 5.6% and 6.6%, respectively.

The maximum 24 h-average $PM_{2.5}$ concentrations obtained in REF, WSR, WSS1, and WSS2 on any day of the 14d sensitivity study were 51.1, 47.6, 26.9, and 47.5 $\mu g.m^{-3}$ on October 14, 2008. The 24 h-average $PM_{2.5}$ differences of REF-WSS1 were appreciably higher than those of REF-WSR

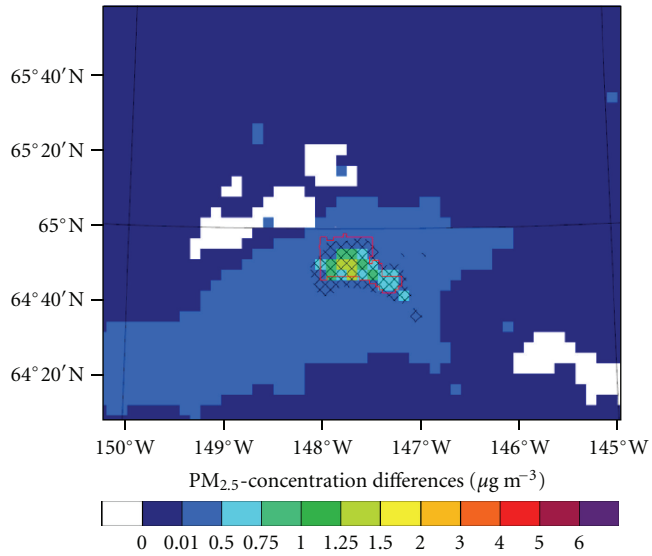

Figure 5: Zoom-in on the average differences of $PM_{2.5}$ concentration between REF and WSR for October to March. Hashed shading indicates grid cells with significant differences at the 95% or higher level of confidence.

or REF-WSS2 because the emission reduction was the highest in WSS1 (Figures 2 and 6). The maximum 24 h-average $PM_{2.5}$ differences obtained on any day in WSS1 was 24.9 $\mu g.m^{-3}$. On the contrary, the maximum 24 h-average $PM_{2.5}$ difference obtained on any of the 14 days in WSS2 was 3.6 $\mu g.m^{-3}$, which was only marginally higher than that obtained in WSR (3.5 $\mu g.m^{-3}$) for the same timeframe. About 16.7, 25.3, 18.2, 8.8, 13.1, 13.4, and 5.5% of the 24 h-average $PM_{2.5}$ differences REF-WSS1 fall within <1, 1-2, 2-3, 3-4, 4–6, 6–10, and >10 $\mu g.m^{-3}$, respectively. During the same 14d period, about 77.0 (80.2), 18.4 (17.1), 3.5 (2.3), 1.2 (0.5), and 0 (0)% of 24 h-average $PM_{2.5}$ differences of REF-WSS1 (REF-WSR) fell between <1, 1-2, 2-3, 3-4, and >4 $\mu g.m^{-3}$, respectively.

The average RRFs of the 24 h-average $PM_{2.5}$ concentrations obtained at the grid-cell of the monitoring site for WSS1, WSS2, and WSR were 0.543, 0.913, and 0.930, respectively, for the 14d episode. The RRFs of NH_4 were about 1 in all sensitivity simulations. The RRFs of NO_3 were 0.471, 0.815, and 0.818 in WSS1, WSS2 and WSR, respectively, while those of SO_4, OC, EC, and others were similar to those for $PM_{2.5}$.

The spatial variations of RRFs were within ±0.1 of the RRF at the grid-cell of the monitoring site for any species at any grid-cell in the nonattainment area for both WSS2 and WSR. On the contrary, in WSS1, the spatial variations of RRFs reached from no difference to 0.4 greater RRF values than the RRF-value at the grid-cell of the monitoring site. On six and five out of the 14 days of the sensitivity study, the highest response, that is, highest reduction in the nonattainment area, occurred at the grid-cell of the monitoring site and other grid-cells of the site group. The highest response (RRF = 0.821) occurred at the grid-cell of

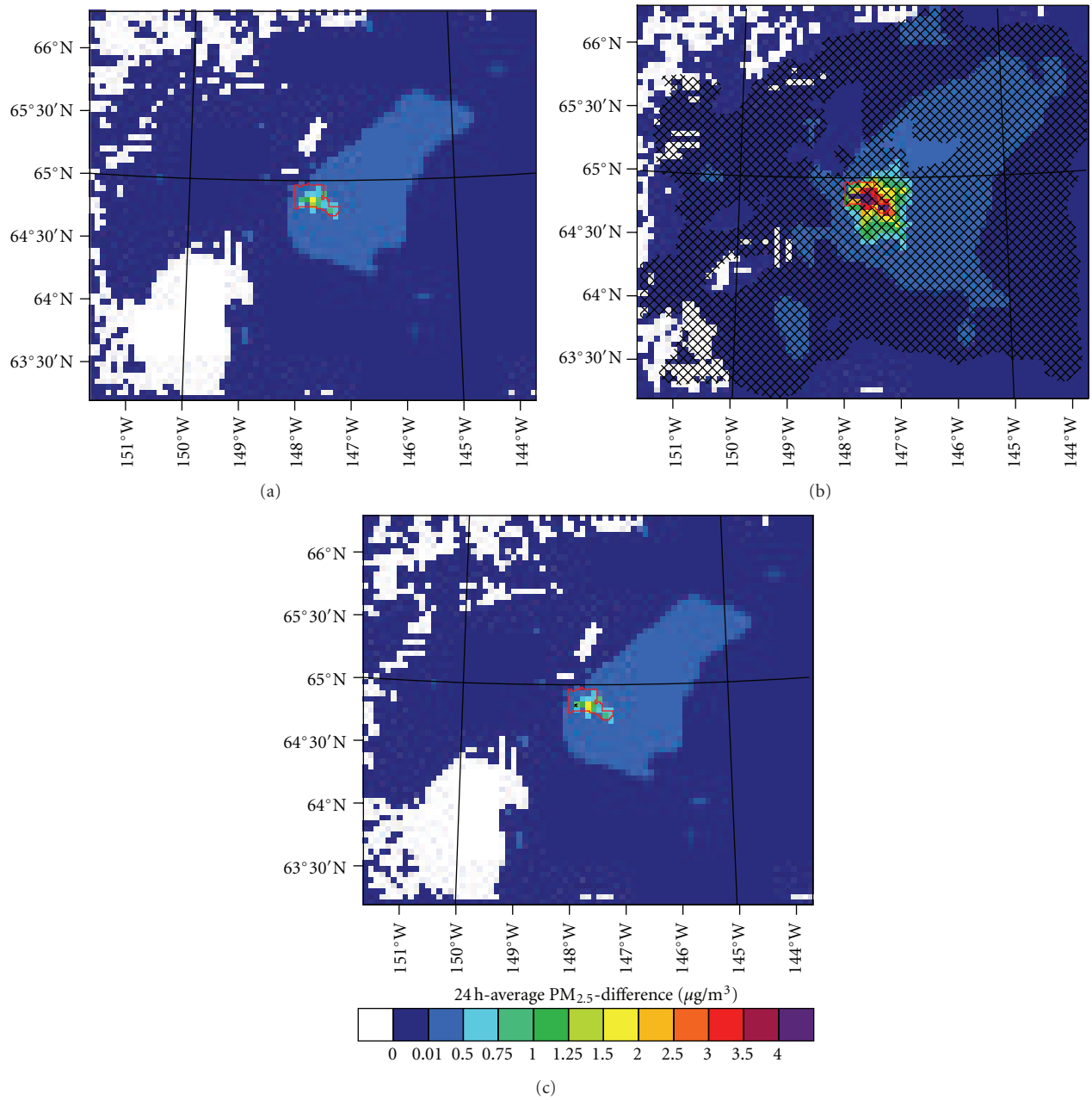

Figure 6: Like Figure 5, but for 24 h-average $PM_{2.5}$ differences (a) REF-WSR, (b) REF-WSS1, and (c) REF-WSS2 from October 1 to October 14 2008 AST.

the monitoring site on one day in WSS2. However, on no day the strongest response occurred at the grid-cell of the monitoring site in WSR.

The high number of wood-burning devices changed out in WSS1 led to avoidance of all 4 (6) exceedance days (exceedances) that occurred in REF during the same time. No exceedances were avoided in both WSS2 and WSR during these 14 days. The highest (lowest) 24 h-average $PM_{2.5}$ difference obtained at any exceedance location in WSS1 was 24.9 (16.8) $\mu g.m^{-3}$. The locations of exceedances were the same in REF, WSS2, and WSR and all occurred in the nonattainment area.

4. Conclusions

The effects of exchanging noncertified wood-burning devices with certified woodstoves on reducing the 24 h-average $PM_{2.5}$ concentrations at breathing level in the Fairbanks nonattainment area were investigated for October 1, 2008, to March 31, 2009, using results from WRF/Chem simulations. The results indicated that the assumed wood-burning device changeouts helped to reduce the 24 h-average $PM_{2.5}$ concentrations at breathing level in the nonattainment area. However, the reduction effectiveness depends on the number of wood-burning devices changed out and what kinds of devices

are changed out. The wood-burning device changeout scenario based on data reported by [2] yielded only a 3.7% $PM_{2.5}$ emission reduction from the reference scenario and consequently a low decrease of 24 h-average $PM_{2.5}$ concentrations. On average over the nonattainment area and October to March, the 24 h-average $PM_{2.5}$ differences (REF-WSR) were 0.6 $\mu g.m^{-3}$, which equals a 6% $PM_{2.5}$ concentration reduction. About 79% of the 24 h-average $PM_{2.5}$ differences were less than 1 $\mu g.m^{-3}$. This means given a design value of 44.7 $\mu g.m^{-3}$ the assumed changeout does not lead to compliance and may only reduce the number of exceedances on days with concentrations slightly higher than the NAAQS.

The magnitude of the 24 h-average $PM_{2.5}$ differences REF-WSR differed strongly among days and locations. High 24 h-average $PM_{2.5}$ differences (>3 $\mu g.m^{-3}$) often occurred in October, January, and February. Wind speed and wind direction were the key factors that governed the distribution of the maximum 24 h-average $PM_{2.5}$ difference. The magnitude of the 24 h-average $PM_{2.5}$ difference depended more on the $PM_{2.5}$ emission reduction at grid-cells having relative strong than relative low $PM_{2.5}$ emissions. The maximum 24 h-average $PM_{2.5}$ differences typically occurred in the grid-cells of the site group on days having calm wind ($v < 0.5\,m.s^{-1}$) or wind speeds exceeding 2 $m.s^{-1}$. Under other wind conditions, the maximum 24 h-average $PM_{2.5}$ differences typically occurred at grid-cells in the downwind of the site group. Based on these findings one has to conclude that mitigation is spatially heterogeneous and local emission conditions together with the meteorological conditions strongly govern the magnitude of mitigation.

The wood-burning device changeout assumed in WSR only effectively helped to avoid 7 out of 55 exceedance days that occurred in REF. Moreover, this avoidance occurred only on days with 24 h-average $PM_{2.5}$ concentration slightly above 35 $\mu g.m^{-3}$. The RRFs of $PM_{2.5}$ concentration and its major components typically varied between 0.950–0.965 and were relatively consistent throughout October to March. The lowest RRFs, that is, highest reductions, were not obtained at the grid-cell of the monitoring site but at other grid-cells in the nonattainment area. These findings support the above conclusion that the assumed changeout is not sufficient to achieve compliance. Thus, one has to conclude that the changeout of wood-burning devices may improve the air quality locally in large parts of the nonattainment area without becoming obvious at the monitoring site. Based on the relative consistency of RRF one has to conclude that wood-burning changeout provides a relative reliable reduction.

The 14d sensitive simulations assuming the number of wood-burning devices reported by [29] (WSS1) yielded up to a 39.8% $PM_{2.5}$ emission reduction as compared to the baseline simulation (REF) and a much higher 24 h-average $PM_{2.5}$ concentration reduction over the nonattainment area than WSR and WSS2. In total four of the exceedance days that were simulated in REF during these 14 days were avoided in WSS1 and the maximum 24 h-average $PM_{2.5}$ difference (REF-WSS1) at any exceedance location was 24.9 $\mu g.m^{-3}$. The relative response factors of $PM_{2.5}$ concentrations obtained at the grid-cell of the monitoring site were as high as 0.543 on average and the highest RRFs were frequently obtained at the grid-cell of the monitoring site and other grid-cells of the site group. The results of the sensitivity study WSS2 only marginally differed from those of WSR. Based on the 14d sensitivity study WSS1, one has to conclude that if the number of uncertified wood-burning devices assumed in WSS1 could be changed out, the number of exceedances in the nonattainment area could effectively be reduced. On the contrary, changing out wood-burning devices at the comparatively low numbers assumed in WSR and WSS2 seems not to be sufficient to achieve compliance with the NAAQS. Together the results of the sensitivity studies suggest that accurate knowledge on the number of noncertified devices that have to be or can be changed out is of greatest importance to assess the potential benefits of a changeout program on the 24 h-average $PM_{2.5}$ concentrations.

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