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Research Article

Potential Impacts of the Introduction of Low-Sulfur Fuel on $PM_{2.5}$ Concentrations at Breathing Level in a Subarctic City

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The effects of using low-sulfur fuel for oil-heating and oil-burning facilities on the $PM_{2.5}$ concentrations at breathing level in an Alaska city surrounded by vast areas were examined with the Weather Research and Forecasting model coupled with chemistry packages that was modified for the subarctic. Simulations were performed in forecast mode for a cold season using the National Emission Inventory 2008 and alternatively emissions that represent the use of low-sulfur fuel for oil-heating and oil-burning facilities while keeping the emissions of other sources the same as in the reference simulation. The simulations suggest that introducing low-sulfur fuel would decrease the monthly mean 24 h-averaged $PM_{2.5}$ concentrations over the city's $PM_{2.5}$ nonattainment area by 4%, 9%, 8%, 6%, 5%, and 7% in October, November, December, January, February, and March, respectively. The quarterly mean relative response factors for $PM_{2.5}$ of 0.96 indicate that with a design value of 44.7 μg/m^3 introducing low-sulfur fuel would lead to a new design value of 42.9 μg/m^3 that still exceeds the US National Ambient Air Quality Standard of 35 μg/m^3. The magnitude of the relation between the relative response of sulfate and nitrate changes differs with temperature. The simulations suggest that, in the city, $PM_{2.5}$ concentrations would decrease stronger on days with low atmospheric boundary layer heights, low hydrometeor mixing ratio, low downward shortwave radiation, and low temperatures.

1. Introduction

In 2009, Fairbanks—a city in Alaska that is the only precursor-source area within a region of hardly any anthropogenic emissions—was designated a $PM_{2.5}$-nonattainment area (NAA) due to its frequent exceeding of the 24 h-average National Ambient Air Quality Standard (NAAQS) of 35 μg/m^3 for particulate matter of diameter less than 2.5 μm ($PM_{2.5}$) during past winters [1]. High concentrations of $PM_{2.5}$ suspended in the urban air are health adverse [2] and have led to increased hospital admissions for cerebrovascular and respiratory diseases in Fairbanks [3].

Fairbanks has hills to the North, East, and West (Figure 1) that along with strong inversions from radiative cooling and calm winds (<0.5 m/s at 10 m) limit the horizontal and vertical exchange of air. Extremely low temperatures ($\leq -20^\circ$C at 2 m) and the long dark nights cause high emissions from traffic, power generation, and heating during the cold season (October to March) that lead to accumulation of PM and other pollutants under the inversion [1]. Observations combined with trajectory and air-quality modeling studies showed that advection of pollution plays no role for Fairbanks' $PM_{2.5}$ exceedances in winter [4–6]. Fairbanks is the only city within 578 km radius; that is, local emissions are the main contributor to $PM_{2.5}$ concentrations [5, 7].

$PM_{2.5}$ can be emitted directly into the atmosphere or formed in the atmosphere by gas-to-particle conversion [8, 9]. Emitted gases, such as reactive organic gases, can be oxidized at sufficiently low vapor pressure to form secondary organic aerosols. Precursor gases such as NH_3 (ammonia), NO_x (= NO+NO_2 sum of nitric oxide and nitrogen dioxide), and sulfur dioxide (SO_2) are oxidized and form inorganic

aerosols. Fuel combustion releases SO_2 into the atmosphere where it can contribute to sulfate formation. Sulfate besides organic aerosol is the second major component of atmospheric aerosols in the Fairbanks NAA [7].

In the presence of reactive radicals and water vapor, SO_2 oxidation produces sulfuric acid (H_2SO_4). Since H_2SO_4 has a very low vapor pressure, it is assumed to be in the aerosol form under all atmospheric conditions; the sulfate-related aerosol acidity may be further neutralized by NH_3 to form ammonium sulfate aerosol ($(NH_4)_2SO_4$) [10, 11]. Ammonia can also neutralize nitric acid (HNO_3), which is the product of oxidized NO_x, and form ammonium nitrate aerosol (NH_4NO_3). The sulfate aerosol scatters radiation, can also be dissolved and act as cloud-condensation nuclei, and consequently may alter cloud albedo [12, 13].

To improve air quality and reduce $PM_{2.5}$ concentrations, various countries (e.g., Canada and countries of the European Union) introduced regulations and/or incentives to lower fuel-sulfur content in heating oil. Residential heating oil (number 2 fuel oil), which has an average sulfur content of about 2,500 ppm, is normally used for residential and commercial heating and power generation in Fairbanks. Thus, reductions of precursor SO_2 emission can decrease the $PM_{2.5}$ mass. However, the response to the emission reduction might be nonlinear; in the eastern United States, for instance, a reduction of SO_2 emissions could reduce sulfate concentrations by 50%, but the potential increase of particulate nitrate may decrease the effectiveness of reducing the annual average $PM_{2.5}$ concentrations by up to 24% [14]. The reduction of SO_2 emissions may increase particulate nitrate, as the replacement of one molecule of ammonium sulfate by two molecules of ammonium nitrate increases the total PM mass; this replacement of sulfate by nitrate can increase at low temperatures [15, 16] such that they occur in the cold season in Fairbanks.

One mitigation strategies discussed for Fairbanks is to reduce the sulfur content in fuel used for oil-fired furnaces and facilities. With a design value for 2008 of 44.7 $\mu g/m^3$, an emission-control measure to be efficient has to reduce the $PM_{2.5}$ concentrations by about 22%. Note that a design value describes the air-quality status relative to the NAAQS expressed as a concentration instead of an exceedance.

Fairbanks' low insolation, temperatures, moisture, and wind speeds in winter and the frequent existence of inversions provide quite different environmental conditions for gas-to-particle conversion than found in the eastern US. Since low temperatures favor nitrate formation [15, 16], using low-sulfur fuel may not provide as large reductions as found for the eastern US. The low humidity also hinders particle growth to PM_{10} (PM with diameter >10 μm).

This study tests the hypothesis that, under the meteorological conditions during the cold season in Fairbanks, reducing the fuel-sulfur content is not sufficient to achieve the required reduction. In doing so, we turn to numerical modeling as it permits us to assess the response of $PM_{2.5}$ concentrations at breathing level under the same meteorological conditions. Since Fairbanks is the only major anthropogenic emission source within the area, responses to any local emission-control measures are not diluted by advection of anthropogenic pollutants. We examined the potential effects of utilizing low-sulfur fuel for power generation and heating on the $PM_{2.5}$ concentrations at breathing level in Fairbanks by using the Weather Research and Forecasting model [17] inline coupled with chemistry packages (WRF/Chem; [18]) with the modifications for the subarctic introduced by [7]. WRF/Chem had recently been successfully used to assess the response to the emission controls implemented for the 2008 Olympic Games [19, 20].

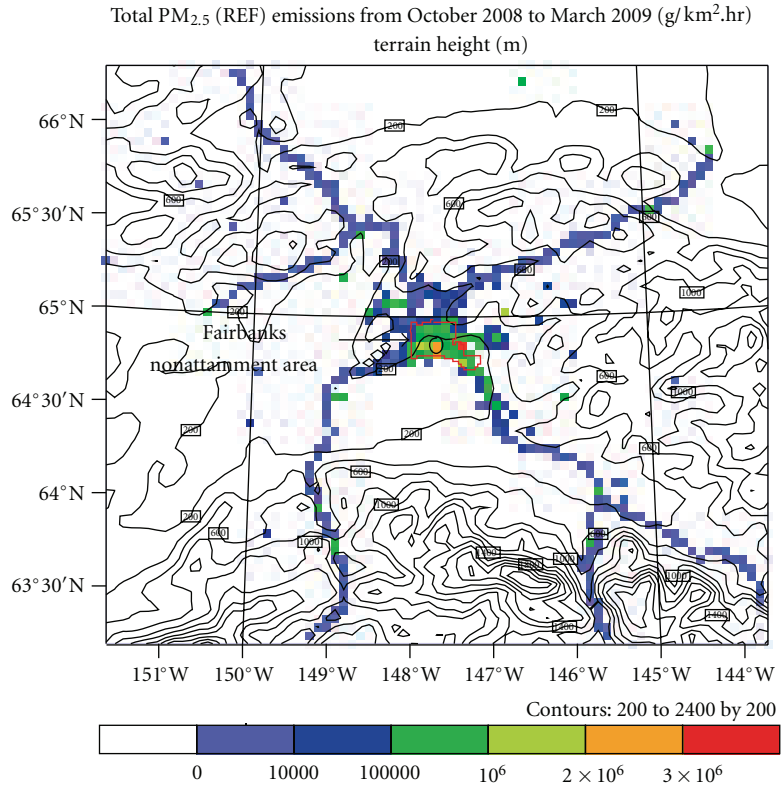
2. Experimental Design

2.1. Model Setup. We used the physical and chemical packages as described in [6]. This model setup includes the six water-class cloud microphysical scheme [21], the further-developed Grell-Dévényi cumulus-ensemble scheme [22] in its 3D version, the Goddard shortwave radiation scheme [23], and the radiative transfer model for long-wave radiation [24]. The processes in the atmospheric boundary layer (ABL) and sublayer were considered following Janjić [25]. The exchange at the surface-atmosphere interface is determined using a modified version of the Rapid Update Cycle land-surface model [26]. The chemistry package considered radiative feedback from aerosols [27]. The gas-phase chemistry by Stockwell et al. [28] with photolysis frequencies calculated following [29] was used. Aerosol dynamics, physics, and chemistry were described by the Modal Aerosol Dynamics Model for Europe (MADE; [30]) and the Secondary ORGanic Aerosol Model (SORGAM; [9]). For secondary organic formation, WRF/Chem considers the OH-radical, the nitrate-radical, and ozone as oxidants for ROG [9]. For aerosol inorganic chemistry, the model includes sulfate, ammonium, and nitrate for thermodynamic of gas/aerosol equilibrium. Dry deposition of trace gases was determined in accord with Wesely [31], with the modifications by [7].

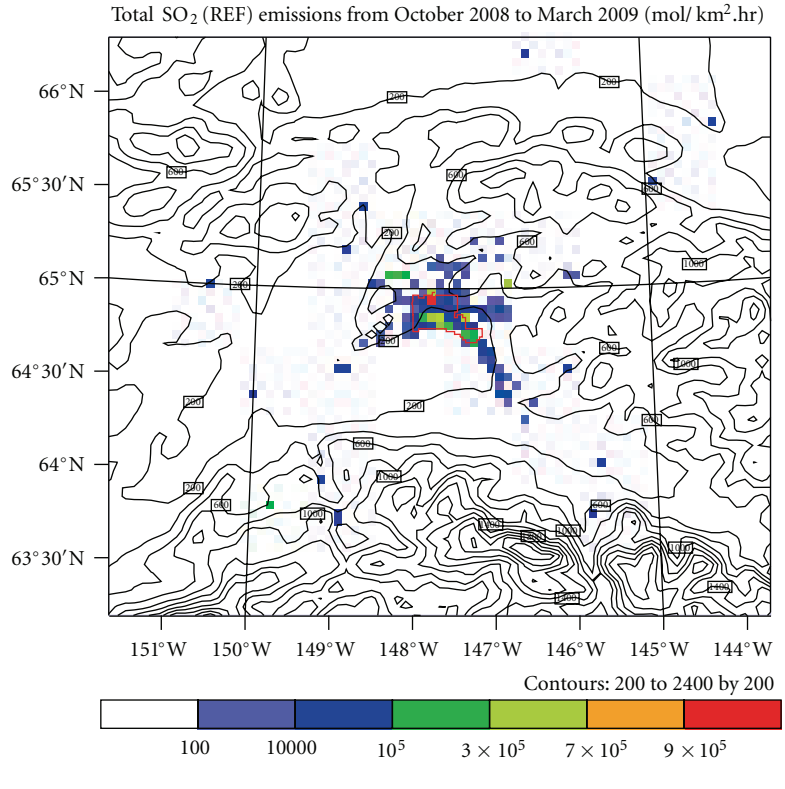
2.2. Simulations. The area for our analysis encompasses 80 × 70 grid points with a grid increment of 4 km centered over Fairbanks (Figure 1). The vertically stretched grid had 28 layers up to 100 hPa. The initial meteorological conditions, including snow and soil variables, were downscaled from the 1° × 1°, 6 h-resolution National Centers for Environmental Prediction global final analyses. This meteorological data was also downscaled as lateral boundary conditions.

The vertical profiles of Alaska-typical background concentrations served to initialize the chemical fields. Since Fairbanks is the only city and major emission source [4–6], Alaska background concentrations served as lateral boundary conditions.

The simulations were performed in forecast mode for October 1, 2008 0000UTC to April 1, 2009 0000UTC, and analyzed for October 1 to March 31 Alaska Standard Time (AST = UTC-9 h). The meteorological fields were initialized every five days. The chemical distributions at the end of each simulation served as the initial contributions for the next simulation.



(a)



(b)

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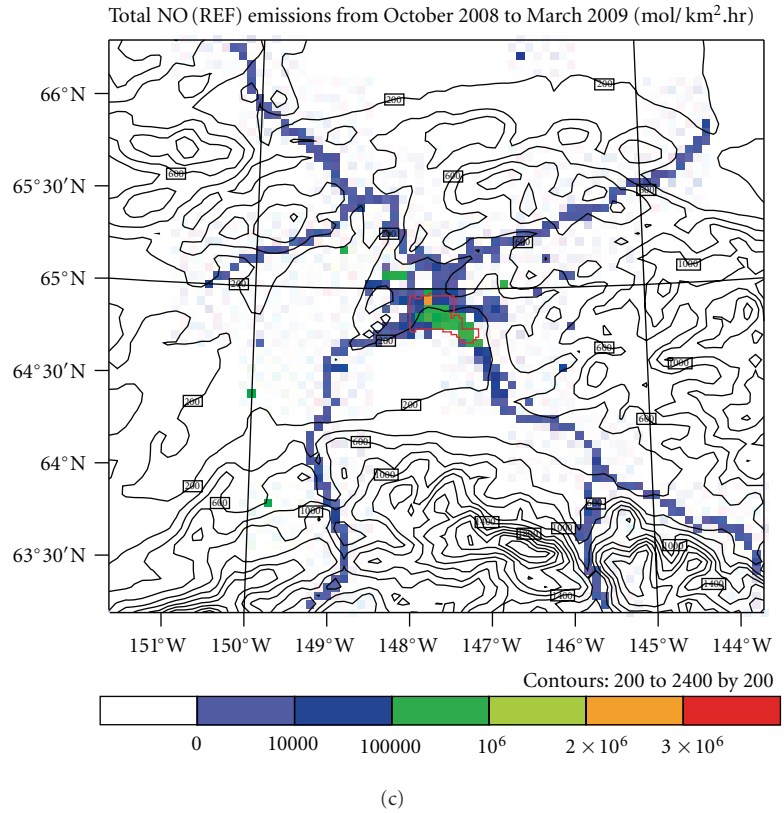


(c)

Figure 1: Total emission of (a) $PM_{2.5}$, (b) SO_2, and (c) NO_x from October 1, 2008 to March 31, 2008 (color) from all layers in the analysis domain. Terrain height is superimposed (contour lines). In (a), the red polygon and black circular shape indicate the boundaries of the Fairbanks nonattainment area and the location of the official $PM_{2.5}$-monitoring site, respectively.

2.3. Emissions. Biogenic emissions were calculated depending on temperature and radiation flux density [32]. Anthropogenic emissions were obtained from the National Emission Inventory of 2008 with updates for point-source and nonpoint source sectors using data from point-source facility operators and local agencies if available. Otherwise, a 1.5% increase per year from the point-source emissions of the previous inventory was assumed. The anthropogenic emissions were allocated according to the source-specific activity in space (e.g., point-source coordinates, population and traffic density) and time (month, day of-the week, hour). Empirical functions [6, 33, 34] were used to allocate emissions from power generation, commercial and residential heating, and traffic (cold-starts) temperature dependent. These parameterizations ensured higher (lower) emissions on days with daily mean temperatures below (above) the 1971–2000 average. The temperature-dependency used the downscaled final analysis temperatures to avoid that errors in WRF/Chem-predicted temperatures affect the anthropogenic emissions.

In the reference simulation (REF), emissions from oil-burning facilities and furnaces represent emissions with the current sulfur content. The mitigation simulation (LSF) assumed the use of low-sulfur fuel for these sources. A reduction of sulfur in fuel from 2500 to 500 ppm means emission reductions from oil furnaces, oil-burning facilities, and power generation of 75%, 80%, and 10% for SO_2, PM and NO_x, respectively; the decrease of NO_x emissions is due to the reduced nitrogen content of low-sulfur fuel [35]. Following [35] we assumed no reduction for the emissions of volatile organic compounds (VOCs) and CO. VOCs include all alkanes, alkenes, aromatics, organic acid, and carbonyl groups [7].

2.4. Analysis. Our analysis focused on the changes in precursor emissions of SO_2, and NO and their effect on the simulated concentrations and composition of $PM_{2.5}$. We tested the hypothesis that the use of low-sulfur fuel does not affect the $PM_{2.5}$ concentrations using a t-test. The word "significant" is used only when data pass this test at the 95% confidence level. To compare the simulation results in a relative rather than absolute sense, we calculated the relative response factors (RRFs) as the ratio of the 24 h-average concentration obtained by LSF to that obtained REF. Multiplication of the RRF with the design value provides the new design value that represents the conditions that would be found if the measure was in place.

The thermodynamic equilibrium between the gas phase and particle-phase shifts toward the gas-phase when temperature increases and vice versa. Water in the atmosphere can change the activity of organic substances [36] and affect the phase transition for inorganic aerosols. As humidity decreases, drops evaporate, and solid particles are formed.

Table 1: Total emissions of REF (first value) and LSF (second value) and percent reduction (in brackets) in the nonattainment area, and monthly mean temperatures (T_{mon}) and frequency of days with temperatures lower than the 1971–2000 mean (T_{30}). Bold values indicate significant changes.

	October	November	December	January	February	March	October to March
$PM_{2.5}$ (tons)	**4.34** | **3.86** (−11%)	2.83 | 2.30 **(−19%)**	2.92 | 2.44 (−16%)	3.69 | 3.21 (−13%)	2.84 | 2.44 (−14%)	3.05 | 2.63 (−14%)	23 | 19.8 (−14%)
$PM_{10} \times 10^4$ (mol)	**1.48** | **1.31** (−11%)	1.26 | 1.04 **(−18%)**	1.25 | 1.04 (−17%)	1.38 | 1.18 (−15%)	1.13 | 0.97 (−14%)	1.22 | 1.05 (−13%)	8.99 | 7.70 (−14%)
$SO_2 \times 10^5$ (mol)	9.39 | 6.59 **(−30%)**	8.13 | 6.61 (−19%)	7.96 | 6.33 (−21%)	**9.42** | **7.34** (−22%)	7.13 | 5.57 (−22%)	7.49 | 5.87 (−22%)	57.4 | 44.4 (−23%)
$NO \times 10^5$ (mol)	14.4 | 14.1 **(−3%)**	13.9 | 13.8 (−1%)	13.5 | 13.2 (−2%)	**15.3** | **15.2** (−1%)	11.7 | 11.7 (+<1%)	12.4 | 12.2 (−2%)	94.2 | 92.8 (−1%)
T_{avg} (°C)	−8.1	−14.7	−17.5	−18.2	−13.7	−13.5	
$T_{30y\text{-average}}$ (°C)	−4.4	−16.7	−21.1	−23.3	−19.9	−23.9	
Frequency of days, with $T_{mon} < T_{30}$ (%)	61	23	29	42	17	6	

These particles remain solid until the relative humidity increases to the deliquescence [11]. In view of these meteorological effects on particle formation, we examined how differences between REF and LSF change with the meteorological conditions as well.

3. Reference Simulation

3.1. Emissions. In REF, the total monthly $PM_{2.5}$ emissions in the NAA were 4.34, 2.83, 2.92, 3.69, 2.84, and 3.05 tons in October to March (Table 1). Except for October monthly mean temperatures exceeded their 30-year average (1971–2000). Although October was the warmest month of winter 2008/09, it was much colder than the 30-year average and had the highest frequency of daily mean temperatures below that average. Thus, the temperature dependency of the emissions led to higher emissions than they would have occurred in October with normal mean temperatures. Consequently, October had the highest $PM_{2.5}$ emissions in REF and LSF, and the lowest relative $PM_{2.5}$ emission reduction. January had high total emissions as it was the coldest month. In March, recreational use of snow machines as the temperature and daylight hours increased led to an increase in $PM_{2.5}$ emissions.

3.2. Evaluation. The evaluation of REF by data from 23 surface meteorological sites, 9 $PM_{2.5}$ sites, 4 specification sites, and mobile $PM_{2.5}$-concentration and temperature measurements provides on average over October to March biases of 2 m-temperature, 2 m-dewpoint temperature, sea-level pressure, 10 m-wind-speed and direction of 1.3 K, 2.1 K, −1.9 hPa, 1.55 m/s, and −4°, respectively [6]. The wind errors explain some of the underestimation of the $PM_{2.5}$ concentrations. The overestimation of temperatures led to biases of 0.5 K, 0.8 K, 2 K, 2.6 K, 1.6 K, and 0.3 K and root-mean square errors (RMSEs) of 3.8 K, 4.8 K, 6.1 K, 4.3 K, 5.2 K, and 4.1 K in October to March, respectively; 2 m-dewpoint-temperature RMSEs were less than 5 K except November (6.2 K). Performance was better on relatively warmer than colder days and in the rural than urban areas [6]. The mobile temperature measurements indicated that, in the NAA, simulated temperatures were about 1.4, 2.4, 1.2, and 2.2 K too high in November, December, January, and February, and 0.9 K too cold in March [6]. No mobile measurement data existed for October.

In Fairbanks during winter, the low-incoming solar radiation yields to radiative cooling, low daily mean temperatures, and inversions [1, 37, 38]. The strength of low level inversions depends on the net radiation loss and marginal to no cloudiness [37]. WRF/Chem well captured this typical behavior of inversion events, low ABL-heights with usually hardly any cloud or ice particles as indicated by low integrated hydrometeor mixing ratio (e.g., Figure 2).

The $PM_{2.5}$ evaluation used the fractional bias $FB = (2/N)\sum_{i=1}^{N}((C_s - C_o)/(C_s + C_o)) \times 100\%$, fractional error $FE = (2/N)\sum_{i=1}^{N}|(C_s - C_o)/(C_s + C_o)| \times 100\%$, normalized mean bias $NMB = (\sum_{i=1}^{N}(C_s - C_o)/\sum_{i=1}^{N} C_o) \times 100\%$, and normalized mean error $NME = (\sum_{i=1}^{N}|C_s - C_o|/\sum_{i=1}^{N} C_o) \times 100\%$. On average over October to March and all sites, the FB, FE, NMB and NME for 24 h-average $PM_{2.5}$ concentrations were 22%, 67%, 13%, and 71%, respectively, which is slightly weaker than the performance found for various air-quality model applications in midlatitudes [6]. WRF/Chem performed best for $PM_{2.5}$ concentrations between 15 and 50 $\mu g/m^3$. Performance was best for organic carbon (OC) followed by sulfate. Ammonium was strongly underestimated. The errors in predicted $PM_{2.5}$ were due to errors in emissions and simulated meteorological conditions (mistiming of fronts, underestimation of inversion strength, overestimation of wind speed), measurement errors and, on some days in March, the chemical boundary conditions [6].

For application in air-quality mitigation studies, a model must perform well around the NAAQS and the design value. Since (1) WRF/Chem achieved the best results for $PM_{2.5}$ concentrations between 15 and 50 $\mu g/m^3$, (2) performed acceptable for sulfate species in $PM_{2.5}$, and (3) LSF and REF

Table 2: Monthly mean of 24 h-averaged $PM_{2.5}$, PM_{10}, sulfate, and nitrate concentrations in the nonattainment area as obtained with REF (first value) and LSF (second value) and percent change (in brackets). Reductions are presented negative. Bold values indicate significant changes.

	October	November	December	January	February	March	October to March
$PM_{2.5}$ ($\mu g/m^3$)	**13.0** | **12.5** (−4%)	11.0 | 10.0 (−**9**%)	9.2 | 8.5 **(−8%)**	11.0 | 10.4 (−6%)	9.8 | 9.3 (−5%)	5.7 | 5.3 (−7%)	9.5 | 8.9 (−6%)
PM_{10} ($\mu g/m^3$)	**29.4** | **28.6** (−3%)	28.2 | 25.9 (−**8**%)	24.1 | 22.4 (−7%)	26.6 | 25.2 (−6%)	24.2 | 23.0 (−5%)	15.3 | 14.3 (−7%)	23.6 | 22.2 (−6%)
Sulfate ($\mu g/kg \cdot$dryair)	**2.15** | **2.07** (−3%)	1.79 | 1.64 (−**8**%)	1.49 | 1.38 (−7%)	1.76 | 1.67 (−5%)	1.61 | 1.52 (−6%)	0.98 | 0.91 (−7%)	1.56 | 1.47 (−6%)
Nitrate ($\mu g/kg \cdot$dryair)	0.09 | 0.10 (+3%)	**0.12** | **0.11** (−**10**%)	0.05 | 0.04 (−**10**%)	0.06 | 0.06 (−8%)	0.06 | 0.06 (+10%)	0.02 | 0.02 (−10%)	0.06 | 0.06 (−4%)
Ammonium $\times 10^{-3}$	2.64 | 2.75 (+4%)	2.84 | 2.83 (0%)	1.71 | 1.71 (0%)	2.09 | 2.28 (+9%)	**2.50** | **2.34** **(−6%)**	1.36 | 1.35 (+1%)	2.19 | 2.21 (−1%)
Elemental carbon	0.92 | 0.89 (−4%)	**0.77** | **0.70** (−**9**%)	0.62 | 0.58 (−8%)	0.75 | 0.71 (−6%)	0.68 | 0.64 (−6%)	0.40 | 0.37 (−7%)	0.69 | 0.65 (−6%)
Organic carbon	4.72 | 4.55 (−4%)	**3.91** | **3.57** (−**9**%)	3.19 | 2.94 (−8%)	3.83 | 3.61 (−6%)	3.49 | 3.28 (−6%)	2.02 | 1.87 (−7%)	3.53 | 3.31 (−6%)

are affected by errors in the same way; that is, errors may cancel out in the differences, we can expect that WRF/Chem is suitable to assess the impact of low-sulfur fuel on $PM_{2.5}$ concentrations. Furthermore, we discuss the results in a relative sense by means of RRF and relative responses.

3.3. Urban Air Quality and Meteorology. On average over the analysis domain and October to March, the simulated 24 h-average $PM_{2.5}$ concentration was 0.4 $\mu g/m^3$ in the lowest layer in REF. In the NAA, $PM_{2.5}$ concentrations were highest. Here, in REF, the monthly averages of 24 h-average $PM_{2.5}$-concentrations were 13.0, 11.0, 9.2, 11.0, 9.8, and 5.7 $\mu g/m^3$ for October to March, respectively.

According to the model, in the NAA, $PM_{2.5}$ strongly depended on temperature, relative humidity, and wind speed (Figure 2). Low temperatures and high emissions led to increased gas-to-particle conversion (e.g., the peak of simulated $PM_{2.5}$ during the cold snaps at the end of October or the beginning of January and March). The highest and second highest sulfate concentrations were simulated for October and January (Table 2). This behavior of WRF/Chem well agrees with regression analysis of observational data [1] that identified low temperatures as one of the main factors for increased 24 h-average $PM_{2.5}$ concentrations in the NAA.

Atmospheric moisture affects aerosol formation, and its impact varies with temperature [37]. WRF/Chem simulated low hourly $PM_{2.5}$ concentrations in the NAA when simulated vapor pressure and relative humidity were high which well reflects the typically observed hygroscopic growth of particles under these conditions [1].

Observations showed that winds with daily average speeds >0.5 m/s dilute the $PM_{2.5}$ concentrations, while calm winds (<0.5 m/s) build up the $PM_{2.5}$ concentrations in the NAA [1]. WRF/Chem showed this behavior during October to March (Figure 2). During these months, the monthly average simulated wind speeds in the NAA were 2.27, 1.93, 2.68, 2.62, 2.18, and 3.74 m/s, respectively. The relatively stronger wind simulated for March than in other months resulted in the lowest monthly average of 24 h-average simulated $PM_{2.5}$ concentrations, and aerosol compositions including nitrate, ammonium, sulfate, EC, and OC in the NAA (Table 2).

Since Fairbanks is the only major emission source, advection generally brings clean air, except when the aged Fairbanks pollution is advected back into the NAA [6]. Such advection occurred 27 times during winter 2008/09. October, November, and February had the highest frequency of advection of aged Fairbanks urban air (6-7 times/month).

Our analysis showed that the simulated low ABL-heights (<100 m) limited the vertical mixing tremendously, resulting in high $PM_{2.5}$ concentrations. For example, the peak of $PM_{2.5}$ at the beginning of January occurs when the ABL height is lower than 100 m for many days. November had the lowest monthly average simulated ABL-height of winter 2008/09 and the highest simulated monthly nitrate and ammonium concentrations and the second highest simulated monthly concentrations of PM, sulfate, EC, and OC (Table 2).

4. Low Sulfur Fuel

4.1. Emissions. The assumed emission reductions due to low sulfur-fuel usage differ among hours, days, and months as the emissions related to oil-burning furnaces and facilities were prepared temperature dependent for use in WRF/Chem. Compared to REF, assuming that the rates given by [35] for low sulfur fuel reduced the total $PM_{2.5}$ emissions in the NAA by 11%, 19%, 16%, 13%, 14%, and 14% for October to March, respectively, with similar reductions in PM_{10} emissions. On average over October to March, the PM-emission reduction would be 14%. On average, over these months, the total SO_2 emission would be reduced by ~23% (Table 1). Emissions from all other sources than oil-furnaces and oil-burning facilities were identical to those in REF.

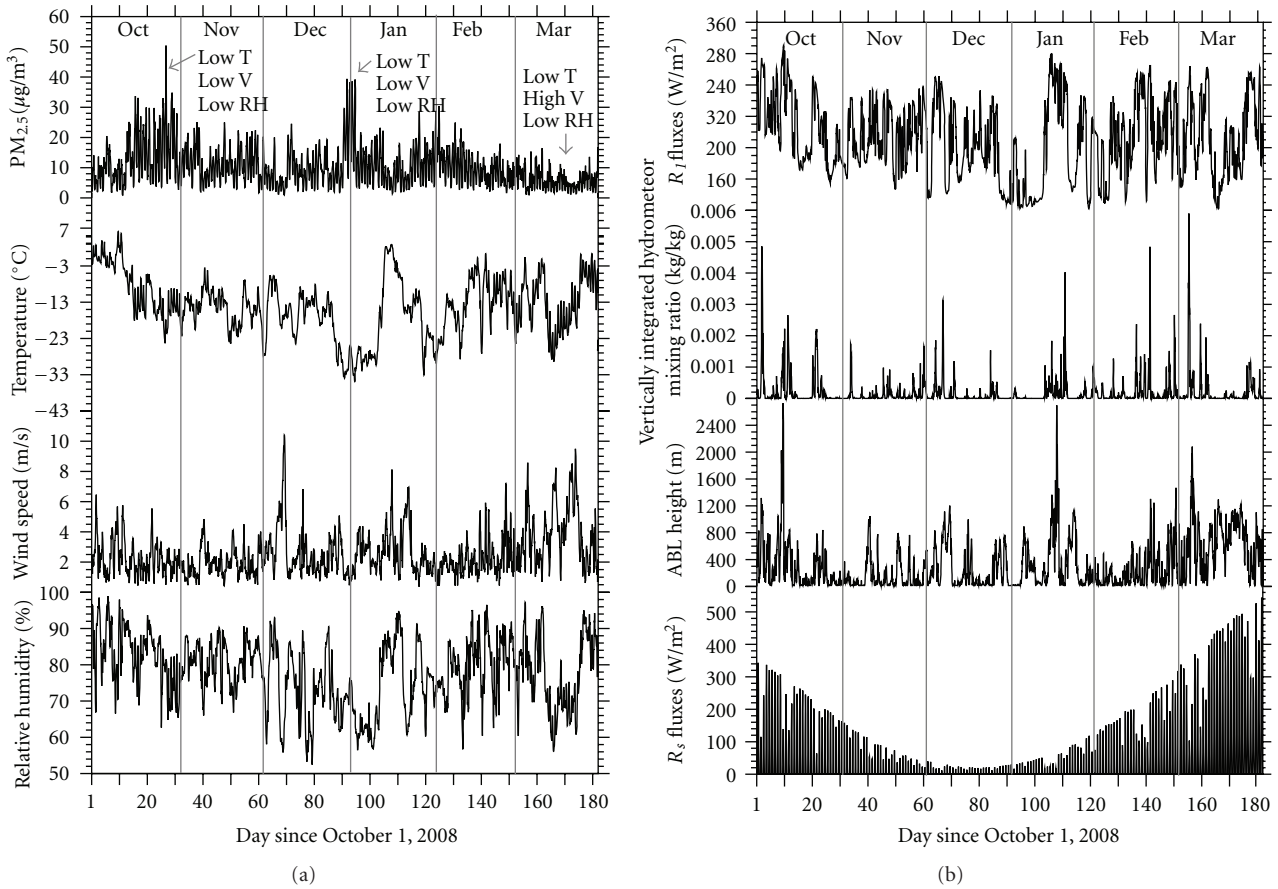


Figure 2: Temporal evolution of simulated hourly (a) $PM_{2.5}$-concentration, temperature, wind-speed, relative humidity, and (b) downward long-wave radiation, hydrometeor mixing ratios, ABL-height and downward shortwave radiation averaged over the Fairbanks nonattainment area for each of the 182 simulation days.

4.2. Impacts on Urban Air Quality. On average over the first layer of the analysis domain and October to March, the assumed usage of low sulfur fuel reduced the simulated $PM_{2.5}$ concentrations by 5%. In LSF, the hourly $PM_{2.5}$-concentrations significantly decreased in some areas in the first layer as compared to REF (Figure 3). In the NAA, in response to the assumed emission changes, the simulated $PM_{2.5}$-concentrations decreased by 0.5 $\mu g/m^3$, 1.0 $\mu g/m^3$, 0.7 $\mu g/m^3$, 0.6 $\mu g/m^3$, 0.5 $\mu g/m^3$, and 0.4 $\mu g/m^3$ in October to March, respectively, and by 0.6 $\mu g/m^3$ on average over these months. These simulated $PM_{2.5}$-concentration reductions were significant in November, December, and March (Figure 3). The relative monthly mean of 24 h-average $PM_{2.5}$-concentration reductions would vary between 4% and 9% (Table 2). At the grid cell of the monitoring site, the October to March monthly averaged 24 h-average $PM_{2.5}$ concentrations decreased from 40.2, 30.3, 25.8, 33.9, 27.1, and 17.1 $\mu g/m^3$ in REF, respectively, to 39.2, 28.6, 24.4, 32.7, 26.0, and 16.2 $\mu g/m^3$ in LSF, that is 1.2 $\mu g/m^3$ (~4%) on average.

The simulations suggested that introduction of low sulfur fuel would reduce the number of exceedance days (days with 24 h-average $PM_{2.5}$-concentrations >35 $\mu g/m^3$). The simulated number of exceedance days went down from 20, 10, 5, 15, and 5 days to 19, 8, 4, 14, and 5 days for October to February, respectively. No exceedances were simulated for March. The highest frequency of exceedance days (52 in REF, 47 in LSF) was simulated for the grid-cell that holds the official monitoring site. On most of these days, this grid-cell had the highest 24 h-average $PM_{2.5}$ concentrations in the NAA.

The 24 h-average $PM_{2.5}$-concentration difference between REF and LSF for each of the 182 simulation days was calculated and sorted from highest to lowest. The investigation of the top 20% (37 days) showed that 14 of the days with the highest concentration differences occurred in November. In November, wind speeds and ABL height, on average, were the lowest of all months (e.g., Figure 2, Table 3). Thus, pollutants accumulated and had enough time for chemical conversion. The changed composition and reduced amount of precursors in LSF as compared to REF, and hence, became most effective due to the relatively long retention of pollutants in the NAA in November. November had the highest monthly average $PM_{2.5}$-emission reduction and concentration reduction (Tables 1 and 2). Of the 20% days with the lowest concentration differences, 14 days occurred in March. March had the lowest difference between REF- and LSF-simulated $PM_{2.5}$ concentrations. WRF/Chem (correctly)

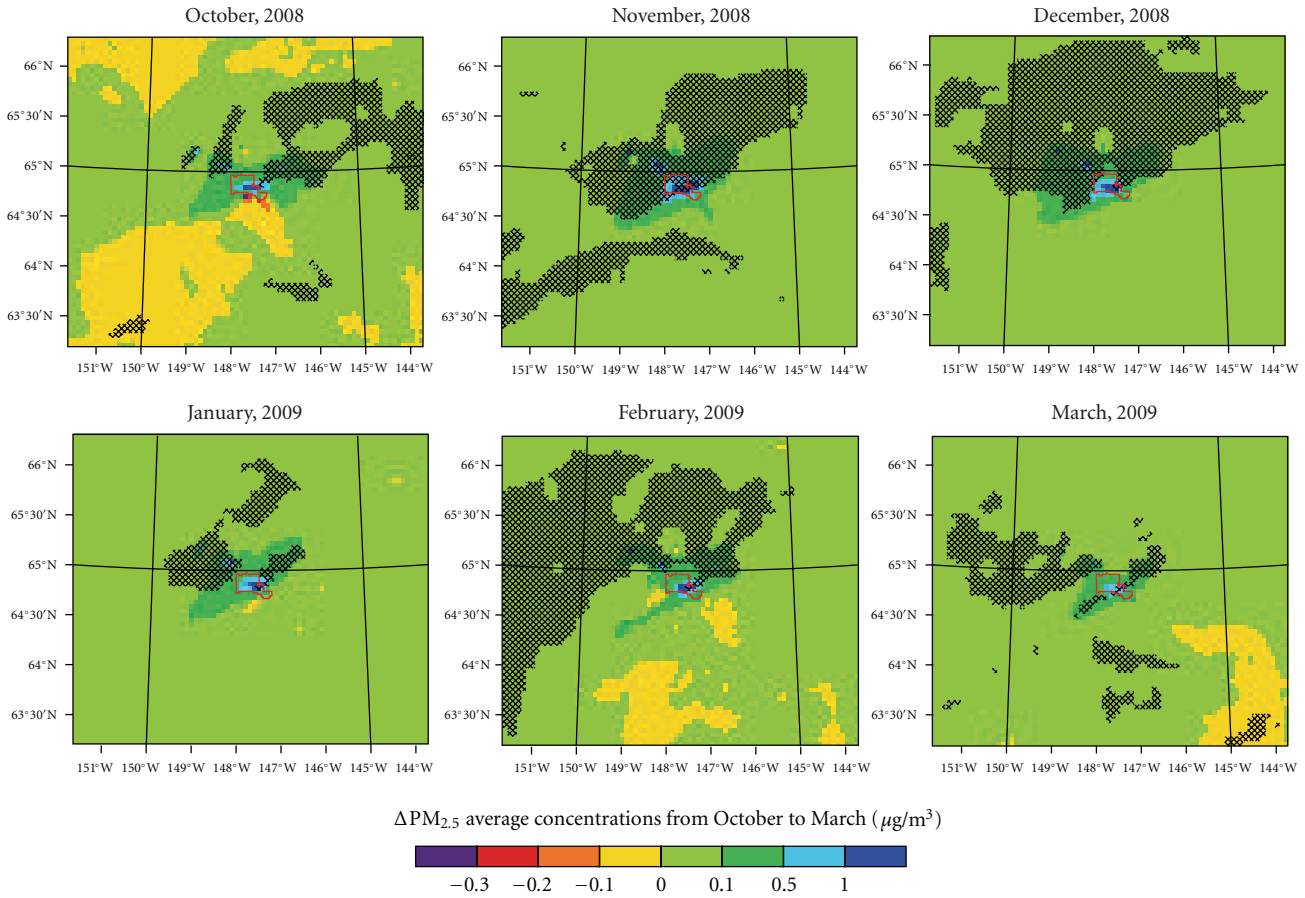


Figure 3: Difference REF-LSF of monthly averaged 24 h-average $PM_{2.5}$ concentrations (color) from October to March. The hashed shading indicates significant (95% or higher confidence level) differences. The red polygon indicates the boundaries of the Fairbanks nonattainment area.

Table 3: Monthly average of near-surface air temperature (T), dewpoint temperature (T_d), wind speed (v), relative humidity (RH) and downward shortwave radiation (R_s), ABL-height (h) and sea-level pressure (SLP), and precipitation (P) in the nonattainment area as obtained by REF (first value) and LSF (second value). Relative differences are in brackets. The letters L and N represent changes <0.001 and no change, respectively.

	October	November	December	January	February	March
T (°C)	−8.1 | −8.5 (−0.3)	−14.7 | −14.7 N	−17.5 | −17.5 (−L)	−18.2 | −18.2 (−L)	−13.7 | −13.9 (−0.2)	−13.5 | −13.1 (+0.4)
T_d (°C)	−10.5 | −10.8 (−0.3)	−17.3 | −17.3 N	−21.2 | −21.2 (−L)	−21.7 | −21.7 (−L)	−16.6 | −16.8 (−0.2)	−16.6 | −16.2 (+0.5)
v (m/s)	2.27 | 2.23 (−0.04)	1.93 | 1.93 N	2.68 | 2.68 (+L)	2.62 | 2.62 (+L)	2.18 | 2.17 (−0.01)	3.74|3.74 (−L)
RH (%)	81 | 81 (+L)	79 | 79 N	72 | 72 N	72 | 72 (−L)	78 | 78 (+L)	76 | 76 (+L)
R_s (W/m^2)	50 | 51 (+1)	12 | 12 N	2 | 2 N	8 | 8 (+L)	38 | 38 (L)	103 | 108 (+5)
R_L (W/m^2)	229 | 227 (+2)	215 | 215 N	196 | 196 (−L)	194 | 194 (−L)	215 | 215 (+L)	212 | 212 (−L)
h (m)	306 | 284 (−21)	157 | 157 N	258 | 258 (L)	340 | 340 (L)	237 | 233 (−4)	622 | 630 (+8)
SLP (hPa)	1006.8 | 1006.9 (+0.1)	1005.9 | 1005.9 N	1018.6 | 1018.6 N	1013.0 | 1013.0 N	1015.2 | 1015.2 (+0.0)	1012.3 | 1012.2 (−0.1)
P (mm)	0.5 | 1.4 (+0.9)	0.4 | 0.4 N	0.6 | 0.6 N	0.5 | 0.5 (+L)	0.7 | 0.9 (+0.2)	0.6 | 0.6 (+L)

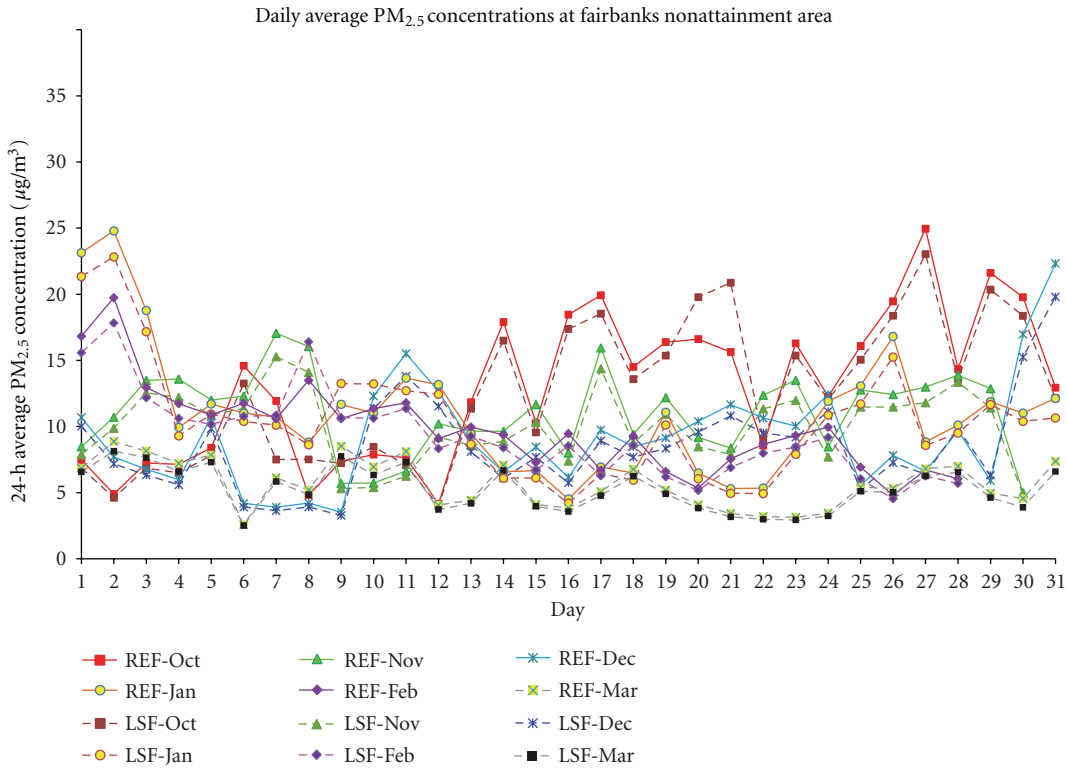


FIGURE 4: Temporal evolution of simulated 24 h-average $PM_{2.5}$ concentrations averaged over the nonattainment area for October to March as obtained by REF and LSF.

simulated the highest wind speeds and ABL heights for March (Figure 2). These relatively stronger wind speeds resulted in quick transport of pollutants out of the NAA and left only short time for aerosol formation from precursor SO_2 than in other months.

On October 8, 10, 20, 21, and 22, December 28, and 29, January 9, and 10, and February 7, 8, and 9, which account for 7% of the 182 days studied, the 24 h-average simulated $PM_{2.5}$ concentrations averaged over the NAA increased in response to the assumed usage of low sulfur fuel (Figure 4). The maximum increases of $PM_{2.5}$ in the NAA and at the grid cell of the monitoring site occurred on October 21 and were 5.2 $\mu g/m^3$ and 13.3 $\mu g/m^3$, respectively. The reasons for these increases are discussed in Section 4.4.

The RRFs of the 24 h-average $PM_{2.5}$ concentrations vary only marginally over the NAA in all months (not shown). At the grid cell of the monitoring site, the RRFs were 0.97, 0.94, 0.94, 0.97, 0.96, and 0.95 for October to March, respectively. The quarterly mean RRFs were 0.96 for both the first (January to March) and fourth quarter (October to December). The relatively high RRFs indicate a low sensitivity of simulated $PM_{2.5}$ concentrations to the assumed emission-control measure. Given that the 2008 design value was 44.7 $\mu g/m^3$, introducing low sulfur fuel would lead to a new design value of 42.9 $\mu g/m^3$. Thus, the simulations suggest that reducing the sulfur content in fuel alone for the targeted emission sources would not lead to air quality in Fairbanks that is in compliance with the NAAQS.

4.3. Role of Meteorology on the $PM_{2.5}$-Concentration Reductions. Investigation of the relation between the $PM_{2.5}$-concentration reductions and the meteorological conditions showed the following. In general, the simulated $PM_{2.5}$-concentration reductions increased at low near-surface temperatures, low ABL heights, low hydrometeor mixing ratio (cloud, rain, ice, and snow-mixing ratio integrated over all levels), and low downward shortwave radiation (R_s) (Figures 2, 5(a)–5(d)). The highest absolute correlation existed between simulated ABL-height and $PM_{2.5}$-concentration reductions ($|-0.28|$, significant at the 95% confidence level). For low ABL heights, the atmosphere typically is very stable [1]. Hence, the emitted precursors and $PM_{2.5}$ stay in a relatively thin layer. Consequently, the assumed emission reductions led to relatively high reduction in simulated $PM_{2.5}$ concentrations. The low insolation and relatively strong radiative cooling, low hydrometeor ratio (i.e., marginal cloudiness) also contributed to low ABL-heights. On the contrary, high ABL heights allow mixing of emitted gases and particles over a thicker layer, leading to a seemingly lower impact of the assumed emission reduction on the simulated near-surface $PM_{2.5}$ concentrations.

On some days the simulated meteorological conditions changed slightly in response to the assumed introduction of low-sulfur fuel (Figures 5(e)–5(h)). In the NAA, changes in the simulated meteorological quantities were relatively high in October, February, and March (Table 3). These months have relatively high insolation as compared to November to January (Figure 2) for which the simulated aerosol-radiation

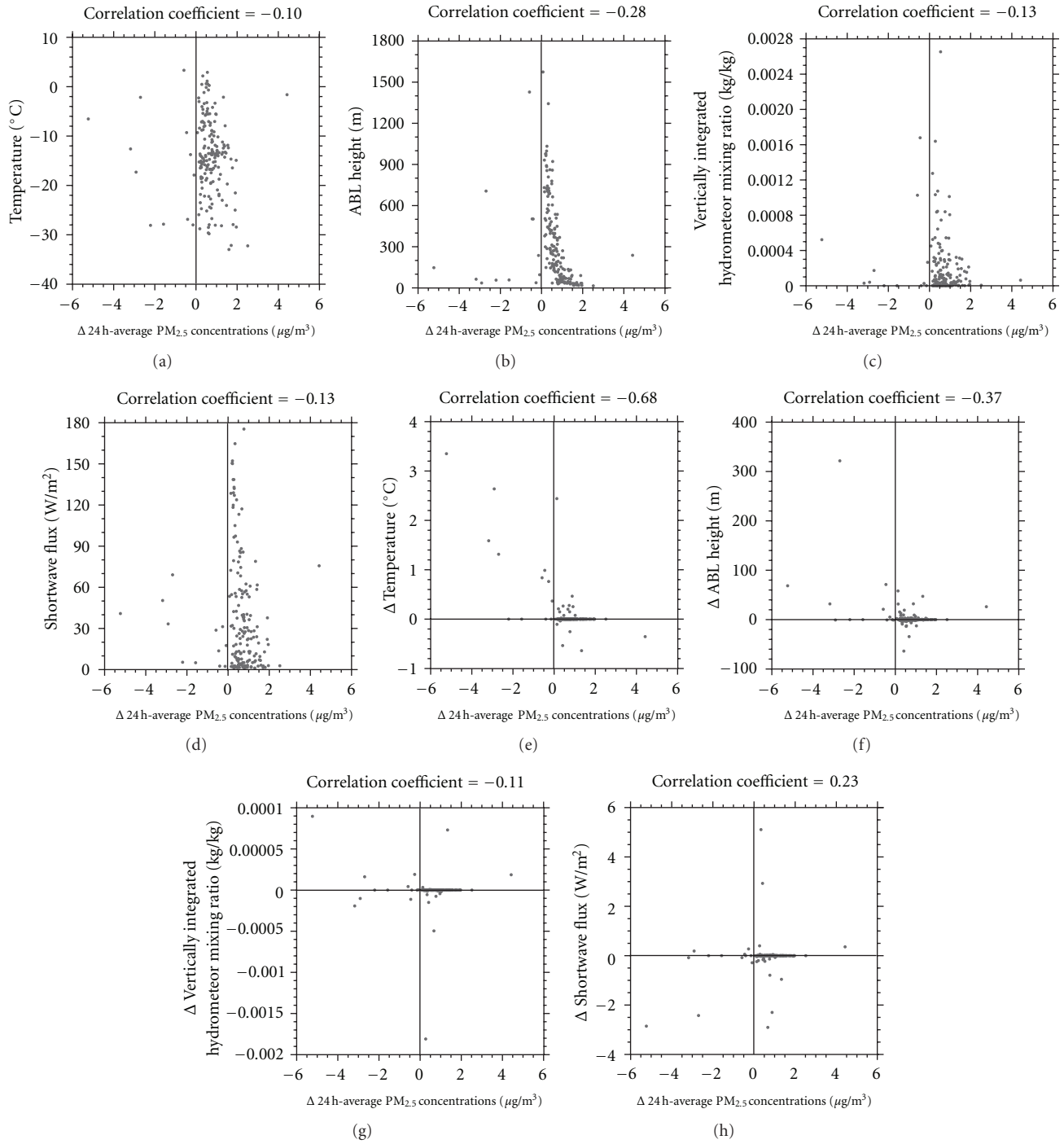


Figure 5: Scatter plots of the REF simulation of 24 h-average $PM_{2.5}$ concentrations versus (a) near-surface temperature, (b) ABL height, (c) vertically integrated hydrometeor mixing ratio, and (d) downward shortwave radiation, and differences REF-LSF of 24 h-average $PM_{2.5}$ concentrations versus difference (REF-LSF) of daily mean (e) near-surface temperature, (f) ABL height, (g) vertically integrated hydrometeor mixing ratio, and (h) downward shortwave radiation in the nonattainment area for the 182 simulation days. The lines crossing at zero indicate nondifferences with respect to the values at the x and y axis.

feedbacks can become more obvious. The changes in simulated meteorological quantities in October and February led to a more stable atmosphere, that is, reduced vertical and horizontal mixing. In March, the changes enhanced thermal turbulence and hence vertical mixing.

4.4. Speciation. In REF, the speciation of total dry $PM_{2.5}$ on average over November to March was 1% ammonium, 1.4% nitrate, 11.8% EC, 25.8% sulfate, and 60.1% OC. According to the simulations, introducing low-sulfur fuel would increase the absolute nitrate-aerosol concentrations in the

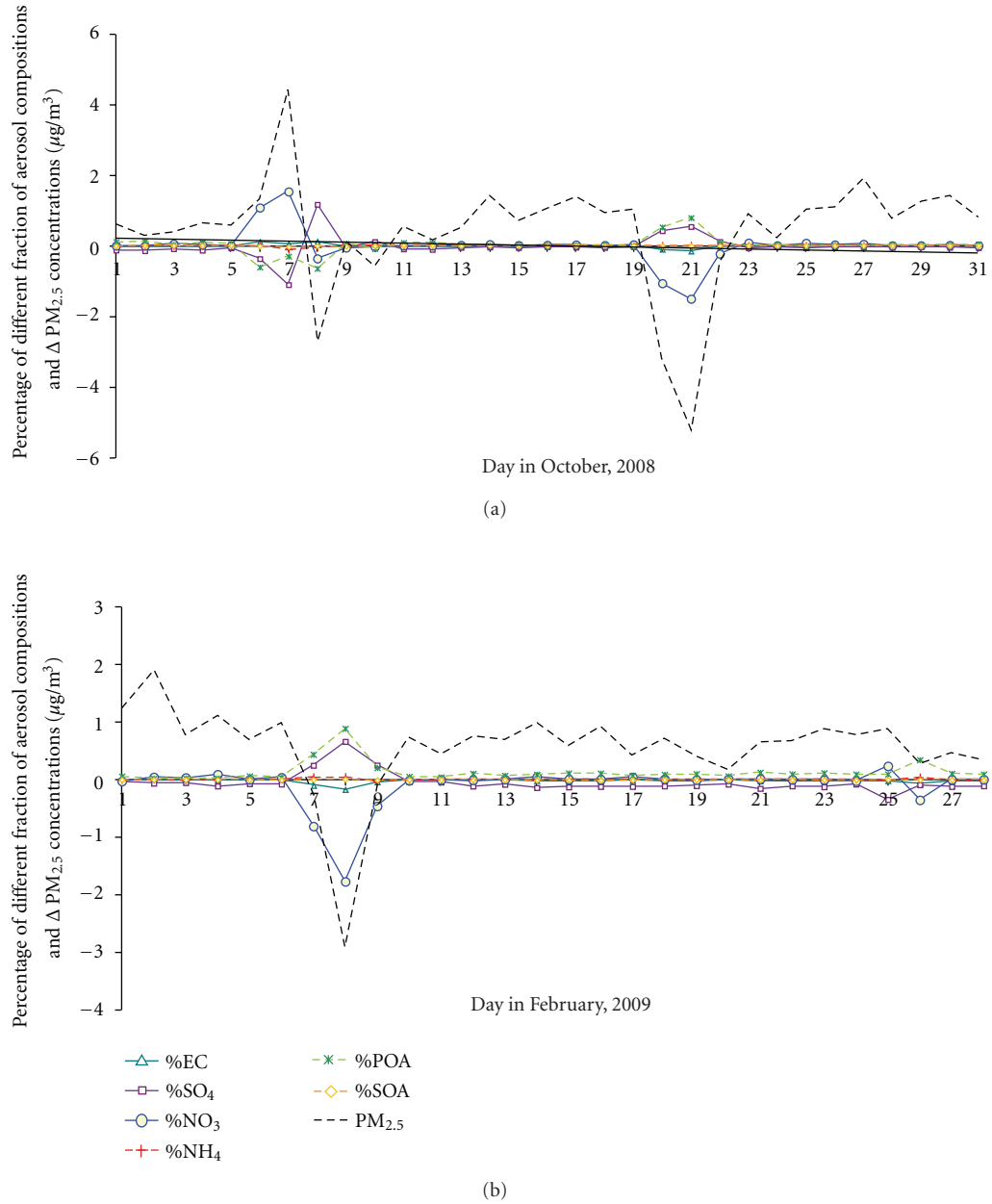
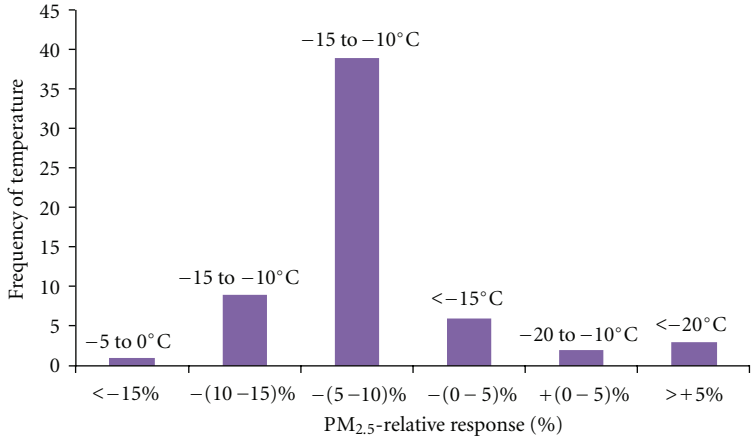


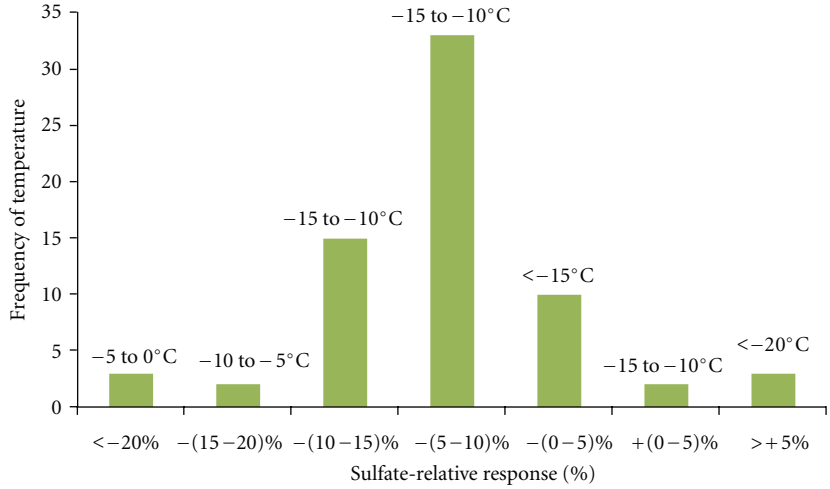
Figure 6: Temporal evolution of daily average percent differences in simulated aerosol compositions in the nonattainment area as obtained for October and February.

NAA by 3% and 10% in October and February, respectively (Table 2). Though nitrate makes up only a small fraction of the total $PM_{2.5}$, its increasing affected the reduction of $PM_{2.5}$ concentrations in these two months notably (Figure 6). On average over October to March and the NAA, the LSF-simulated OC, sulfate, EC, and emitted $PM_{2.5}$ decreased by 6%, nitrate decreased by 4% and ammonium by 1% as compared to REF. The percent reductions of OC, sulfate, EC, and emitted $PM_{2.5}$ are similar to the percent reduction of $PM_{2.5}$ and PM_{10} in the NAA (Table 2).

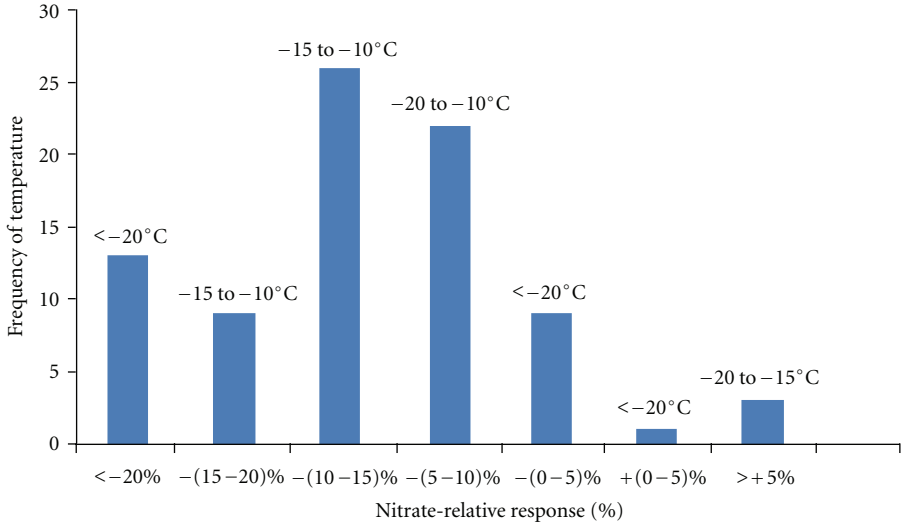
To assess how the low Fairbanks temperatures affect the relative responses of the of the total simulated $PM_{2.5}$ concentrations and its speciation in the NAA, we determined the daily relative response RR $= ((PM_{2.5,LSF} - PM_{2.5,REF})/PM_{2.5,REF}) \times 100\%$ following [15, 16]. Here $PM_{2.5,REF}$ and $PM_{2.5,LSF}$ are the 24 h-average $PM_{2.5}$ concentrations averaged over the NAA for REF and LSF, respectively. The RR of total simulated $PM_{2.5}$ and its speciation were grouped according to their magnitude in classes of 5% increments. We then identified the most frequent occurrence of daily mean temperatures in each group and calculated the frequency of that temperature. According to the simulations, the highest relative reduction of $PM_{2.5}$ (>15%) occurs between −5 and 0°C (Figure 7(a)). $PM_{2.5}$ reductions of 5 to



(a)

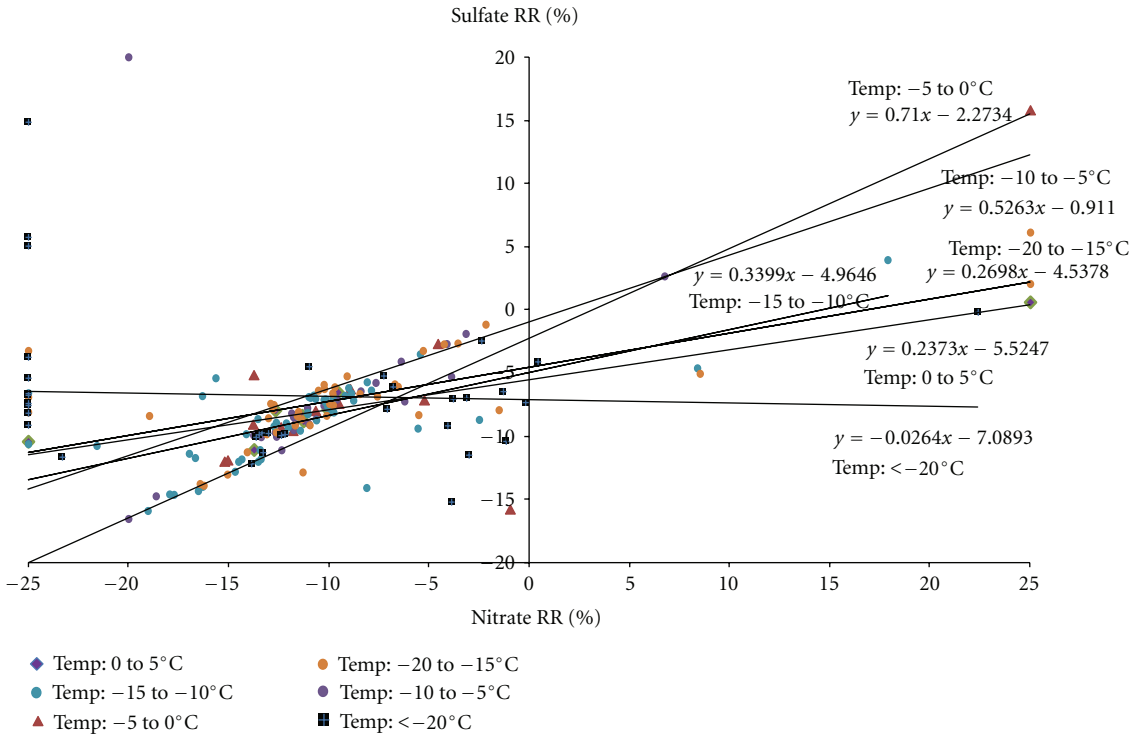


(b)



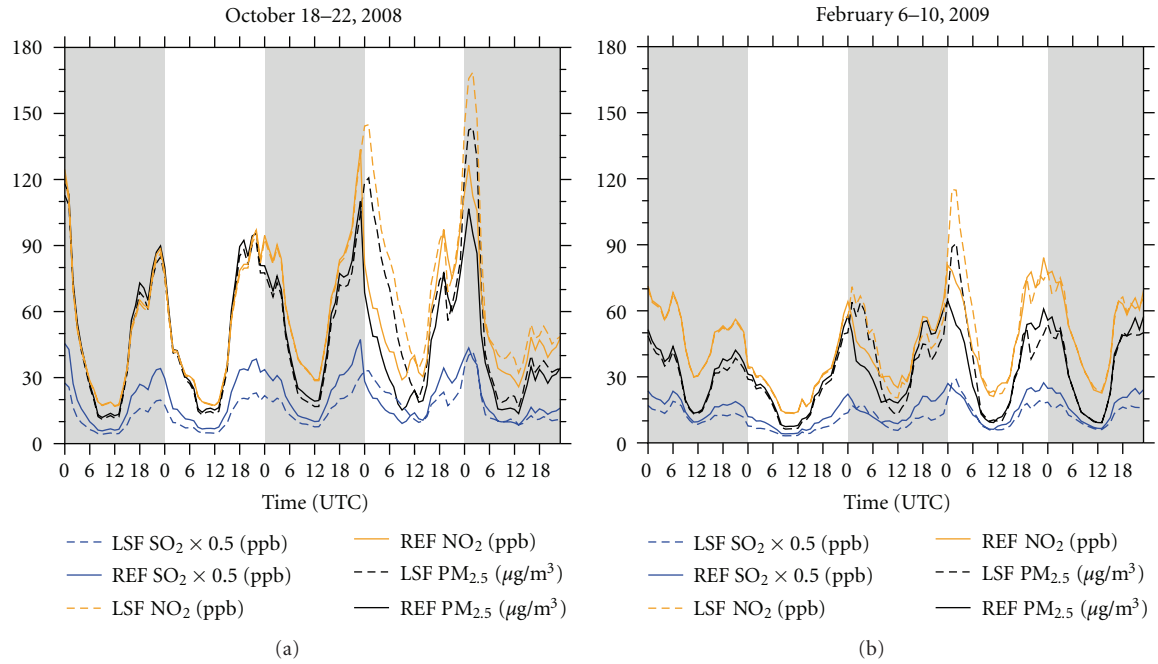
(c)

Figure 7: Continued.



(d)

Figure 7: Relative responses of (a) total $PM_{2.5}$, (b) sulfate and (c) nitrate concentrations to the assumed fuel sulfur content reductions and (d) relation of relative responses of sulfate and nitrate at different temperature ranges. The temperature ranges on each bar are the ranges of temperature which has the most frequent occurrence, and the y axis indicates that frequency.



(a) (b)

Figure 8: Mixing ratios of chemical species and $PM_{2.5}$ concentrations as obtained by REF and LSF for the gird cell that holds the monitoring site for October 18–22, 2008, and February 6–10, 2009 (UTC). The grey color serves to better distinguish among days.

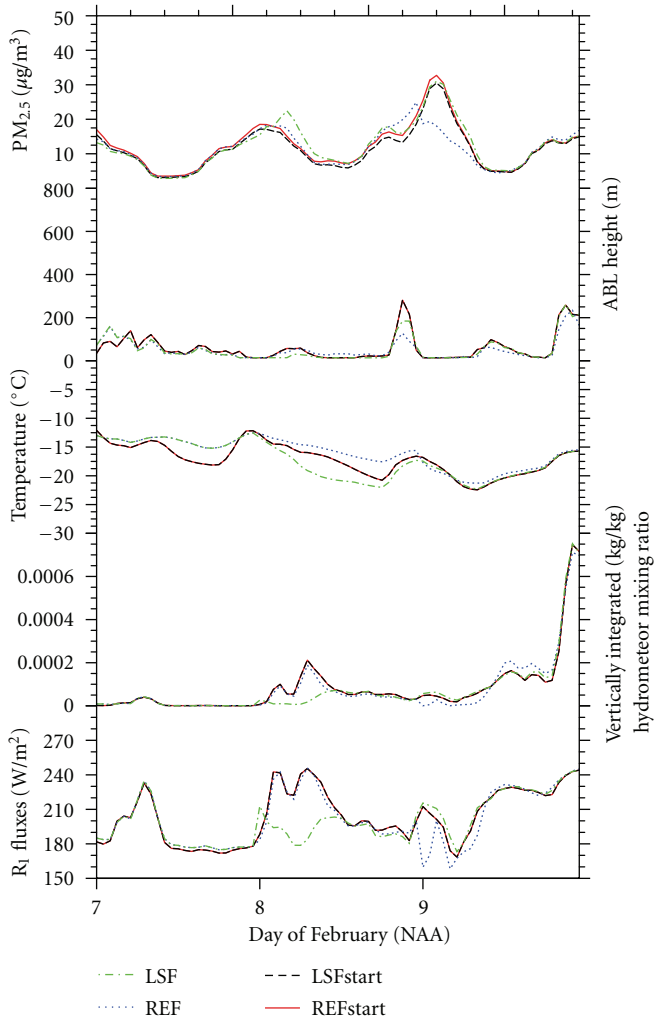


Figure 9: Meteorological variables and $PM_{2.5}$ concentrations simulated by REF, LSF, and REFstart and LSFstart averaged over the nonattainment area during February 7–9, 2009 (UTC).

10% would occur most frequently between −15 and −10°C. The same would be true for sulfate (Figure 7(b)). $PM_{2.5}$ and sulfate would decrease 0–5% for temperatures below −15°C (Figures 7(a), 7(b)). At temperatures between −15 and −10°C, nitrate would be reduced most frequently by 10 to 15%. At daily mean temperatures below −20°C, the relative reduction of nitrate would exceed 20% most of the time. However, sometimes at temperatures below −20°C, $PM_{2.5}$, sulfate and nitrate would increase (Figures 7(a), 7(b), and 7(c)). The relative nitrate changes differ from the relative sulfate changes (Figure 7(d)). In the temperature range −15 to −10°C, for instance, nitrate would decrease stronger for a given decrease in sulfate than in the range −20 to −15°C. These findings mean that at low temperatures of Fairbanks' winters, the $PM_{2.5}$ reductions in response to reduced SO_2-emissions are quite nonlinear. This finding agrees with the response of the particulate nitrate to the SO_2 reductions found in the relative warmer eastern US [14, 15].

The change in simulated meteorological quantities occurred at 1500 AST (0000 UTC) on the days with increased nitrate and relates to the increase of $PM_{2.5}$ in the NAA. They coincided with changes in various pollutants (e.g., Figure 8). On these days and time, the meteorology was initialized. Note that when running a model in forecast mode for six months, the meteorological conditions have to be initialized on a regular basis as frequent meteorological reinitializations result in improved model skills [39]. However, a reinitialization approach may lead to a discontinuity, which takes a few hours or two days to reach dynamical equilibrium [40, 41]. Discarding the first 6 h after re-initializing the meteorology yields discrepancies between the meteorological fields and the chemical fields initialized from the previous run [7] for which we did not use that approach.

We examined all 37 days with reinitialization of meteorology and found that only 6 days showed increased $PM_{2.5}$ concentrations. On these days also the $PM_{2.5}$-composition changed. These days were characterized by strong stability ($\gamma = 2.28$ K/1 km) over the NAA and averages of simulated ABL heights as low as 107 m and 80 m, in REF and LSF, respectively. Observations showed that on these days a front

came in. At the beginning of a reinitialization, cloud and ice mixing ratios are zero. It takes about 3–6 hours for the clouds and precipitation species to spin up in the model. When on an initialization day a front approaches and fogs and clouds form, downward shortwave radiation can be overestimated during the spinup [7].

To investigate whether the increased $PM_{2.5}$ on these days results from spinup effects, we reinitialized the simulations three days earlier. These simulations as their results are called REFstart and LSFstart. The temporal evolution of hourly average $PM_{2.5}$ concentrations and meteorological quantities hardly differed between REF, REFstart, and LSFstart (Figure 9). Obviously, while the clouds had not yet fully spunup, the radiative feedback with the modified aerosols led to higher long-wave radiation loss in LSF than in REF. Thus, in LSF temperatures decreased, and saturation was reached quicker than in REF. Subsequently gas-to-particle conversion increased, and thermal turbulence and the ABL-height decreased as compared to REF. Thus, $PM_{2.5}$ concentrations increased in LSF. The simulated temperature decrease supported particulate nitrate of formation (Figure 7(c)). Later the enhanced cloudiness decreased the long-wave radiation loss as compared to REF (e.g., Figure 9). These findings attribute the increased $PM_{2.5}$ concentrations and changed meteorological quantities to spinup effects.

When excluding six days that had increased $PM_{2.5}$ concentrations due to spinup effects, the RRFs were 0.95, 0.94, 0.93, 0.94, 0.94, and 0.95 for October to March, respectively, at the grid cell of the monitoring site. The quarterly mean RRFs were 0.94 for both quarters. Multiplication of the RRFs with the 2008 design value yielded 42.0 $\mu g/m^3$ which is also higher than the NAAQS. These results confirm the findings above that the assumed introduction of low-sulfur fuel alone would not yield compliance.

5. Conclusions

We examined the response of $PM_{2.5}$ concentrations at breathing level to the reduction of sulfur in heating oil and fuel used for oil-burning facilities for a subarctic city surrounded by an area with hardly any anthropogenic emission sources. In doing so, simulations were performed with the subarctic-modified WRF/Chem in forecast mode for October to March (a full-cold season). According to the simulation results, the introduction of low-sulfur fuel would lead to an average decrease of $PM_{2.5}$ concentrations of 0.6 $\mu g/m^3$ (6%) and 1.2 $\mu g/m^3$ (4.2%) in the nonattainment area and the grid-cell holding the monitoring site, respectively; it also would avoid five exceedance days. According to the simulations, the monthly average relative $PM_{2.5}$-concentration reductions varied between 4% and 9%. The quarterly average RRFs of 0.96 at the grid cell of the monitoring site indicate a low response of $PM_{2.5}$ concentrations to the assumed emission reductions. Given a design-value of 44.7 $\mu g/m^3$ and these RRFs, one has to conclude that introducing low-sulfur fuel without other emission-control measures will not achieve compliance with the NAAQS of 35 $\mu g/m^3$.

Investigation of the relationship between the simulated meteorological conditions and the $PM_{2.5}$-concentration reduction showed that the measure would be most efficient on very cold days with low ABL heights, low shortwave radiation, and low hydrometeor mixing ratios.

Running WRF/Chem in forecast mode with reinitialization of the meteorology every 5 days for an entire cold season meant 37 initializations. On six of these initialization days simulated $PM_{2.5}$ concentrations increased despite reduced sulfur fuel content. Investigation showed that on these days, the spinup of meteorology, and the aerosol-radiation feedback led to nonlinear processes that favored nitrate-aerosol formation. When removing this artifact, the RRFs decreased to 0.94; that is, the model artifact did not affect the above conclusions.

Acknowledgments

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