INVESTIGATION ON THE IMPACTS OF LOW-SULFUR FUEL USED IN RESIDENTIAL HEATING AND OIL-FIRED POWER PLANTS ON $PM_{2.5}$-CONCENTRATIONS AND ITS COMPOSITION IN FAIRBANKS, ALASKA

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A

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Abstract

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 forested areas were examined with the Weather Research and Forecasting model coupled with chemistry packages that were 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 24h-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 44.7μg/m^3 introducing low-sulfur fuel would lead to a new design value of 42.9μg/m^3 that still exceeds the US National Ambient Air Quality Standard of 35μg/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 more on days with low atmospheric boundary layer heights, low hydrometeor mixing ratio, low downward shortwave radiation and low temperatures. Furthermore, a literature review of other emission control measure studies is given, and recommendations for future studies are made based on the findings.

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CHAPTER 1

Introduction

1.1 Background

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 high frequency of exceedances of the 24h-average National Ambient Air Quality Standard (NAAQS) of 35µg/m^3 for particulate matter (PM) of a diameter less than 2.5µm ($PM_{2.5}$) during past winters [1]. High concentrations of $PM_{2.5}$ suspended in the urban atmosphere are hazardous to human health [2]. In Fairbanks, these high concentrations have led to an increase in hospital admissions for cerebrovascular and respiratory diseases [3].

The nonattainment designation led to the obligation to develop an approvable State Implementation Plan (SIP) by December, 2012, which must demonstrate attainment in this area by December, 2014. To ensure compliance with the NAAQS, the State of Alaska's Department of Environmental Conservation (DEC), the Fairbanks North Star Borough (FNSB), a private for profit company (Sierra Research Inc.), and the University of Alaska Fairbanks (UAF) have been working cooperatively to assemble information about the causes of the Fairbanks' area air quality problem. These activities included analysis of weather situations that lead to high $PM_{2.5}$-concentrations [1, 4, 5], documentation of the trends in ambient $PM_{2.5}$-concentrations [6], emission sources and rates [7-8]; and scientific investigations on the contribution of various sources (e.g.

[9-10]), and on the impacts of potential emission control measures [11-14]. These analyses provide part of the framework for the development a SIP.

1.2 Applications of Air Quality Model for Investigating the Impacts of Emission Reduction Measures

In order to apply air quality models to generate the information used in the model attainment demonstration, the simulated 24h-averaged $PM_{2.5}$-concentrations were suggested to be used by [15], as they are the baseline design value calculations. The baseline design value is an average of several design values of monitored 24h-averaged $PM_{2.5}$-concentrations at each monitoring site, which is calculated from the 5-year base period centered around the modeling year [15]. For Fairbanks, the 5-year base period is 2006-2010, and it has a 2008 design value of 44.7μg/m^3. It should be noted that 2008 is the middle year of the base period.

This baseline design value is used to project future year concentrations by multiplying it by the relative response factors (RRFs). The RRFs are calculated as the ratio of the 24h-average concentration obtained by the alternative emission scenario to the concentration that was obtained by the reference or base case scenario [15]. Multiplication of RRF with the design value for Fairbanks provides the new design value that represents the conditions that would be found if the emission control measure was in place. In the case of studies on the contribution of emission sources, the multiplication of

RRF provides the design value representing the conditions without the contribution of the emission sources that were switched off or changed.

1.3 Alaska-adapted WRF/Chem

The Weather Research and Forecasting model inline coupled with a chemistry package (WRF/Chem), which was adapted to simulate Alaska conditions during winter [16-18]. In winter 2008-09, WRF/Chem performs best for $PM_{2.5}$-concentrations between 15 and 50µg/m^3 [16]. For $PM_{2.5}$-species, the performance is best for organic carbon (OC) followed by sulfate. Ammonium was strongly underestimated by WRF/Chem. 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 [17-18].

Based on the WRF/Chem simulations, the contributions of different emission sources, as well as the impacts of different emissions scenarios, were studied for the Fairbanks area. RRFs were used to compare the impacts of each emission control measure or contribution of an emission source on the $PM_{2.5}$-concentrations at breathing level (2m). Mölders et al. (2011, [19]) and Tran and Mölders (2012, [12]) found that point sources contributed 0.7µg/m^3 (5%) of the 24h-average $PM_{2.5}$-concentrations on average over the NAA, and 1.2µg/m^3 (4%) at the State Office Building site for the 2005-

2006 winter, respectively. Note that the State Office Building is the official monitoring site which is located in the middle of the central business district of Fairbanks (Figure 1.1). Point sources provided the additional amount that led to exceedances on 13% of the exceedance days. On the other days, exceedances would have occurred even without the presence of point source emissions. For the point source scenario case, RRF is 0.97 for the 2005-2006 winter [12]. This means that point sources, on average, contribute 1.3µg/m^3 to the Fairbanks' 2008 design value of 44.7µg/m^3.

The effects of wood stove replacements for the year 2008-2009 were examined by [13]. They found that the assumed wood stove replacements would lead to a decrease of the 24h-average $PM_{2.5}$-concentrations of 0.6µg/m^3 (1.5µg/m^3), which corresponds to a 6% (5%) reduction of the 24h-average $PM_{2.5}$-concentrations on average over the NAA (State Office Building site) for the 2008-2009 winter. The assumed replacement could avoid 13% of the exceedance days. The average RRF for the wood stove replacement scenario is 0.95. However, the results of sensitivity simulations showed that the average RRF varied significantly with the number of non-certified wood stoves and the number of wood stoves that would have to be replaced. When the number of wood-burning devices and uncertified wood stoves were estimated following the assumption of [20], the average RRF could be as low as 0.54, which would achieve compliance with the NAAQS in the two week sensitivity test performed by [10, 13].

This number of devices is much larger than those found in a later survey by [21]. Nevertheless, the higher number of wood stoves is probably more accurate and representative of the real emissions in the FNSB during the 2008-2009 winter. In

Fairbanks' households, the use of wood stoves increased in response to the increased price of heating fuel since 2007, which can be derived by the number of wood cutting permits that had tripled in 2009 [10] .

Mölders (2013, [14]) studied the impact of substituting all wood heating, which was calculated from [20] by natural gas in the NAA. This assumption resulted in a decrease of the 24h-average $PM_{2.5}$-concentrations by 1.0µg/m^3 (3.9µg/m^3) which corresponds to 11% (13%) reduction of the 24h-average $PM_{2.5}$-concentrations on average over the NAA (State Office Building site) for the 2008-2009 winter. For the substitution of the use of wood burning by natural gas scenario, the RRF is 0.87, which would lead to a new design value of 38.9µg/m^3. This value is still higher than the NAAQS.

A multiple emission control measure, the combination of a non-certified wood stove replacement measure, and introduction of low-sulfur fuel was also examined [11]. This measure was found to be more effective in the NAA than any single measure, as it decreased the 24h-average $PM_{2.5}$-concentrations by 1.4µg/m^3 (3.6µg/m^3), which corresponds to 15% (12%) of the 24h-average $PM_{2.5}$-concentrations on average over the NAA (at the State Office Building site) for the 2008-2009 winter. The average RRFs at the State Office Building site is 0.88, which leads to a 24h-average $PM_{2.5}$-concentrations of 39.3µg/m^3, which still exceeds the NAAQS.

The magnitude of the $PM_{2.5}$-concentrations also depends on the meteorological conditions. Simulated low temperatures (below -20ºC), calm winds (<0.5m/s), high emission of $PM_{2.5}$ (>0.2g/m^2.h), low vapor pressures (<2hPa), and low mixing heights are

the conditions that support the exceedance of simulated $PM_{2.5}$-concentrations [1, 11]. This behavior of WRF/Chem simulations agrees well with the observational data which showed that during November to Feburary of 1999-2009, the exceedances of $PM_{2.5}$ occurred under similar conditions with inversion conditions lasting multiple days [1].

The stagnant conditions resulted in the accumulation of primary $PM_{2.5}$ ($PM_{2.5}$ that is directly emitted), and increased the time available for the gas-to-particle conversion processes to form secondary $PM_{2.5}$ [11]. Additionally, downward shortwave radiation can cause the increase of the photolysis rates that supports the particle formation process [11, 14], i.e., it is critical to simulate the cloudiness accurately.

Various studies showed that the magnitude of the $PM_{2.5}$-concentrations depends on the emissions and meteorological conditions in Fairbanks [1, 11, 13]. Therefore, accurate emissions, as well as good meteorological information, are keys for any assessment of emission control measures.

1.4 Low-sulfur Fuel Control Measures

Low-sulfur fuel is one of the SIP emission control measures that has been adopted on the Eastern Coast of the U.S. (i.e., New Jersey [22]). This measure reduces the precursors by reducing the sulfur content of fuel oils that is used in residential and commercial heating and power generation.

In Fairbanks, residential heating oil (number 2 fuel oil), which has an average sulfur content of about 2,500 ppm, is normally used. 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 [23]. Introducing a low-sulfur fuel control measures may reduce the sulfate concentrations which is the major $PM_{2.5}$-composition in the Fairbanks nonattainment area (Figure 1.2).

However, the result of the reductions might not be effective in reducing $PM_{2.5}$-concentrations as expected. Reductions in sulfate concentrations may cause inorganic parts of $PM_{2.5}$ to respond nonlinearly. The ammonium nitrate may increase due to the reductions of sulfate concentrations [24]. Low temperature favors the formation of solid aerosol nitrate. The replacement of sulfate by nitrate is eight times higher during the winter than during summer [25]. Therefore, the goal of this thesis is to determine the potential effects of using low-sulfur fuel on the $PM_{2.5}$-concentrations in Fairbanks, a city in subarctic Alaska where winter temperatures are often below -20ºC.

1.5 Hypothesis and Objectives of the Study

The hypothesis of this study is as follows: "Under given meteorological conditions during the cold season in Fairbanks, reducing the fuel-sulfur content is not sufficient to achieve the required reduction".

The objectives of the study are as follows:

- To determine the effects of low-sulfur fuel on $PM_{2.5}$-concentrations and its composition in the Fairbanks nonattainment area
- To determine the relative response factors that indicate how effective of the low-sulfur fuel measure would be in complying with the NAAQS

- To investigate the impact of meteorology on the reductions of $PM_{2.5}$-concentrations.

To meet all of these three objectives, I analyzed the results of simulations of the reference and the low-sulfur fuel case performed with the Alaska-adapted WRF/Chem. The model experimental design, analysis method, and results are presented and discussed in chapter 2. The conclusions are presented in chapter 3. The primary results from chapter 2 of this thesis have been published as a peer-reviewed article (K. Leelasakultum, N. Mölders, H. N. Q. Tran, and G. A. Grell, "Potential impacts of the introduction of low-sulfur fuel on $PM_{2.5}$ concentrations at breathing level in a subarctic city," *Advances in Meteorology,* vol. 2012, pp. 1-15, 2012.). The evaluation of the simulation is published in N. Mölders, H. N. Q. Tran, C. F. Cahill, K. Leelasakultum, and T. T. Tran, "Assessment of WRF/Chem $PM_{2.5}$-forecasts using mobile and fixed location data from the Fairbanks, Alaska winter 2008/09 field campaign," *Atmospheric Pollution Research,* vol. 3, pp. 180-191, 2012.

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Figure 1.1: Fairbanks $PM_{2.5}$-nonattainment area boundary (red polygon) and the locations of the State Office Building site (blue icon). Retrieved from http://maps.google.com.

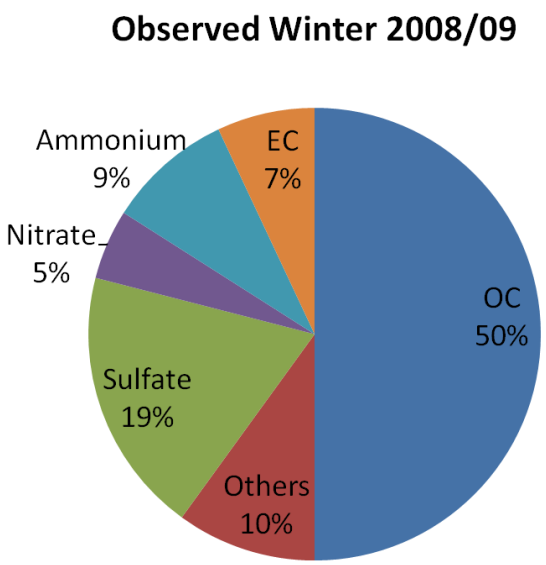

Figure 1.2: Composition of observed 24h-average total $PM_{2.5}$-concentrations in winter 2008/09. EC is elemental carbon and OC is organic carbon. Others include Al, Br, Ca, Cl, Cu, Fe, Pb, Ni, K, Se, Si, Na, S, Sn, Ti, V, and Zn.

CHAPTER 2

Potential Impacts of the Introduction of Low-sulfur Fuel on $PM_{2.5}$-concentrations at Breathing Level in a Remote Subarctic City[1]

Abstract

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 24h-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 44.7μg/m^3 introducing low-sulfur fuel would lead to a new design value of 42.9μg/m^3 that still exceeds the US National Ambient Air Quality Standard of 35μg/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.

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[1] K. Leelasakultum, N. Mölders, H. N. Q. Tran, and G. A. Grell, "Potential Impacts of the Introduction of low-sulfur fuel on $PM_{2.5}$ concentrations at breathing level in a subarctic city," *Advances in Meteorology*, vol. 2012, pp. 1-15, 2012.

2.1 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 24h-average National Ambient Air Quality Standard (NAAQS) of 35µg/m^3 for particulate matter of diameter less than 2.5µm ($PM_{2.5}$) 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 2.1) that along with strong inversions from radiative cooling and calm winds (<0.5m/s at 10m) limit the horizontal and vertical exchange of air. Extremely low temperatures (≤-20ºC at 2m) 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 particulate matter (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 578km radius, i.e. 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 as they occur in the cold season in Fairbanks.

One mitigation strategy 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 44.7μg/m^3, for an emission-control measure to be efficient it 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 reduction as large as those found for the eastern US. The low humidity also hinders particle growth to PM_{10} (PM with diameter >10μm).

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 measure 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].

2.2 Experimental Design

2.2.1 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 Reactive Organic Gases (ROG) [9]. For aerosol inorganic chemistry, the model includes sulfate, ammonium, and

nitrate for thermodynamic gas/aerosol equilibrium. Dry deposition of trace gases was determined in accord with Wesely [31], with the modifications by [7].

2.2.2 Simulations

The area for our analysis encompasses 80×70 grid-points with a grid-increment of 4km centered over Fairbanks (Figure 1). The vertically stretched grid had 28 layers up to 100hPa. The initial meteorological conditions, including snow and soil variables, were downscaled from the $1^{o}\times1^{o}$, 6h-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 0000 UTC to April 1, 2009 0000 UTC and analyzed for October 1 to March 31 Alaska Standard Time (AST=UTC-9h). 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.

2.2.3 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].

2.2.4 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. 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.

2.3 Reference Simulation

2.3.1 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 2.1). 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 an October with normal mean temperatures. Consequently, October had the highest $PM_{2.5}$-emissions in REF and LSF, and the lowest relative $PM_{2.5}$-emissions 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.

2.3.2 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 2m-temperature, 2m-dewpoint temperature, sea-level pressure, 10m-wind-speed and direction of 1.3K, 2.1K, -1.9hPa, 1.55m/s, and -4^{o}, respectively [6]. The wind-errors explain some of the underestimation of the $PM_{2.5}$-concentrations. The overestimation of temperatures led to biases of 0.5K, 0.8K, 2K, 2.6K, 1.6K and 0.3K and root-mean square errors (RMSEs) of

3.8K, 4.8K, 6.1K, 4.3K, 5.2K and 4.1K in October to March, respectively; 2m dewpoint temperature RMSEs were less than 5K except November (6.2K). Performance was better on relatively warmer than colder days and in the rural than urban areas [6]. Mobile temperature measurements indicated that in the NAA, simulated temperatures were about 1.4, 2.4, 1.2, and 2.2K too high in November, December, January and February, and 0.9K 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 2.2).

The $PM_{2.5}$-evaluation used the fractional bias $FB = \frac{2}{\mathrm{N}} \sum_{\mathrm{i=1}}^{\mathrm{N}} \frac{\mathrm{C_s - C_o}}{\mathrm{C_s + C_o}} \times 100\%$, fractional error $FE = \frac{2}{\mathrm{N}} \sum_{\mathrm{i=1}}^{\mathrm{N}} \left|\frac{\mathrm{C_s - C_o}}{\mathrm{C_s + C_o}}\right| \times 100\%$, normalized mean bias $NMB = \frac{\sum_{i=1}^{N} C_s - C_o}{\sum_{i=1}^{N} C_o} \times 100\%$, and normalized mean error $NME = \frac{\sum_{i=1}^{N} |C_s - C_o|}{\sum_{i=1}^{N} C_o} \times 100\%$. On average over October to March and all sites, the FB, FE, NMB and NME for 24h-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 mid-latitudes [6]. WRF/Chem performed best for $PM_{2.5}$-concentrations between 15 and 50µg/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 50µg/m^3, (2) performed acceptably for sulfate species in $PM_{2.5}$, and (3) LSF and REF are affected by errors in the same way, i.e. 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.

2.3.3 Urban Air Quality and Meteorology

On average over the analysis domain and October to March, the simulated 24h-average $PM_{2.5}$-concentration was 0.4µg/m^3 in the lowest layer in REF. In the NAA, $PM_{2.5}$-concentrations were highest. Here, in REF, the monthly averages of 24h-average $PM_{2.5}$ concentrations were 13.0, 11.0, 9.2, 11.0, 9.8 and 5.7µg/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 2.2). 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

2.2). 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 24h-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 >0.5m/s dilute the $PM_{2.5}$-concentrations, while calm winds (<0.5m/s) build up the $PM_{2.5}$-concentrations in the NAA [1]. WRF/Chem showed this behavior during October to March (Figure 2.2). During these months, the monthly average simulated wind-speeds in the NAA were 2.27, 1.93, 2.68, 2.62, 2.18, and 3.74m/s, respectively. The relatively stronger wind simulated for March than in other months resulted in the lowest monthly average of 24h-average simulated $PM_{2.5}$-concentrations, and aerosol compositions including nitrate, ammonium, sulfate, EC and OC in the NAA (Table 2.2).

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 (<100m) 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 100m 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 2.2).

2.4 Low-sulfur Fuel

2.4.1 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 for temperature dependent use in WRF/Chem. Compared to REF, assuming 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 2.1). Emissions from all other sources than oil-furnaces and oil-burning facilities were identical to those in REF.

2.4.2 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 2.3). In the NAA, in response to the assumed emission changes, the simulated $PM_{2.5}$-concentrations decreased by 0.5µg/m^3, 1.0µg/m^3, 0.7µg/m^3, 0.6µg/m^3, 0.5µg/m^3 and 0.4µg/m^3 in October to March, respectively, and by 0.6µg/m^3 on average over these months. These simulated $PM_{2.5}$-concentration reductions were significant in November, December and March (Figure 2.3). The relative monthly mean of 24h-average $PM_{2.5}$-concentration reductions would vary between 4% and 9% (Table 2.2). At the grid-cell of the monitoring site, the October to March monthly averaged 24h-average $PM_{2.5}$-concentrations decreased from 40.2, 30.3, 25.8, 33.9, 27.1 and 17.1µg/m^3 in REF, respectively, to 39.2, 28.6, 24.4, 32.7, 26.0 and 16.2µg/m^3 in LSF, i.e. 1.2µg/m^3 (~4%) on average.

The simulations suggested that introduction of low-sulfur fuel would reduce the number of exceedance days (days with 24h-average $PM_{2.5}$-concentrations >35µg/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 24h-average $PM_{2.5}$-concentrations in the NAA.

The 24h-average $PM_{2.5}$-concentration differences between REF and LSF for each of the 182 simulation days were 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 2.2, Table 2.3). Thus,

pollutants accumulated and had enough time for chemical conversion. The changed composition and reduced amount of precursors in LSF as compared to REF, 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 2.1, 2.2). 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) simulated the highest wind-speeds and ABL-heights for March (Figure 2.2). 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, 9, which account for 7% of the 182 days studied, the 24h-average simulated $PM_{2.5}$-concentrations averaged over the NAA increased in response to the assumed usage of low-sulfur fuel (Figure 2.4). 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 5.2μg/m^3 and 13.3μg/m^3, respectively. The reasons for these increases are discussed in section 2.4.4.

The RRFs of the 24h-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 44.7µg/m^3 introducing low-sulfur fuel would lead to a new design value of 42.9µg/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.

2.4.3 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 2.2, 2.5a-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 (Figure 2.5e-h). In the NAA, changes in the simulated meteorological quantities were relatively high in October, February and March (Table 2.3). These months have relatively high insolation as compared to November to January (Figure 2.2) for which the simulated aerosol-radiation feedbacks can become more obvious. The changes in simulated meteorological quantities in October and February led to a more stable atmosphere, i.e. reduced vertical and horizontal mixing. In March, the changes enhanced thermal turbulence and hence vertical mixing.

2.4.4 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 NAA by 3% and 10% in October and February, respectively (Table 2.2). 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 2.6). 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 2.2).

To assess how the low Fairbanks temperatures affect the relative responses of the total simulated $PM_{2.5}$-concentrations and its speciation in the NAA, we determined the daily relative response $RR = \frac{(PM_{2.5,LSF} - PM_{2.5,REF})}{PM_{2.5,REF}} \times 100\%$ following [15, 16]. Here $PM_{2.5,REF}$ and $PM_{2.5,LSF}$ are the 24h-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^{o}C (Figure 2.7a). $PM_{2.5}$-reductions of 5 to 10% would occur most frequently between -15 and -10^{o}C. The same would be true for sulfate (Figure 2.7b). $PM_{2.5}$ and sulfate would decrease 0-5% for temperatures below -15^{o}C (Figure 2.7a, b). At temperatures between -15 and -10^{o}C, nitrate would be reduced most frequently by 10 to 15%. At daily mean temperatures below -20^{o}C, the relative reduction of nitrate would exceed 20% most of the time. However, sometimes at temperatures below -20^{o}C, $PM_{2.5}$, sulfate and nitrate would increase (Figure 2.7a, b, c). The relative nitrate changes differ from the relative sulfate changes (Figure 2.7d). In the temperature range -15 to -10^{o}C, for instance, nitrate would decrease more for a given decrease in sulfate than in the range -20 to -15^{o}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 2.8). 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-intializing 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 (γ=2.28K/1km) over the NAA and averages of simulated ABL heights as low as 107m and 80m, 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 2.9). 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 formation (Figure 2.7c). Later the enhanced cloudiness decreased the long-wave radiation loss as compared to REF (e.g., Figure 2.9). These findings attribute the increased $PM_{2.5}$-concentrations and changed meteorological quantities to spinup effects.

When excluding the 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 42.0g/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.

2.5 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 0.6μg/m^3 (6%) and 1.2μg/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 44.7μg/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 35μg/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 ratio.

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.

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Table 2.1: 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} x10^4 (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 x10^5 (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 x10^5 (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} (ºC)	-8.1	-14.7	-17.5	-18.2	-13.7	-13.5	
$T_{30y\text{-}average}$ (ºC)	-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	

Table 2.2: Monthly mean of 24h-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 as 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 (µg/kg.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 (µg/kg.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 x 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%)
Element 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%)

Table 2.3. 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 (ºC)	-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 (ºC)	-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)

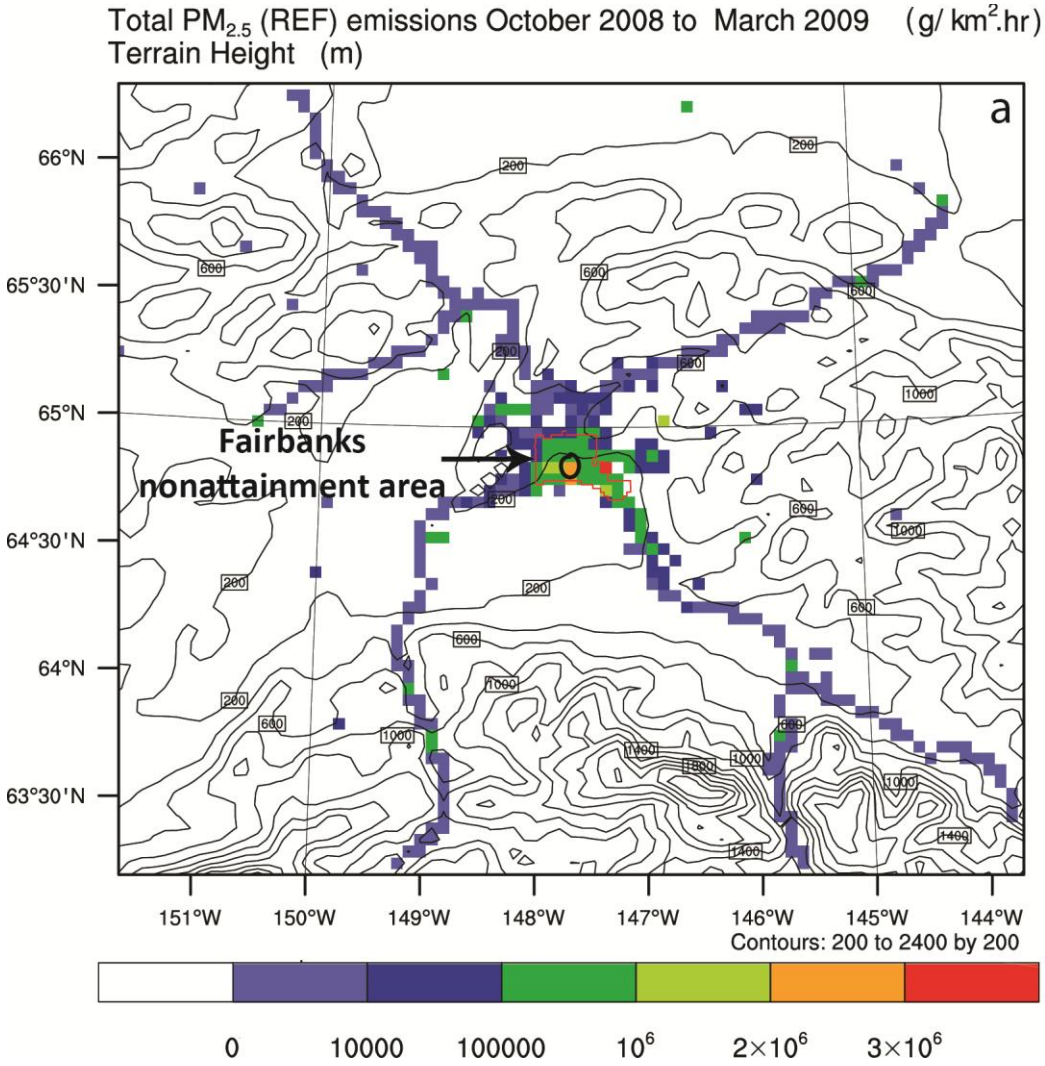

Figure 2.1: Total emission of (a) $PM_{2.5}$, (b) SO_2, and (c) NO_x from October 1, 2008 to March 31, 2008 (color) from all layers in the analysis domain. Terrain height is superimposed (contour lines). In (a), the red polygon and black circular shape indicate the boundaries of the Fairbanks nonattainment area and the location of the official $PM_{2.5}$-monitoring site, respectively.

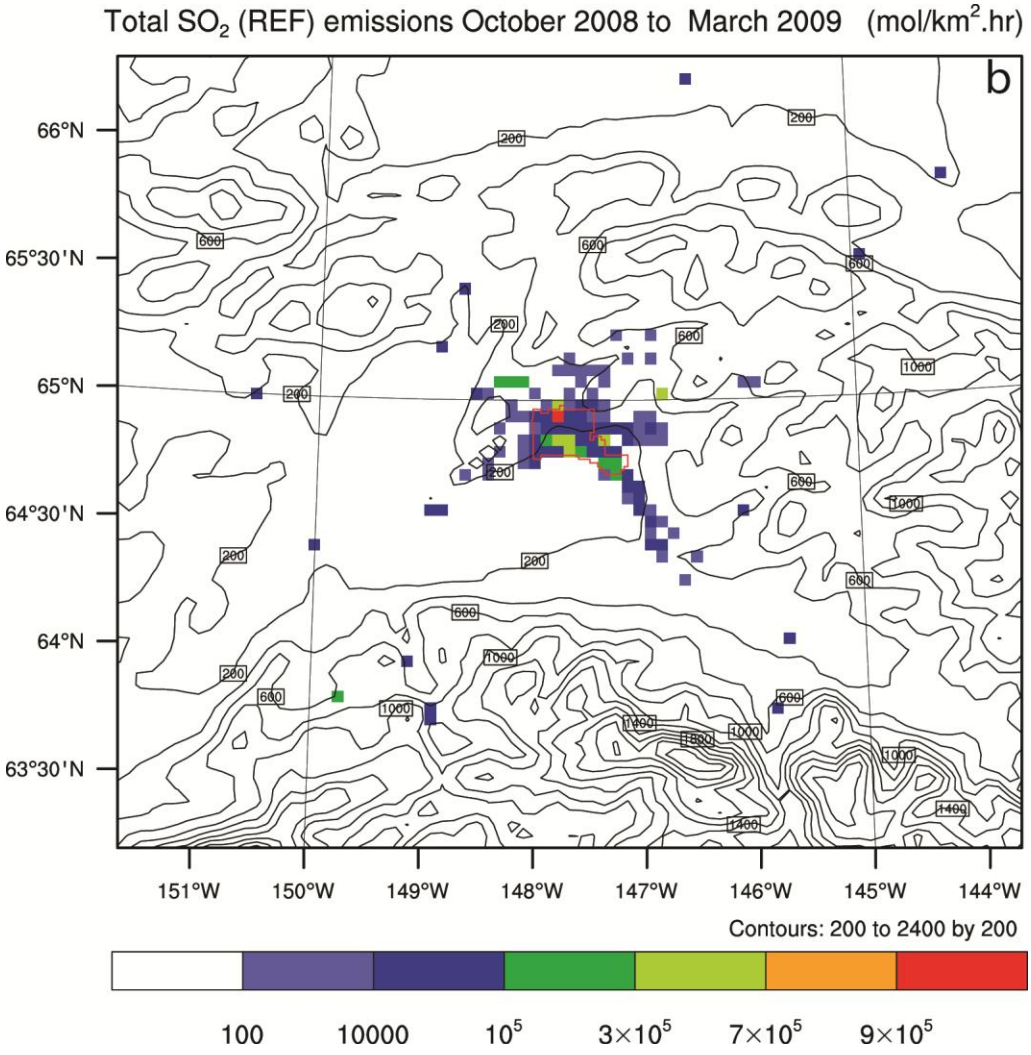

Figure 2.1 (cont.)

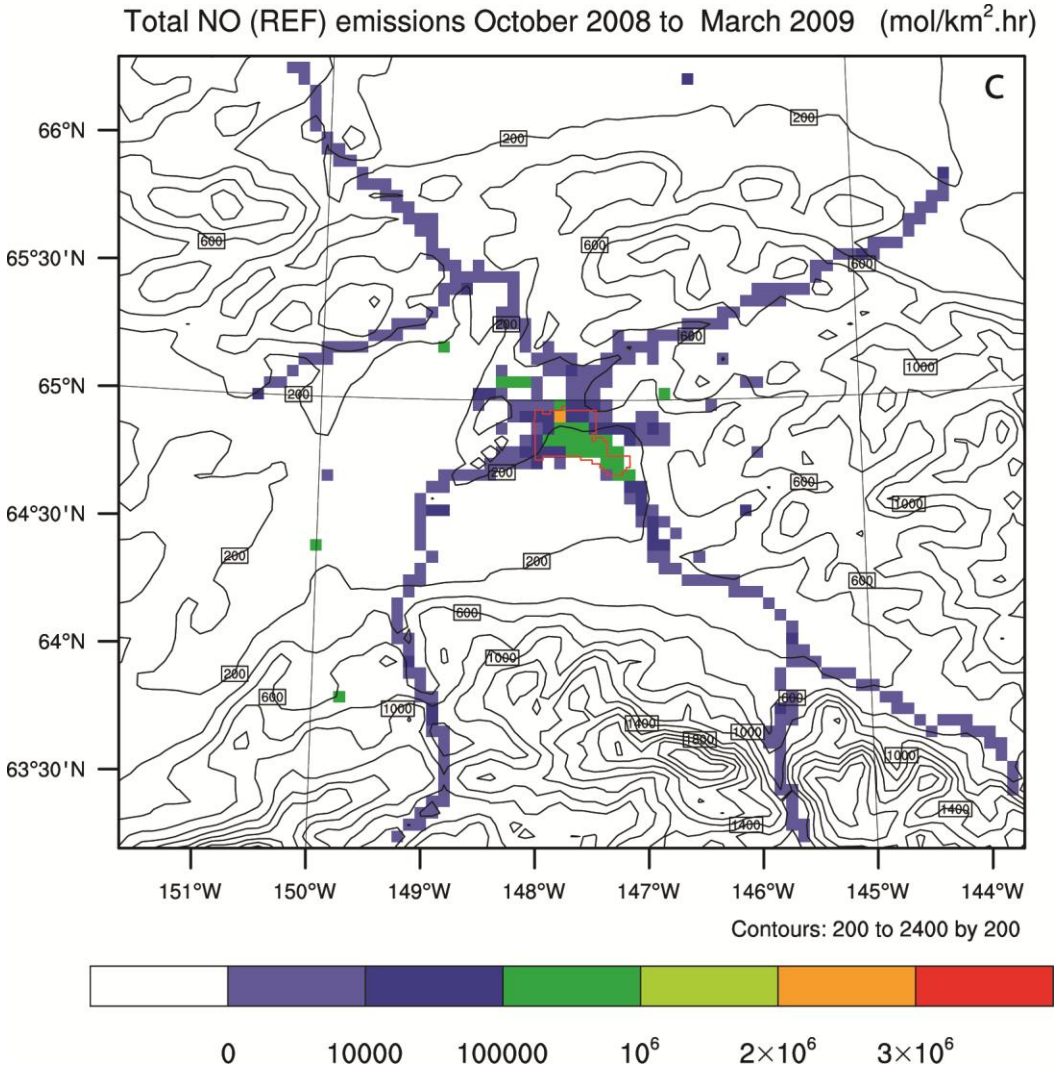

Figure 2.1 (cont.)

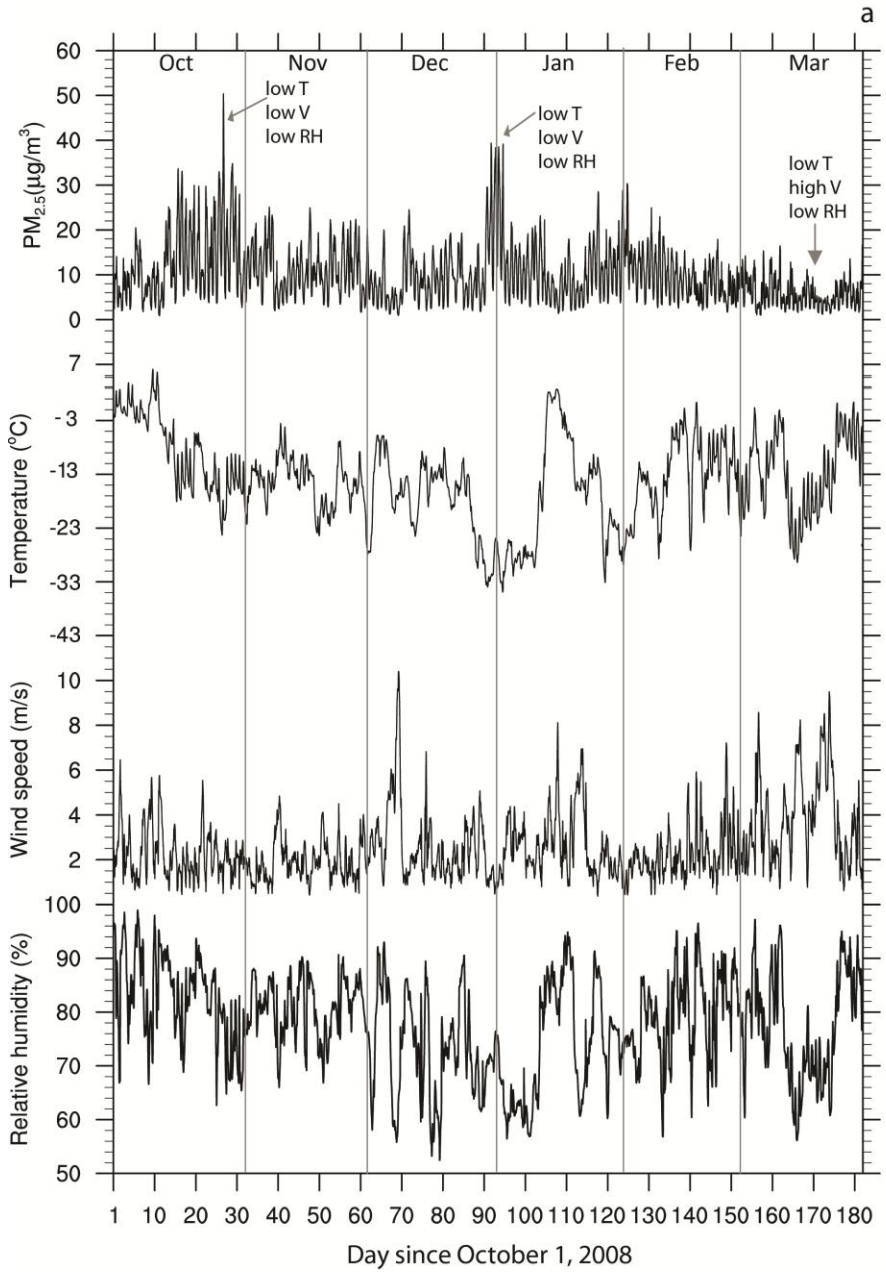

Figure 2.2: Temporal evolution of simulated hourly (a) $PM_{2.5}$-concentrations, temperature, wind-speed, relative humidity, and (b) downward long-wave radiation, hydrometeor mixing ratios, ABL-height and downward shortwave radiation averaged over the Fairbanks nonattainment area for each of the 182 simulation days.

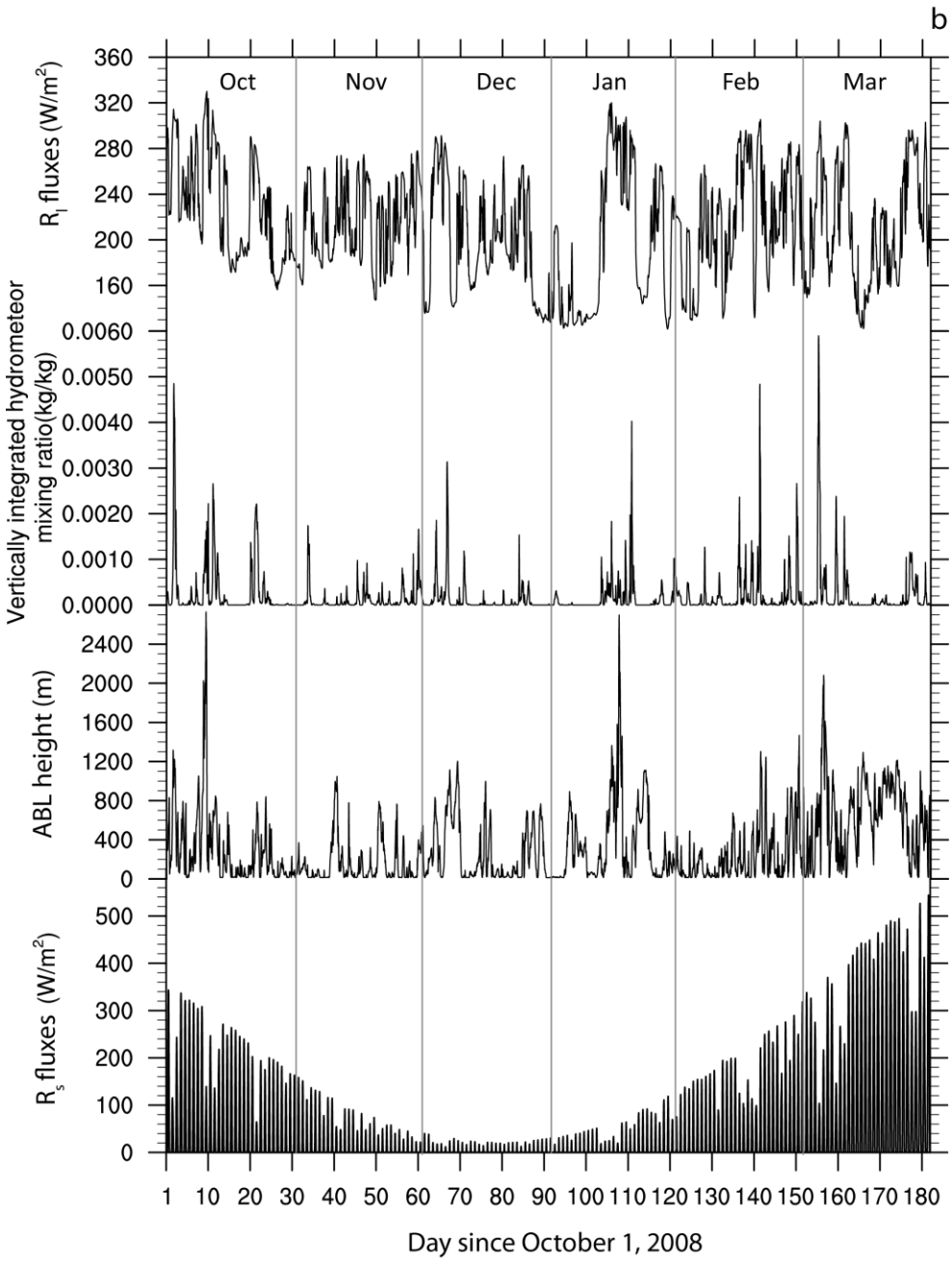

Figure 2.2 (cont.)

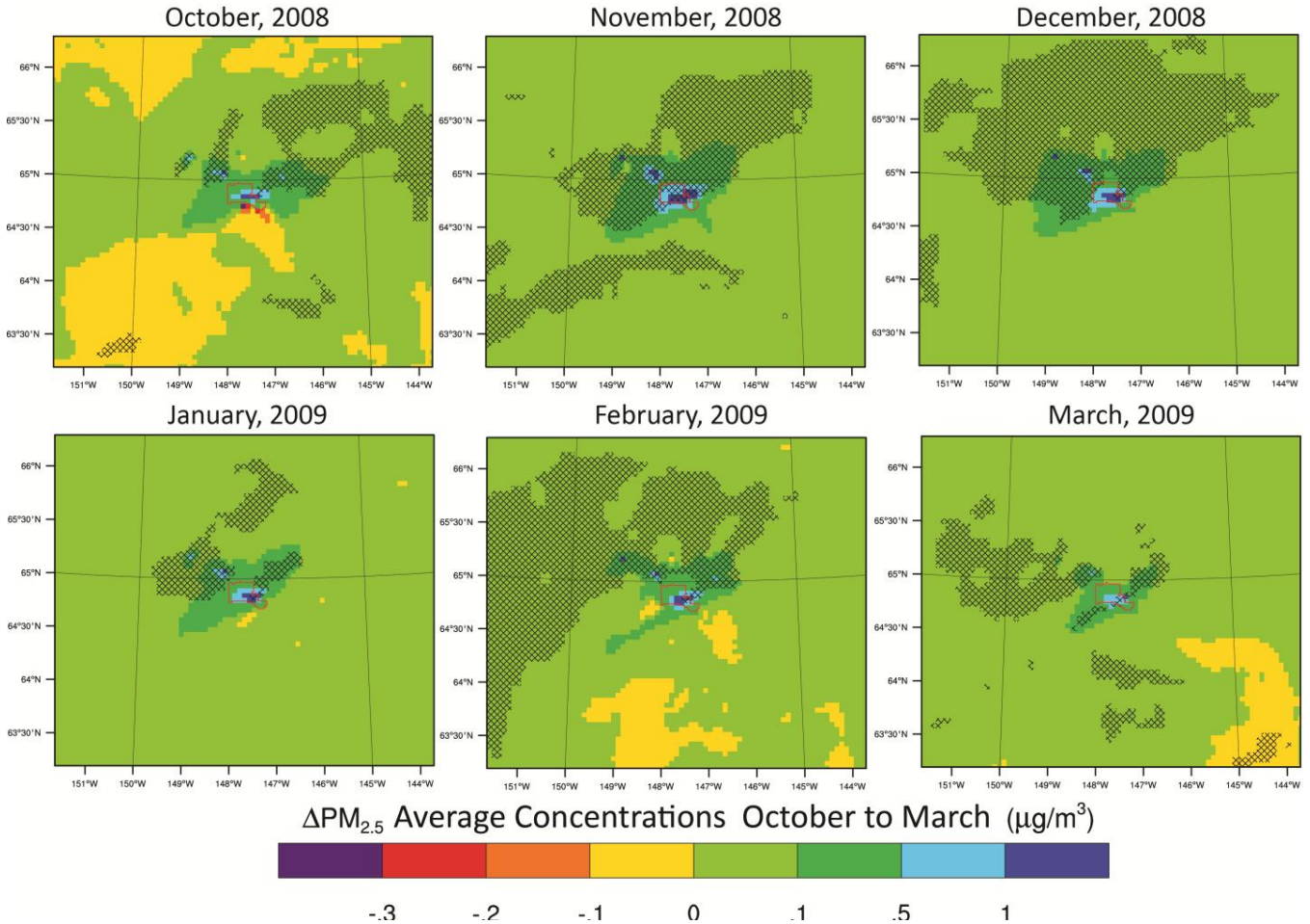

Figure 2.3: Difference REF-LSF of monthly averaged 24h-average $PM_{2.5}$-concentrations (color) from October to March. The hashed shading indicates significant (95% or higher confidence level) differences. The red polygon indicates the boundaries of the Fairbanks nonattainment area.

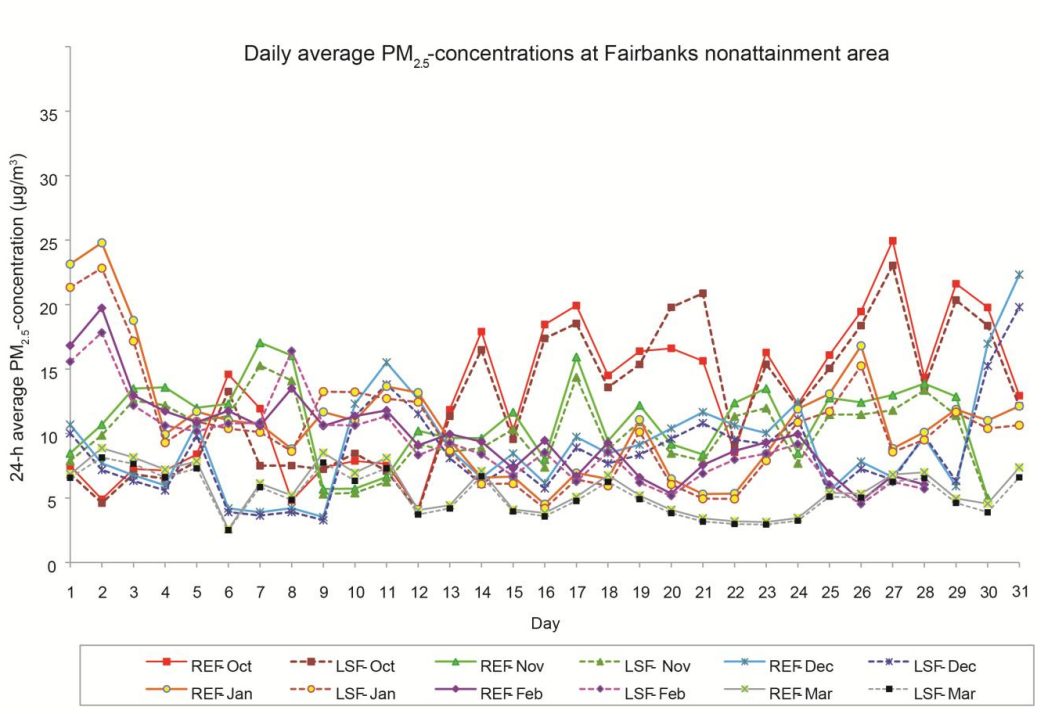

Figure 2.4: Temporal evolution of simulated 24h-average $PM_{2.5}$-concentrations averaged over the nonattainment area for October to March as obtained by REF and LSF.

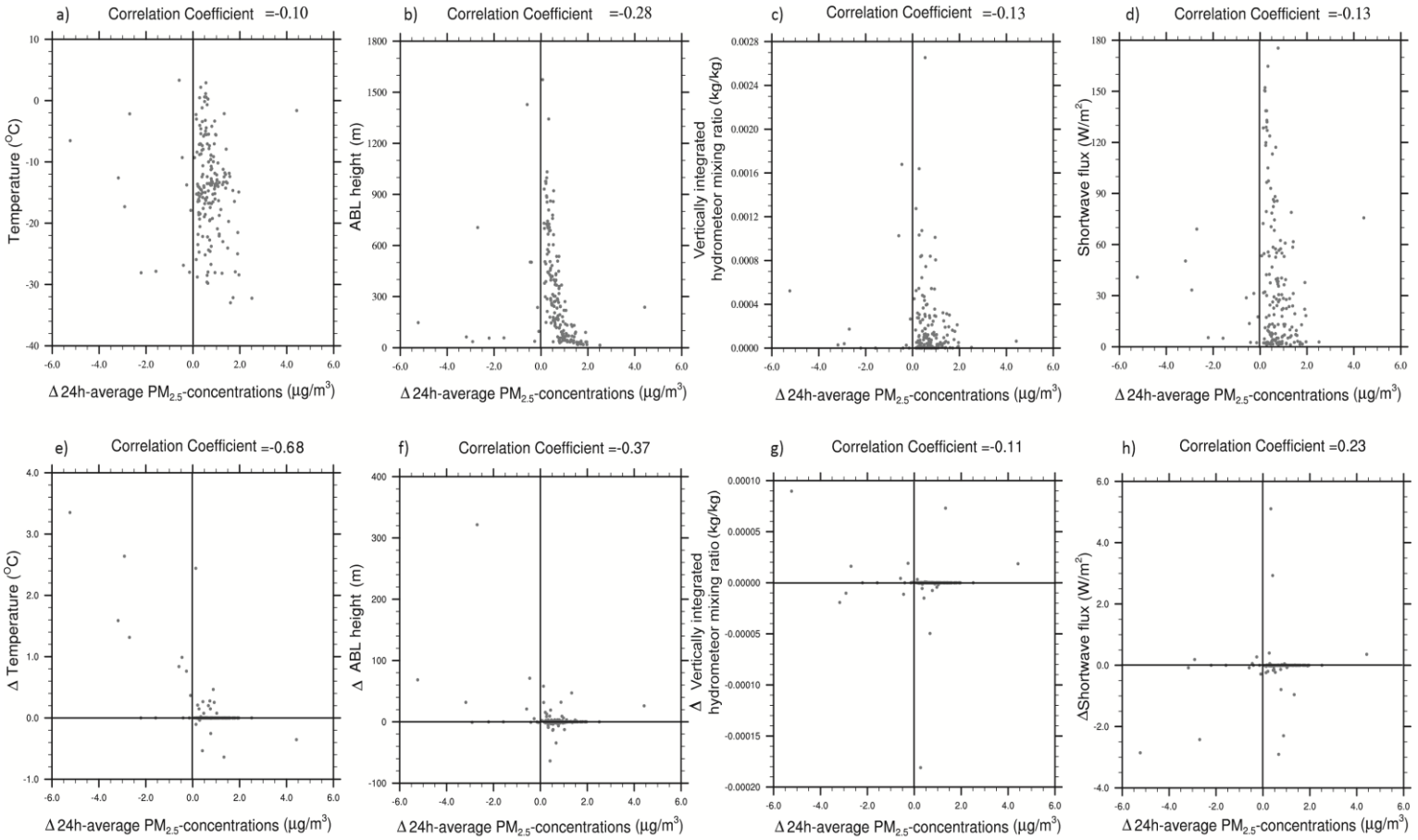

Figure 2.5: Scatter plots of the REF-simulation of 24h-average $PM_{2.5}$-concentrations vs. (a) near-surface temperature, (b) ABL-height, (c) vertically integrated hydrometeor mixing ratio, and (d) downward shortwave radiation, and differences REF-LSF of 24h-average $PM_{2.5}$-concentrations vs. difference (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 non-differences with respect to the values at the x- and y-axis.

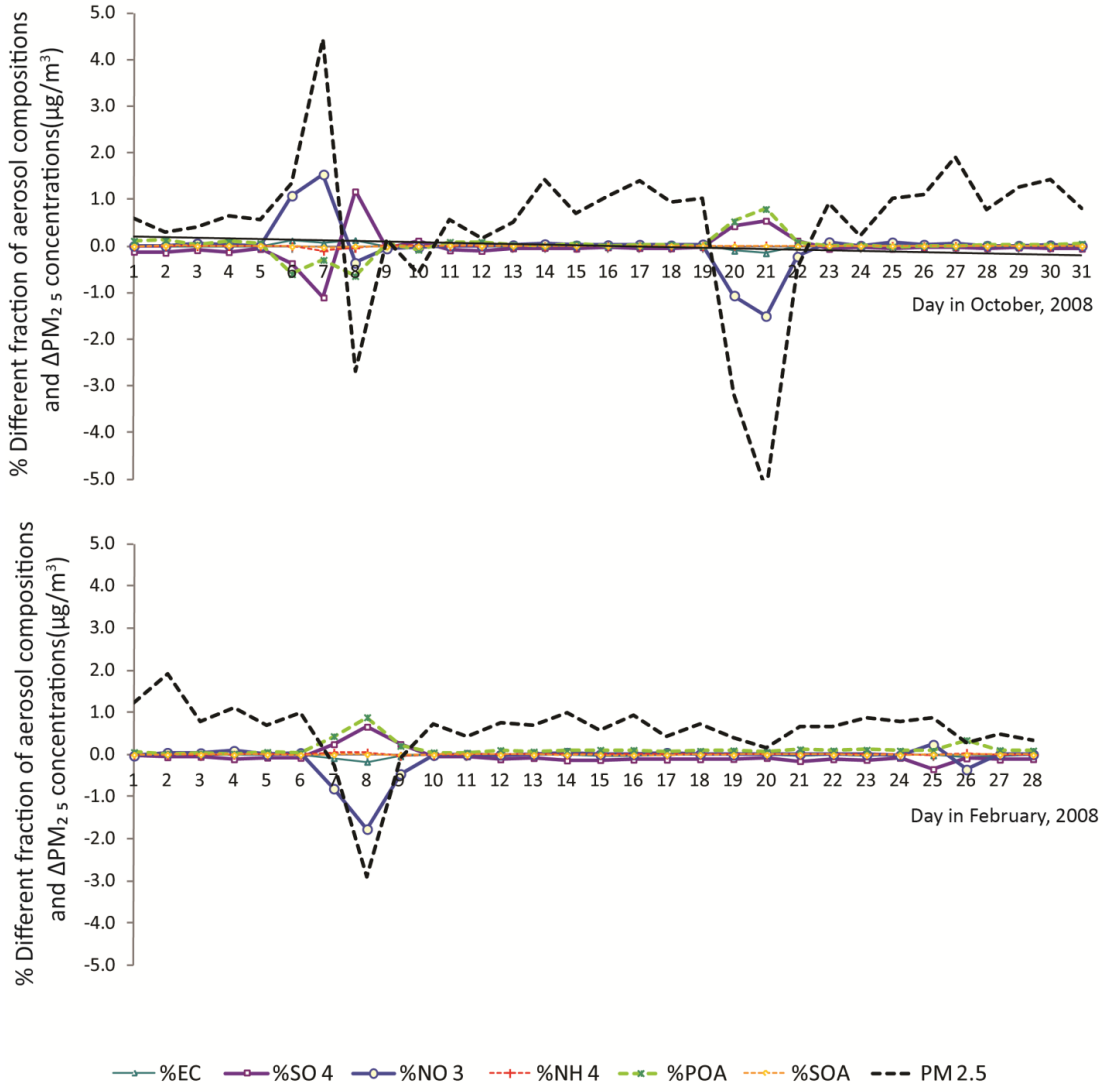

Figure 2.6: Temporal evolution of daily average percent differences in simulated aerosol compositions in the nonattainment area as obtained for October and February.

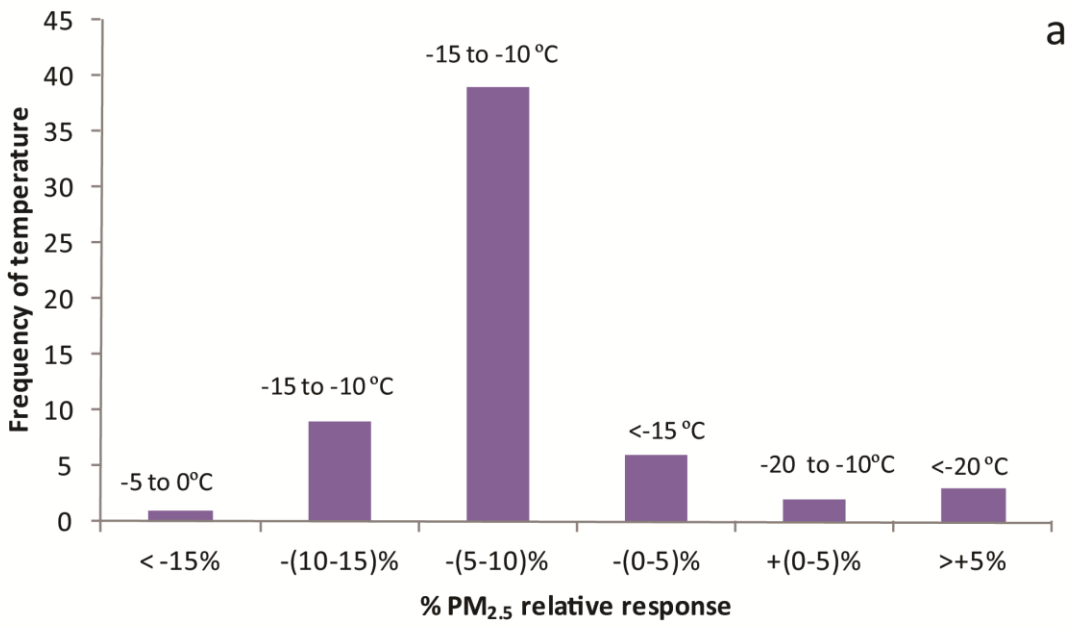

Figure 2.7: Relative responses of (a) total $PM_{2.5}$-, (b) sulfate-, and (c) nitrate-concentrations to the assumed fuel sulfur content reductions and (d) relation of relative responses of sulfate and nitrate at different temperature ranges. The temperature ranges on each bar are the ranges of temperature which has the most frequent occurrence, and the y-axis indicates that frequency.

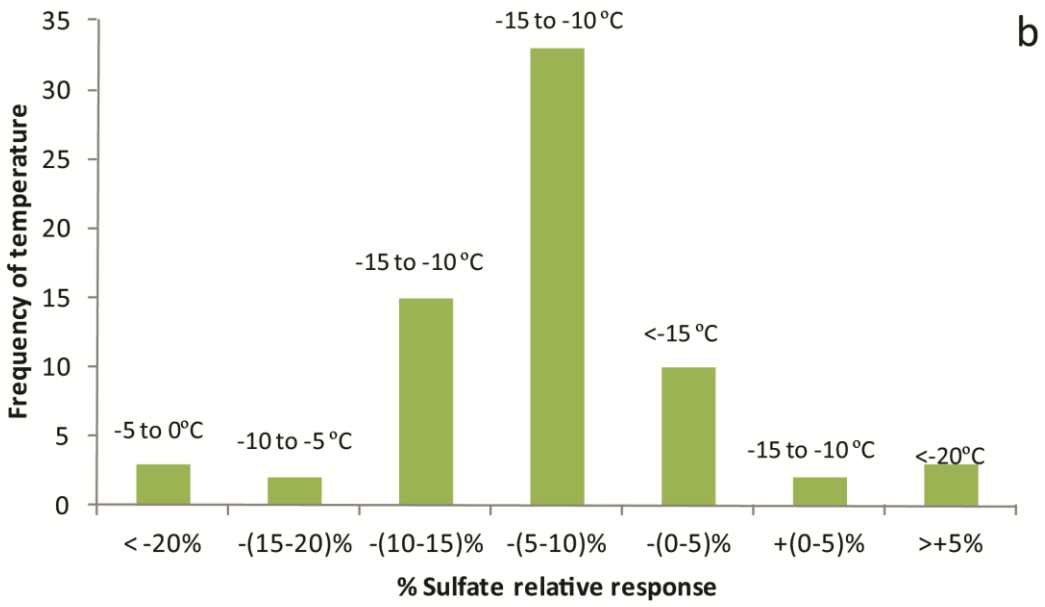

Figure 2.7 (cont.)

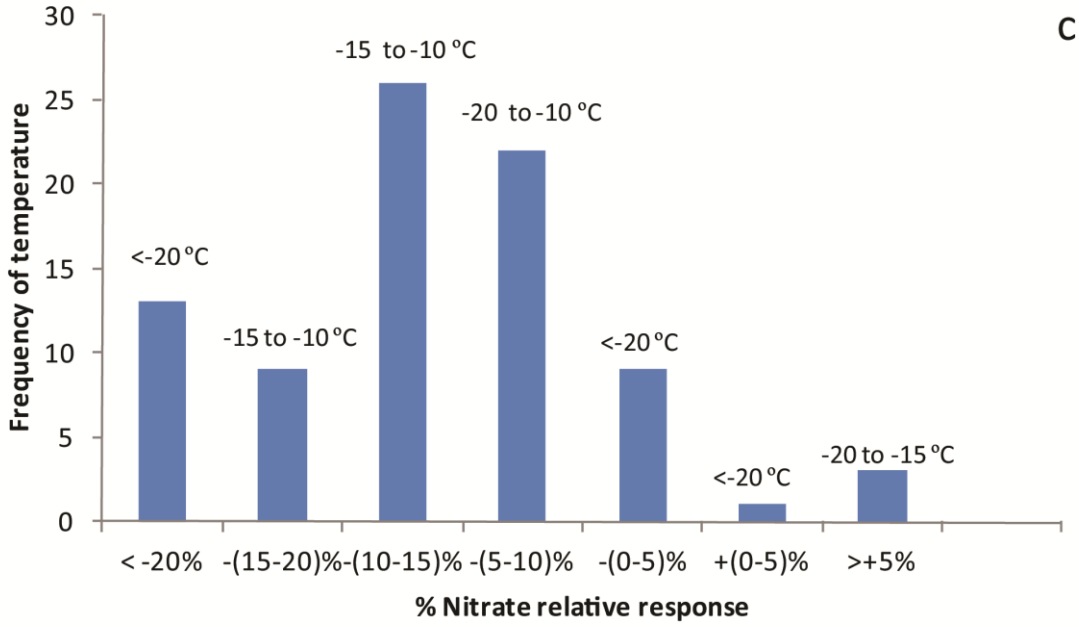

Figure 2.7 (cont.)

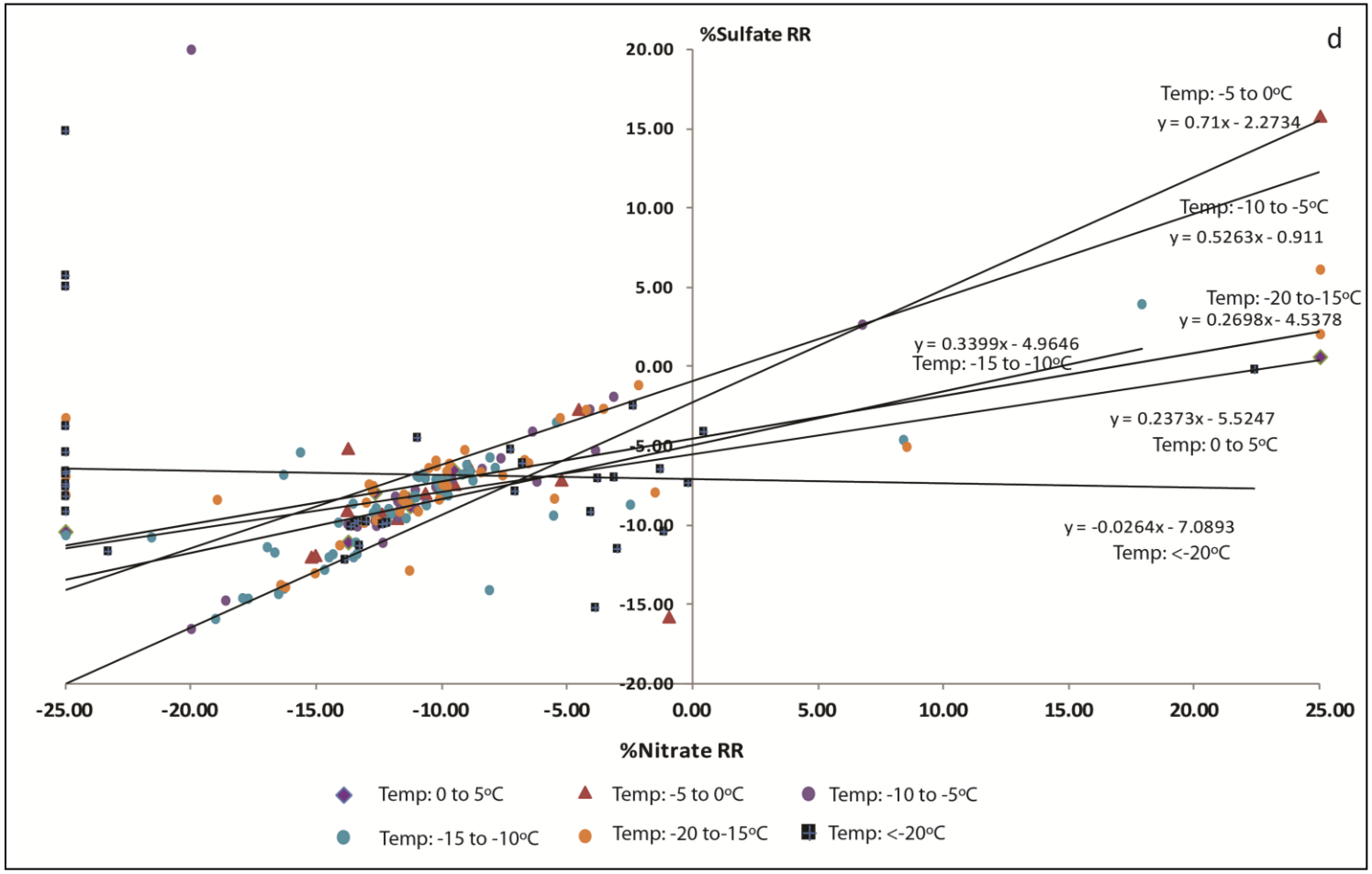

Figure 2.7 (cont.)

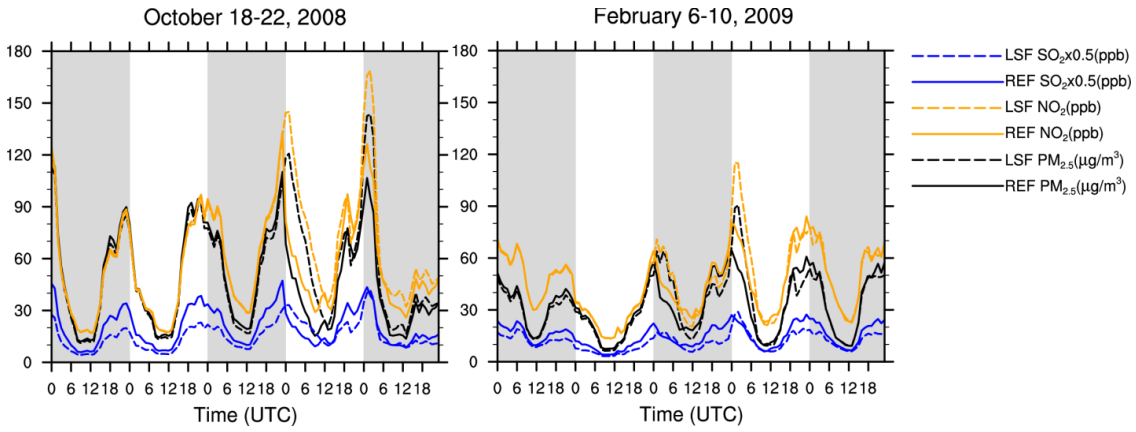

Figure 2.8: Mixing ratios of chemical species and $PM_{2.5}$-concentrations as obtained by REF and LSF for the gird-cell that holds the monitoring site for October 18-22, 2008 and February 6-10, 2009 (UTC). The grey color serves to better distinguish among days.

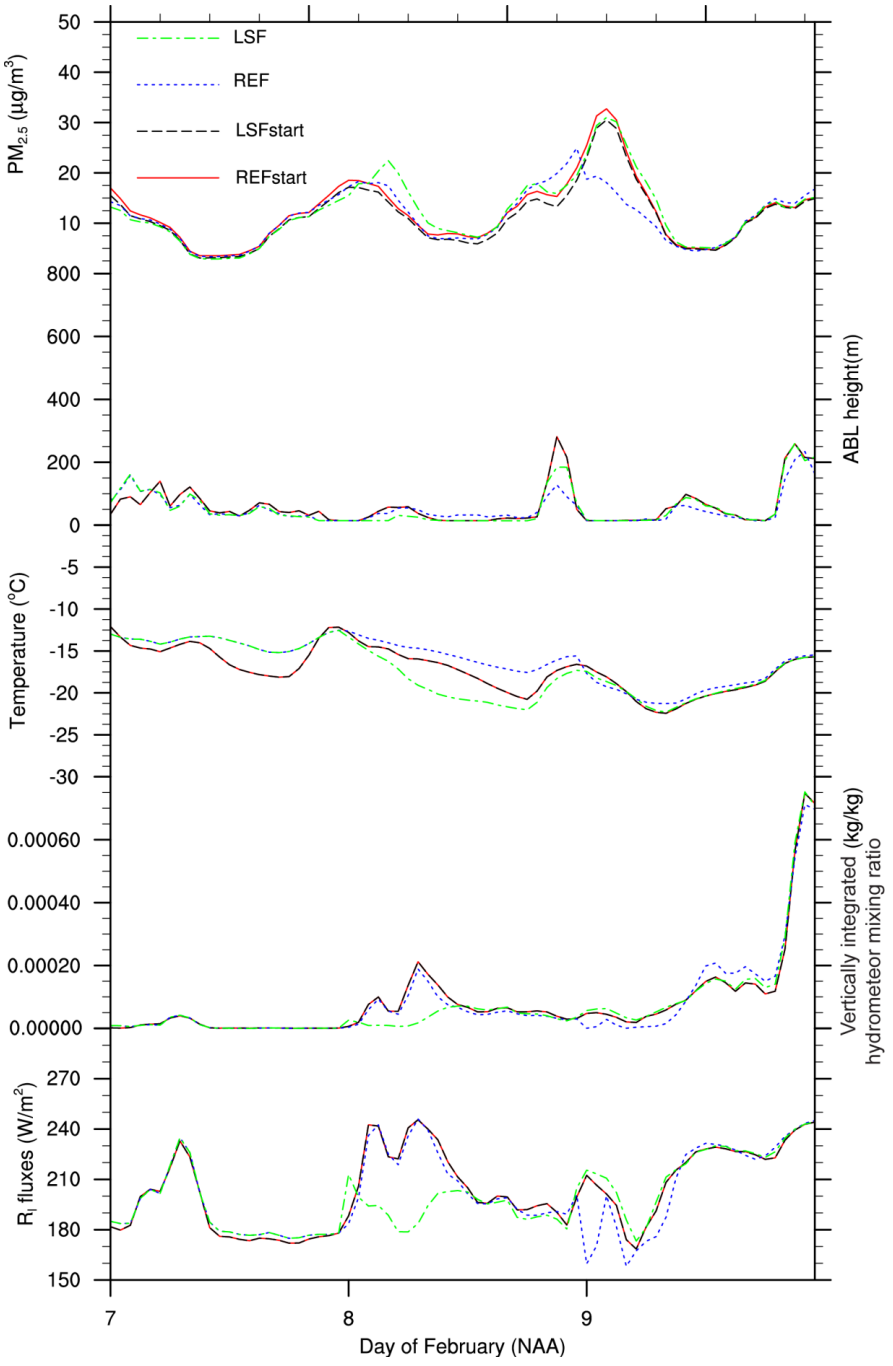

Figure 2.9: Meteorological variables and $PM_{2.5}$-concentrations simulated by REF, LSF and REFstart and LSFstart averaged over the nonattainment area during February 7-9, 2009 (UTC).

CHAPTER 3

Conclusions and Recommendations

This thesis study 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 Fairbanks, Alaska that is surrounded by a taiga landscape with hardly any anthropogenic emission sources. The episode of interest covered 1 October 2008 to 31 March 2009. In doing so, simulations were performed with the Alaska-modified WRF/Chem in forecast mode for October to March (a full cold season). WRF/Chem was found to perform acceptably for $PM_{2.5}$ and acceptably to well for the meteorological quantities [1].

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 Fairbanks nonattainment area and the grid-cell holding the monitoring site at the State Office Building, respectively; it also would avoid five exceedance days. 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 $44.7\mu g/m^3$ for Fairbanks and these RRFs, one has to conclude that introducing low-sulfur fuel will achieve a concentration of $42.9\mu g/m^3$. Thus without other emission-control measures, the Fairbanks nonattainment area 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 this emission control measure would be most efficient on very cold days with low ABL-heights, low shortwave radiation and low hydrometeor mixing ratios.

For the study in this thesis, WRF/Chem was run in forecast mode with reinitialization of the meteorology every five days for the entire cold season from 1 October 2008 to 31 March 2009 which 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 six days, the spinup of meteorology, and the aerosol-radiation feedback led to nonlinear processes that favored nitrate-aerosol formation. When removing these six days from our analysis, the RRFs decreased to 0.94; that is, this model artifact did not affect the conclusions of this study.

It is noteworthy that both the reference simulation as well as the low sulfur fuel scenario simulation data were affected by the initialization. However, it is the comparison of the simulations that made the model artifact obvious. Therefore, it would be worth investigating whether other ways to "patch" long-term simulations together would provide similar artifacts and to find a way that has the least impacts on the simulated chemical fields.

To avoid these artifacts due to model spinup effect in future studies, it should be tested to perform meteorological simulations in an overlapping manner (e.g. [2]. [3]). In this manner, typically the first six to 12 hours or so are discarded to allow clouds to spinup and, hence, to obtain more realistic radiation flux densities. This means in a

simulation a given amount of initializing hours is discarded from the analysis. This overlapping technique can be applied for both forecast mode [2] and nudging mode [3].

One recommendation for future investigations is to examine the impacts of such artifacts in the meteorological fields on the simulated $PM_{2.5}$-concentrations and composition with a different air quality model (e.g., Community Multiscale Air Quality model [4]).

Furthermore, to confirm the results and conclusions, a low sulfur scenario should be performed with another air quality model to examine whether the response to the changed emissions is independent of the model. Future work in this area should also be done at a finer scale (e.g. 1.3km) to see how the response varies with the model resolution.

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APPENDIX A

Contributions to Thesis Chapters

A.1 Chapter 2

The key topic of this chapter was adapted from Professor Nicole Mölders' grant LGFEEQ. The Alaska adapted WRF-Chem simulations (both reference simulation and mitigation simulation) were performed by Professor Nicole Mölders. The annual emission inventory for year 2008 was conducted by Huy N.Q. Tran. The literature research, analysis, text and figures were prepared by Ketsiri Leelasakultum. Professor Nicole Mölders helped, guided, and mentored Ketsiri Leelasakultum in the physical interpretation and refining of the text and the figures.