Numerical investigations on the contribution of point source emissions to the PM$_{2.5}$ concentrations in Fairbanks, Alaska

Huy N.Q. Tran, Nicole Mölders

University of Alaska Fairbanks, Geophysical Institute and College of Natural Science and Mathematics, Department of Atmospheric Sciences, 903 Koyukuk Dr., Fairbanks, AK 99775–7230, USA

ABSTRACT

Simulations with and without consideration of emissions from point sources were performed with the Weather Research and Forecasting model with online chemistry (WRF/Chem) to examine the contribution of point source emissions to the PM$_{2.5}$ concentrations at breathing level in Fairbanks, Alaska during winter. On days and at locations where PM$_{2.5}$ concentrations exceed the National Ambient Air Quality Standard of 35 $\mu$g m$^{-3}$, emissions from point sources account for 4% of the 24 h–average PM$_{2.5}$ concentration on average. The locations of highest concentrations were the same in both simulations. Point source emissions induced only five additional exceedance days in the nonattainment area. The magnitude of the PM$_{2.5}$ concentrations depended on meteorological conditions (temperature, wind speed, mixing height) and emissions. The radius of impact of point source emissions on the PM$_{2.5}$ concentration at breathing level of about 10–12 km downwind results as a combination of low emission heights, low wind speeds and the presence of inversions.

1. Introduction

Various studies showed epidemiological relationships between particulate air pollution and mortality and/or morbidity due to cardiovascular and pulmonary diseases, and adverse health effects caused by particulate matter under both short–term and long–term exposure (Dominici et al., 2006; Pope and Dockery, 2006). In response to these findings, the U.S. Environmental Protection Agency has tightened the National Ambient Air Quality Standard (NAAQS) for the 24 h–average concentration of particulate matter with diameters of 2.5 $\mu$m or less (PM$_{2.5}$) to 35 $\mu$g m$^{-3}$ in 2006. Thus, days with PM$_{2.5}$ concentration exceeding this NAAQS at the official monitoring site in a community are considered as exceedance days.

In Fairbanks, the PM$_{2.5}$ concentrations monitored at the official monitoring site have frequently exceeded the new NAAQS in the cold season, especially from November to February, in the previous years (Tran and Mölders, 2011). Thus, Fairbanks was assigned as a PM$_{2.5}$ nonattainment area. Achieving and remaining in compliance with the new NAAQS requires developing strategies for emission reduction. Such strategies require detailed knowledge about the emission sources, behavior and fate of PM$_{2.5}$. In the atmosphere, PM$_{2.5}$ may stem from direct emission (primary particles) or gas–to–particle conversion (secondary particles). The secondary particles comprise mainly ammonium sulfate and ammonium nitrate from reactions between ammonia and sulfuric and nitric acids.

Numerical modeling is a useful tool to assess the contribution of different emission sources to the pollutants’ concentrations. Cheng et al. (2007), for instance, applied the Mesoscale Model generation 5 (MMS) and the Advanced Regional Prediction System coupled with the Models–3/Community Multiscale Air Quality model to assess the emission source contributions to the PM$_{10}$ concentrations in the Beijing area. They identified emissions from industries, construction sites and road dusts as the major contributors. A study conducted for the Pearl River Delta region, China with the MM5–STEM–2K1 modeling system identified power plants as the major contributors to sulfur dioxide (SO$_2$) concentrations, and traffic as the main contributor to the NO$_x$ (NO+NO$_2$), nitric oxide and nitrogen dioxide), carbon monoxide (CO) and ozone (O$_3$) concentrations (Wang et al., 2005). Frost et al. (2006) applied the Weather Research and Forecasting model (Skamarock et al., 2008) with online chemistry (Grell et al., 2005) to investigate the impact of decreased power plant NO$_x$ emissions on O$_3$ concentrations. They found that O$_3$ concentrations generally decreased with the magnitude of the NO$_x$ emissions and depended on whether the NO$_x$ emission reduction yielded a plume that was in a high or low NO$_x$ regime. Ying et al. (2009) used WRF/Chem to investigate the sensitivity of O$_3$ concentrations to the diurnal variations of surface emissions in Mexico City. They found that morning emissions of volatile organic compounds (VOCs) and NO$_x$ both determined daytime O$_3$ concentrations, and that the O$_3$ production in Mexico City is VOC–limited. Chapman et al. (2009) performed WRF/Chem simulations to assess the impact of altered emissions from elevated point sources on aerosol radiative forcing.

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Corresponding Author:

Nicole Mölders

Tel: +1-907-474-7910

Fax: +1-907-474-7290

E-mail: molders@gi.alaska.edu

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Knowledge on air quality in high–latitude cities, especially in Alaska, is scarce (Mölders et al., 2011; Mölders et al., 2012). Fairbanks and its vicinity have four power plants and various other point sources. In Fairbanks during winter, surface–based and low–level inversion layers frequently exist (Mölders and Kramm, 2010; Tran and Mölders, 2011). These inversions may either enhance or reduce the impacts of point source emissions on the PM$_{2.5}$ concentrations at breathing level depending on whether the point sources emit into, above or below the inversion layer.

The National Emission Inventory of 2005 (NEI, 2005) shows that in Fairbanks, point source emissions contributed up to 15% of the total PM$_{2.5}$ emission. If point source emissions were found to tremendously contribute to the 24–h average PM$_{2.5}$ concentrations in the Fairbanks nonattainment area, controlling these emissions would be an effective tool to reduce the number of exceedance days. Advanced pollution control techniques for point sources are namely easier to implement and manage than controlling area emissions (e.g. residential heating, traffic).

The goal of this study is to examine the contribution of point source emissions on the 24–h average PM$_{2.5}$ concentrations at breathing level in the Fairbanks nonattainment area. In doing so, we performed and analyzed WRF/Chem simulations with and without inclusion of point source emissions.

2. Experimental Design

2.1. Simulations

We used the WRF/Chem with the modifications for Alaska and the physical and chemical schemes described and evaluated in Mölders et al. (2011). The WRF Single–Moment six–class cloud–microphysics scheme (Hong and Lim, 2006) served to simulate cloud and precipitation formation. This scheme considers mixed–phase processes and the coexistence of super–cooled water and ice. Cumulus convection was treated using the 3D–version of the cumulus–ensemble approach available in WRF (Skamarock et al., 2008). This scheme is a further development of Grell and Devenyi (2002) parameterization. Heat and moisture exchange at the land–atmosphere interface was treated with a modified version of the Rapid Update Cycle Land–Surface Model (Smirnova et al., 2000). Turbulent processes in the atmospheric boundary layer and surface layer were calculated in accord with Janjic (2002). Atmospheric radiative transfer was determined by the Rapid Radiative Transfer Model (Mlawer et al., 1997) for long–wave radiation and by the Goddard scheme (Chou and Suarez, 1994) for shortwave radiation. Gas–phase chemistry was represented by Stockwell et al. (1990) chemical mechanism which includes 21 inorganic and 42 organic species, and considers 156 chemical reactions. Dry deposition of trace gases was treated following Wexley (1989) with the modification by Mölders et al. (2011). The Secondary Organic Aerosol Model (Schell et al., 2003) and Modal Aerosol Dynamics Model for Europe (Ackermann et al., 1998) served to describe aerosol chemistry and physics including inorganic and secondary organic aerosols, wet and dry removal of aerosols. Direct and indirect feedbacks of aerosols to radiation schemes were considered (Barnard et al., 2010).

The domain of interest for the analysis encompasses the Fairbanks nonattainment area and its adjacent land with 80×70 grid cells and a 4 km increment (Figure 1). There are 28 stretched vertical layers from the surface to 100 hPa. The first layer is 8 m thick and referred to as breathing level, hereafter. The 1×1° and 6 h–resolution global final analyses data obtained from the National Centers for Environmental Prediction was downscaled to provide the meteorological initial and boundary conditions. The meteorology was initialized every five days. The initial conditions for the chemical fields stemmed from a simulation started with Alaska typical background concentrations 14 days prior to November 1, 2005.

Pleim (2011) showed that advection can strongly impact the pollutants’ concentrations. Numerical studies (Tran et al., 2011) as well as observational studies with backwards trajectory modeling (Cahill, 2003; Mölders et al., 2012) showed that in Alaska, advection of pollutants marginally affects the background concentrations. In March, when advection is the largest it elevates the PM$_{2.5}$ concentrations at Denali Park from less than 0.5 µg m$^{-3}$ to about 2 µg m$^{-3}$. Furthermore, the next closed city to the Fairbanks nonattainment area (Anchorage) is 578 km away on the other side of the Alaska Range of which the highest peak is 6 193 m (Mt. McKinley). Therefore, and as the focus of this study is on the impact of point sources in the vicinity of Fairbanks on the PM$_{2.5}$ concentrations at breathing level in the nonattainment area, we assumed Alaska–typical background concentrations (e.g., acetylene, CH$_3$CHO, CH$_3$OOH, CO, ethane, HCHO, HNO$_3$, H$_2$O$_2$, isoprene, NO$_x$, O$_3$, propene, propane, SO$_2$) as lateral boundary conditions.

Anthropogenic emissions from the NEI2005 for Alaska were allocated into space dependent on point source facility coordinates, land use, road network, and population density data, and into time (month, day of the week, hour) according to source profiles’ specific local activities. Plume rise calculations were based on Peckham et al. (2009) which considered stack height, exit velocity, exit temperature, ambient temperature and wind speed. The assumed split for emitted PM$_{2.5}$ was 46% organic carbon (OC), 20% sulfate (SO$_4$), 5% nitrate (NO$_3$), 9% elemental carbon (EC) and 20% other fine primary PM$_{2.5}$ aerosols. Biogenic emissions were calculated online according to Simpson et al. (1995).

WRF/Chem simulations were analyzed for November 1, 2005 0000 Alaska Standard Time (AST) to March 1, 2006 0000 AST with (REF) and without (NPE) inclusion of emissions from point sources.

2.2. Analysis

The number, frequency and locations of grid cells with PM$_{2.5}$ exceedences in REF and NPE were compared to assess the contributions of point sources to exceedances. We considered a grid cell as experiencing an exceedance when its 24–h average PM$_{2.5}$ concentration was greater or equal to 35 µg m$^{-3}$. We counted a day as an exceedance day when it had an exceedance at least at one grid cell in the nonattainment area.

In the following, the 24–h average PM$_{2.5}$ concentrations refer to AST. We tested the hypothesis that point source emissions do not govern the 24–h average PM$_{2.5}$ concentrations at breathing level at the 95% confidence level according to a t-test. In addition, a false–ensemble analysis was applied to further examine whether the point source emissions affect the PM$_{2.5}$ concentrations at breathing level. Moreover, we examined the various correlations for their significance. In the following, the word significant is only used when data passed the t–test at the 95% level of confidence.

The contributions of point source emissions to the 24–h average and hourly PM$_{2.5}$ concentrations were assessed by the concentration differences (REF–NPE) called 24–h differences and 1 h–differences hereafter, respectively. We assessed the effects of the meteorological conditions (wind speed–v, temperature–T, mixing height–h$_{mix}$, sea level pressure, relative humidity,
downward shortwave radiation), point source and non-point source emissions on the 24 h–average PM$_{2.5}$ concentrations and 24 h–differences at breathing level by their cross–correlations. We used a linear regression analysis to evaluate the importance of the meteorological conditions and emissions. We started this analysis with the “predictant” (simulated PM$_{2.5}$ concentrations) and all “predictors” (point source emissions, non–point source emissions, simulated $T$, $v$, $h_{mix}$, relative humidity, sea level pressure, downward shortwave radiation) of interest as variables. We repeated the analysis by alternatively removing one of the “predictors” from the analysis and evaluated the coefficient of determination ($R^2$). The largest decrease of $R^2$ in response to the removal of a “predictor” identifies that “predictor” as the one with highest impact on the PM$_{2.5}$ concentration.

We investigated the impact radius of the point sources by analyzing the 24 h–differences along the cross–sections through the downwind of each point source, and by analyzing the correlation between the point source emissions at the emission level and the 1 h–difference at each model layer below the emission level. Since wind direction determines the pollutants’ transport direction and the locations, the pollutants’ impact, we only considered the 1 h–differences in grid cells located downwind of the grid cell that holds the point source. We considered 16 wind direction sectors of 22.5° each. We excluded hours with strong wind direction shears (>90°) at any level of interest from the analysis. Such wind direction shears occurred in less than 5% of the total hours. For each level and sector in steps of 4 km, the 1 h–differences were interpolated and averaged over the area covered by that sector. These values were used to calculate the correlation with the point source emissions for November to February. Distances with continuously significant correlation coefficients were considered as being impacted by the respective point source. The locations closest to the point source of interest with the highest significant correlation coefficient were considered as those that experience the highest impact from the point source’s emissions. Note that other interpolation methods led to similar results.

We examined the correlation behavior of each point source under consideration of potential impacts by other point sources. Once correlation becomes non–significant and then significant again and/or increases in the downwind of point sources that are downwind of the point source of interest, we attributed this change to the impact of the downwind point source(s) rather than the point source examined originally. Note that the diurnal activity allocation functions were the same for all point sources.
3. Results

3.1. Evaluation

Mölders et al. (2011b) evaluated the reference simulation by data from Doppler sound detection and ranging, twice–daily radiosondes, 33 surface meteorological and four aerosol sites. They found average biases over November to February and all meteorological sites of 1.6 K, 1.8 K, 1.85 m s$^{-1}$, $-5^\circ$, and 1.2 hPa for temperature, dew–point temperature, wind speed, wind direction, and sea level pressure, respectively. The Doppler sound detection and ranging data indicated under/over estimation of wind speed in the upper (lower) atmospheric boundary layer and good performance in capturing the presence of low level jets.

Mölders et al. (2011a) evaluated WRF/Chem’s performance in simulating PM$_{2.5}$ by data from the State Office Building site in downtown Fairbanks and a remote site in Denali Park. WRF/Chem simulated PM$_{2.5}$ at the urban site better than at the remote site. It captured the temporal evolution of 24 h–average PM$_{2.5}$ at the Fairbanks site broadly. Here the overall bias and correlation of hourly (24 h–average) observed and simulated PM$_{2.5}$ were 4.9 (4.0) μg m$^{-3}$ and 0.31 (0.59; all statistical significant), respectively. Over November to February, 41% (50%) of the simulated and observed PM$_{2.5}$ (SO$_2$ aerosol) concentrations agreed within a factor of two and the fractional bias was less than 30% on average over the two sites. Note that no other PM$_{2.5}$ data was available for our episode.

Obviously, some bias exists in the PM$_{2.5}$ concentrations (Mölders et al., 2011). Investigations on the sensitivity of PM$_{2.5}$ concentrations to biases in temperature showed marginal impact of temperature errors on simulated PM$_{2.5}$ concentrations except for temperatures close to the temperature threshold for particle formation (Mölders et al., 2012). Since REF and NPE used the same model setup, and the radiation aerosol feedback hardly impacted the meteorological quantities most of the time, biases in PM$_{2.5}$ concentrations due to errors in simulated meteorological quantities can be assumed to be similar in REF and NPE. Bias due to errors in biogenic emissions would be similar too as both simulations calculated biogenic emissions inline depending on the meteorological conditions. Both simulations also used the same emissions for the non–point source sector. Thus, we can assume that REF and NPE were affected the same by errors from these sources. This means that biases in PM$_{2.5}$ concentrations due to errors in simulated meteorological conditions, biogenic and area emissions cancel each other out when differences are examined. Point source emissions are the best regulated, controlled and verified emissions, for which we can assume that biases in PM$_{2.5}$ concentrations due to errors in point source emissions are marginal.

3.2. Point source emissions

In the domain of interest, 27 stacks emit into the levels between the second (8–16 m) and the seventh model layer (343–478 m). Among these, some stacks belong to the same facility or stacks from different facilities exist in the same grid cell. In WRF/Chem, like other photochemical models, all stacks located within the same grid cell are lumped, but emit into the layers into which the individual stacks would emit. Due to the lumping, only the joint impacts of point sources within a grid column can be investigated. These columns are denoted PS1 to PS9, hereafter (Figure 1). Three point source holding columns (PS4, PS5, and PS6) are located in the nonattainment area. PS6 has the highest PM$_{2.5}$ emission rate (3 g m$^{-2}$ h$^{-1}$), followed by PS7 (1.3 g m$^{-2}$ h$^{-1}$). Within the nonattainment area, PS4 has the second highest, but 19 times lower PM$_{2.5}$ emissions than PS6. PS4 has the highest emissions of SO$_2$ (0.6 g m$^{-2}$ h$^{-1}$) and NO$_x$ (0.5 g m$^{-2}$ h$^{-1}$), which are important precursors for PM$_{2.5}$ formation via gas–to–particle conversion, followed by PS6 with 0.24 g m$^{-2}$ h$^{-1}$ SO$_2$ and 0.18 g m$^{-2}$ h$^{-1}$ NO$_x$ emissions.

On average over November to February and the domain, the PM$_{2.5}$, SO$_2$, NO$_x$ and VOC emissions from point sources made up 15%, 42%, 42% and 0.6% of the total emissions in the domain, respectively. Within the nonattainment area, point source emissions made up 15%, 36%, 35% and 0.4% of the total PM$_{2.5}$, SO$_2$, NO$_x$ and VOC emissions, respectively. During November to February only non–point sources emitted ammonia and their emission rate was low (0.17 kg km$^{-2}$ h$^{-1}$).

3.3. General features

The phase and amplitude of the diurnal cycle of simulated PM$_{2.5}$ concentrations varied strongly among days. In general, the PM$_{2.5}$ concentrations showed a distinct peak around 0300 AST and a stronger, broader peak around 1300 AST. In general, high 24 h–average PM$_{2.5}$ concentrations occurred when PM$_{2.5}$ emissions were relatively strong (>0.2 g m$^{-2}$ h$^{-1}$) and concurrently the wind was calm (<0.5 m s$^{-1}$), air temperatures were low (below −20°C) and mixing heights were shallow (<20 m). In the nonattainment area, calm wind occurred 20% of the time and concentrations >35 μg m$^{-3}$ occurred on 46% of the calm wind events. Out of the 24 h–average PM$_{2.5}$ concentrations >35 μg m$^{-3}$, 62% (81%) occurred when air temperatures (mixing heights) were low (shallow). Shallow mixing heights (low temperatures) existed 33% (40%) of the time in November to February. Such shallow mixing heights typically occurred when WRF/Chem simulated surface–based inversions and calm wind over the nonattainment area.

At breathing level and between 100 and 200 m above ground, three and four distinct circulation patterns, respectively, existed that frequently coincided with exceedance days. In the nonattainment area, exceedances occurred on days with calm winds from various directions when the air remained in town (Figure 2a). Exceedances also occurred under calm wind conditions when the Fairbanks’ air drained toward southwest or air moved into Fairbanks from the southeast (Figure 2b). In the latter case, polluted air advected from the community of North Pole (22 km southeast of Fairbanks in the nonattainment area) may contribute to the exceedances. Simulated exceedances were often associated with the following airflows between 100 and 200 m above ground: (1) air moved slowly above town down the Tanana Valley to the southwest, (2) air slowly moved over Fairbanks from the southeast and down the valley to the southwest (Figure 2c), (3) air moved southeast up the valley, or (4) air drained to both sides of Fairbanks (Figure 2d). For November to February, WRF/Chem simulated 12 exceedances when air masses that passed over Fairbanks and took up pollutants (Figure 2e), moved back into Fairbanks thereby advecting aged polluted air (Figure 2f). The simulations showed that winds from north or northeast with v>2.5 m s$^{-1}$ typically advected clean air into Fairbanks that diluted the pollutants’ concentrations efficiently and/or moved the polluted air out of town to the west or southwest.

3.4. Contribution of point source emissions

In November to February, the highest 24 h–average PM$_{2.5}$ concentrations in REF and NPE anywhere in the domain differed 1 μg m$^{-3}$ on average and barely exceeded 3 μg m$^{-3}$ locally (Figure 3). On 65 out of the 120 days, the highest 24 h–average PM$_{2.5}$ concentration in REF occurred in the grid cell holding the official monitoring site. On 38 and 17 days, highest 24 h–average PM$_{2.5}$ concentrations occurred in the grid cell adjacent to the south and west of the monitoring site, respectively. In NPE, the highest 24 h–average PM$_{2.5}$ concentrations occurred at the same locations and times as in REF except on 7 days. On these 7 days, however, they occurred still within the three grid cells mentioned above.
Figure 2. Circulation pattern of 10 m–wind (barbs) associated with exceedances at breathing level in the nonattainment area and 24 h–average PM$_{2.5}$ concentrations are underlaid for (a) November 26, 2005, (b) December 1, 2005, (c) January 11, 2006, (d) January 20, 2006, (e) January 15, 2006, and (f) January 16, 2006.

The 98%, 90%, 75%, 50% and 25% percentile of the 24 h–average PM$_{2.5}$ concentration in REF (NPE) were 35.7 (33.9), 24.0 (22.5), 17.1 (15.9), 10.8 (10.3), and 7.0 (6.8) μg m$^{-3}$, respectively. When and where the ten highest and ten lowest 24 h–average PM$_{2.5}$ concentrations occurred in the nonattainment area during November to February hardly differed between REF and NPE. These findings suggest that point sources marginally affected the spatial distribution of 24 h–average PM$_{2.5}$ concentrations in the nonattainment area on polluted ($25 \mu g m^{-3} \leq PM_{2.5} < 35 \mu g m^{-3}$) and hardly affected them on clean ($PM_{2.5} < 25 \mu g m^{-3}$) days.

Topography and wind direction influence the distribution of the mean 24 h–difference and its significance. During November to February, winds from east and northeast dominated. Small but statistically significant 24 h–differences occurred over a relatively large area including the nonattainment area and its downwind (Figure 4). Almost all notable 24 h–differences existed for grid cells holding point sources and their adjacent grid cells. On average over the domain, the nonattainment area and at the grid cell holding the official monitoring site, the 24 h–differences were 0.04, 0.8 and 1.2 μg m$^{-3}$, respectively which corresponds to 3.8, 1.2 and 3.9% reduction, respectively. In the nonattainment area, the highest 24
h–difference was 18 μg m\(^{-3}\) and occurred in the grid cell holding PS6 on January 27 2006 (Figure 3), while the highest 24 h–difference averaged over the nonattainment area was 4.5 μg m\(^{-3}\) on November 13, 2005. In 47% of the time, the highest 24 h–differences occurred at PS6 with 7 μg m\(^{-3}\) on average, and 5% of the time at other grid cells in the nonattainment area with 2.3 μg m\(^{-3}\) on average. During 48% of the time, most of the highest 24 h–differences occurred in the grid cells holding PS1, PS2, PS3 and PS8 with about 2.5 μg m\(^{-3}\) on average. Generally, the highest 24 h–difference occurred outside the nonattainment area on clean days when the 24 h–average PM\(_{2.5}\) concentrations in the nonattainment area were less than 25 μg m\(^{-3}\) and vice versa. The highest and second highest 24 h–differences frequently occurred at PS6 and its adjacent grid cells indicating the importance of PS6 for the 24 h–average PM\(_{2.5}\) concentration in the nonattainment area.

Despite the t-test indicated statistically significant concentration differences, a possibility remains that the difference is not due to contributions of point sources, but rather due to some variable random effects between the two simulations (e.g. truncation errors, model sensitiveness). This possibility is most likely for small (<1 μg m\(^{-3}\)) differences (Werth and Avisar, 2002) like they occurred in this study. To further assess whether the differences are due to the contribution of point sources, we adopted a false–ensemble analysis method that was developed and applied successfully in the analysis of climate–model scenarios (Werth and Avisar, 2002). This method bases on the concept that two simulations with no difference in the mean emissions and small random effects differ hardly in their mean concentrations.

For each month, we calculated the difference of the 24 h–average PM\(_{2.5}\) concentrations REF–NPE called the “true” difference hereafter. We created a set of “false REF” and “false NPE” ensembles by randomly replacing results of simulation days of REF (NPE) with the results of the corresponding simulation days of NPE (REF). The replacement was completed when the number of NPE (REF) simulation days made up 50% of the total days of the “false REF” (“false NPE”) ensemble. Since the emission rates differ among days, the generated false ensembles negligibly and non–significant differ in their monthly total emission depending on for which days the data were exchanged. In principle, \(n!(n/2)!\) false ensembles can be generated from \(n\) simulation days in the described way, i.e. in our case 10\(^{6}\) false ensembles for one month. We generated 450 false ensembles for each month to obtain a sufficiently large statistical basis. For each set of “false REF” and “false NPE” ensembles, the difference of the 24 h–average PM\(_{2.5}\) concentration was calculated. Finally, we ranked the true over the 450 “false” concentration differences. This procedure was applied for each grid cell.

The results of the false–ensemble analysis indicated that for most grid cells the true differences fall within the top 5% of all differences although the distribution of these grid cells differs among months (Figure 5). At grid cells inside the nonattainment area, the true concentration differences consistently fell in the top 5% throughout November to February except at 1, 5 and 1 grid cells in December, January and February, respectively. Thus, the false–ensemble analysis supports that the point sources contributed to the PM\(_{2.5}\) concentrations at breathing level, although the contribution was small on average.
During November to February, the NAAQS was exceeded on 10 (7), 6 (5), 22 (21) and 1 (1) days in REF (NPE) in November, December, January and February, respectively. The five exceedance days avoided in NPE had only slightly lower 24 h–average PM$_{2.5}$ concentrations (up to 5 μg m$^{-3}$) than REF. Out of the 104 (80) exceedances that were simulated anywhere in the nonattainment area at any time during November to February by REF (NPE), 37 (34), 29 (20) and 20 (18) exceedances occurred at the grid cell holding the monitoring site, and in the grid cells adjacent to its west and south, respectively (Figure 6). In REF, 3 and 5 exceedances occurred for the grid cell holding PS6 and the grid cells adjacent to it, respectively, and none of them occurred at these locations in NPE. The fractional difference of 24 h–average PM$_{2.5}$ concentrations \([\text{REF} - \text{NPE}] / \text{REF}\) indicated that on exceedances days, point sources contributed up to 42% to the total 24 h–average PM$_{2.5}$ concentration in the grid cell holding PS6 and up to 22% in the grid cells adjacent to it. At other locations, the fractional differences indicated that point sources accounted for 4% of 24 h–average PM$_{2.5}$ concentration on average and barely exceeded 10% on exceedance days. These findings mean that except for PS6 and its adjacent grid cells, non–point source emissions led already to high PM$_{2.5}$ concentrations and the point sources just added the small amount needed to exceed the NAAQS.

The speciation of PM$_{2.5}$ was almost identical in REF and NPE. For example, at the grid cell holding the monitoring site, the overall PM$_{2.5}$ speciation was 20.4, 2.2, 2.6, 9.0, 45.8, 19.9% SO$_4$, NO$_3$, NH$_4$, EC, OC and other fine particles, respectively, in REF, while it was 20.5, 2.1, 2.6, 9, 45.9 and 19.9% in NPE. Similar minor changes in PM$_{2.5}$ speciation were also found for the grid cell holding PS6. Recall that the emitted PM$_{2.5}$ split was 20, 5, 9, 46 and 20% for SO$_4$, NO$_3$, EC, OC and other fine particles, respectively. These values imply that secondary aerosol formation was low during November to February. This fact contributed to the small impact of point source emissions on the PM$_{2.5}$ concentrations at breathing level despite point sources made up 35% of the total SO$_2$ and NO$_x$ emissions.

At breathing level the 24 h–average PM$_{2.5}$ concentrations averaged over the nonattainment area obtained by REF correlated significantly with $v$, $T$, $h_{mix}$ and downward shortwave radiation ($-0.689$, $-0.537$, $-0.671$, $-0.220$), but non–significantly with relative humidity and sea level pressure. The 24 h–average PM$_{2.5}$ concentration in the nonattainment area correlated stronger and significantly with the non–point source emissions ($0.331$) than with the point source emissions ($0.231$). The linear regression analysis showed that non–point source emissions were the most important factors governing the 24 h–average PM$_{2.5}$ concentrations, followed by $T$, $v$, $h_{mix}$ point source emissions and downward shortwave radiation. These findings also support that non–point source emissions mainly contributed to the 24 h–average PM$_{2.5}$ concentration in the nonattainment area.

At the grid cell holding PS6, the 24 h–average PM$_{2.5}$ concentrations obtained by REF showed similar correlation with the emissions from non–point sources ($0.281$) and point sources ($0.275$). At PS6, the correlations of the 24 h–average PM$_{2.5}$ concentrations with $T$, $v$ or $h_{mix}$ were $-0.608$, $-0.628$ and $-0.592$, respectively. The linear regression analysis showed that at PS6, temperature was the most important factor, followed by non–point source emissions, point source emissions, and wind. Mixing height was least important for the 24 h–average PM$_{2.5}$ concentrations. However, $h_{mix}$ strongly correlated with $v$ ($0.874$) and $T$ ($0.507$).
At PS4, the 24 h-average PM$_{2.5}$ concentrations correlated with the non-point source emissions (0.337) but not with the point source emissions. The linear regression analysis indicated that at PS4, wind followed by non-point source emissions and temperature were the most important factors for the 24 h-average PM$_{2.5}$ concentrations. Similar behavior like for PS4 was found for PS1, PS2, PS3 and PS5 that all are outside, but not far from the nonattainment area. At PS7, PS8 and PS9, the 24 h-average PM$_{2.5}$ concentrations correlated significantly neither with the point source nor with the non-point source emissions. Instead, wind speed, temperature, mixing height and sea level pressure mainly governed the 24 h-average PM$_{2.5}$ concentrations. These point sources are located far from the nonattainment area (PS8, PS9) or in mountainous terrain (PS7) upwind of the nonattainment area (PS7, PS8). In their vicinity, winds were relatively strong (on average $v > 6$ m s$^{-1}$) and there were no non-point source emissions or only low point source emissions (e.g. PM$_{2.5} < 0.08$ g m$^{-1}$ h$^{-1}$) at PS8 and PS9). These conditions allowed strong dilution and marginal advection of pollutants from the nonattainment area. Therefore, at PS7, PS8 and PS9, the 24 h-average PM$_{2.5}$ concentration were more sensitive to meteorological than to emission conditions. Generally, at grid cells holding point sources, the 24 h-average PM$_{2.5}$ concentrations were typically stronger related to the meteorological conditions and non-point source emissions than to the point source emissions.

3.5. Radius of point source impacts

The impact radius differs among point sources and depends on emission height, wind speed and inversion conditions. On average over November to February, the 24 h-difference along the cross-sections C1 to C8 (see Figure 4 for location) centered over point sources were highest in the grid cells holding the point sources and at the level into which they emitted the strongest (Figure 7). At breathing level, a general feature was that point sources contributed most to the PM$_{2.5}$ concentration in the grid cell they are located.

Point sources exist at various places. Hence, point sources in their downwind induced interfering effects with the impact of the point source of interest (e.g. C3, C4, C7, C8). For example, in C5 that is centered on PS7, the second maximum located 20 km downwind of PS7 at about 150 m above ground was caused by emissions from PS6. The PM$_{2.5}$ concentration contributed by the point source of interest was highest right in the grid column it emitted into at the emission level. For regulatory questions, however, the concentration at breathing level is decisive. Therefore, we were interested in the impact of the point source emissions on the concentrations at breathing level. Thus, in the following the term “highest impact” refers to the location that has the highest concentration at the breathing level.

Emissions from PS6 (cross-sections C1 and C2 in Figures 4 and 7) had the strongest impact on the PM$_{2.5}$ concentrations in the grid cell where PS6 is located. This impact quickly decreased in its downwind. Cross-sections C7 and C8 document a similar behavior for PS2 like for PS6 (Figure 7). As shown in C5 and C6, at PS7, the polluted air was strongly diluted before reaching the breathing level because in the mountainous terrain of PS7, the wind was relative strong (on average $v > 6$ m s$^{-1}$). Consequently, PS7 rarely contributed to the breathing level PM$_{2.5}$ concentration in the nonattainment area.

At a point source of interest, due to overlapping effects of all emitting levels, correlation patterns of 1 h-differences with point source emissions at each emitting level were quite similar. Therefore, the impact of individual emission levels on the 1 h-differences cannot be clearly distinguished. Generally, the correlation patterns of the 1 h-differences with the point source emissions (Figure 8) agreed with the above findings that point sources contributed most to the PM$_{2.5}$ concentration at breathing in or very close to the grid cell holding it. Highest correlations occurred for PS6 with similar magnitude for all emission levels (0.26) indicating strong downward mixing of PM$_{2.5}$ from the emission levels to the breathing level. Based on our point source impact radius definition, we conclude that the impact radius of PS6 was about 12 km, and the highest impacted location is the grid cells holding PS6.

Lowest correlations between the 1 h-differences and point source emissions occurred at PS7 (Figure 8), PS8 (up to 0.052, significant) and PS9 (up to 0.088, significant). At these point sources, correlations at breathing level were lower than at upper levels. This finding indicates that the polluted air when it reached the breathing level had much lower PM$_{2.5}$ concentration than at the emission level. The impact radius of PS7 was about 10 km. The impact radius of PS8 was about 4 km due to its low height of emission levels (8–16 m) and the weak PM$_{2.5}$ emission rate (0.08 g m$^{-2}$ h$^{-1}$). PS9 had an impact radius >40 km as it emitted into levels up to 219–343 m. PS7, PS8 and PS9 exerted their highest impact at the grid cell holding the respective point sources.
Figure 7. Horizontal–vertical cross–sections C1 to C8 of average PM$_{2.5}$ differences (color) and of highest PM$_{2.5}$ differences (REF–NPE) during November to February (contours in steps of 1 $\mu$g m$^{-3}$). For locations of C1 to C8 see Figure 4. The point–source investigated is located at x=0.

At PS1, PS4 and PS5, interference effects by other point sources close to the point source of interest (Figure 8) made it difficult to determine clearly the impact radius. Typically all point sources had an impact radius of about 10 to 12 km, on average over November to February, but the radius differed with the wind speed at the emission level. Correlation patterns are quite similar for all point sources. Thus, we exemplarily discuss the behavior for PS6. Over November to February, simulated wind speeds at PS6 were $\leq$2 m s$^{-1}$, between 2 and 5 m s$^{-1}$ and $>5$ m s$^{-1}$ for 38%, 30% and 32% of the time, respectively.

Correlation patterns obtained for wind speeds $\leq$2 m s$^{-1}$ indicated a narrow impact radius (<8 km) and correlations were about 0.28 (significant) at all levels. This behavior indicates that PM$_{2.5}$ was distributed almost uniformly from the emission level to the breathing level under this wind condition (Figure 9). For wind speeds between 2 and 5 m s$^{-1}$, correlations were higher at the emission level (113–219 m) than at subsequently lower levels. This fact indicates dilution of the polluted air that led to lower PM$_{2.5}$ concentrations at the breathing level than at the emission level. In this wind speed range, the radius of impact was 8–10 km. Like for wind speeds $<$2 m s$^{-1}$, the correlation peaks indicated the highest impact of the point source emissions on the 24 h–average PM$_{2.5}$ concentrations correlated up to 0.452 (significant) at the emission level and marginally at the breathing level (up to 0.125, significant). This finding indicates a strong dilution of the polluted air. The correlation peaked at 4 km downwind.

Temperature inversions influence the dispersion of pollutants. We refer to an emission being below the inversion when the bottom of any inversion aloft is less than 50 m above the highest emission level. We considered an emission as being above an inversion layer when the top of the inversion layer is below the lowest emission level. We refer to an emission as going into an inversion layer when the lowest and highest emission levels fall into the inversion layer. In this study, non–inversion condition refers to conditions when the highest emission level is at least 300 m below the bottom of any inversion aloft. Theoretically, point sources contribute to PM$_{2.5}$ concentration at breathing level at lowest to highest magnitude when the emission level is above, in–between and below inversion layers, respectively.

During November to February, WRF/Chem simulated emissions to go into, above, and below the inversion 64%, 18%, and 10% of the time, respectively, and “no inversion conditions” occurred 8% of the time. This means the “between–inversion” conditions dominated the correlation pattern in November to February (Figures 8 and 10a). Under “below–inversion” conditions,
at breathing level, correlations between 1 h–differences and the point source emissions were higher than under the other conditions, and the impact radius extended 10–12 km (Figure 10b). Under “below inversion” condition, upward transport of PM$_{2.5}$ was limited which yielded more concentrated polluted air reaching the breathing level than under all other inversion conditions. When the emission level was above the inversion layer, correlations at breathing level (up to 0.157, significant) were much smaller than under the “between–inversion” (up to 0.295, significant) and “below–inversion” conditions (up to 0.416, significant); the correlation peak shifted to 4–6 km downwind of the point source and the impact radius extended to 14–16 km (Figure 10c). Emission into layers above the inversion allowed PM$_{2.5}$ to be transported far downwind and the pollutants had to be mixed down into the inversion to reach the ground. When no inversion existed, mixing strongly diluted the polluted air leading to low and non–significant correlations at breathing level (Figure 10d). On such days, no exceedance occurred in the nonattainment area.

![Figure 8](image1.png)

**Figure 8.** Correlations of emission rates with the PM$_{2.5}$ difference (REF–NPE) in downwind grid cells at subsequently lower levels from the uppermost level that emissions reached due to their buoyancy, to the breathing level (0–8 m) determined for November to February for various point–sources. Open circles indicate the relative position of point–sources around the point source of interest. Closed red circles indicate locations with significant correlations at the 95% confidence level.

![Figure 9](image2.png)

**Figure 9.** Like Figure 8, but for the correlations of emission rates at PS6 with the PM$_{2.5}$ difference (REF–NPE) in downwind grid cells in subsequently lower layers from the uppermost level that emissions reach due to their buoyancy (113–219 m), to the breathing level (0–8 m) as obtained for various wind–speeds. Behavior of other point–sources is similar.
4. Conclusions

The impact of point source emissions on the PM$_{2.5}$ concentrations at breathing level in the Fairbanks nonattainment area was investigated for one cold season using WRF/Chem simulations alternatively performed with (REF) and without (NPE) consideration of point source emissions. The statistical analysis of the simulations showed that point source emissions were minor contributors to PM$_{2.5}$ exceedances in the nonattainment area. Point source emissions are the best known emissions as they are strongly regulated and verified. Given the small absolute differences in PM$_{2.5}$ concentrations at breathing level found between REF and NPE, we have to conclude that even with higher uncertainty in the other emission sectors than the point source sector, point source emissions are not the main cause for the exceedances. In the nonattainment area, the daily maximum 24 h–average PM$_{2.5}$ concentrations obtained by REF and NPE differed about 1.3 $\mu$g m$^{-3}$ on average over November to February, and the highest maximum 24 h–average PM$_{2.5}$ concentration of REF barely exceeded that of NPE by 3 $\mu$g m$^{-3}$. However, during November to February the highest difference in 24 h–average concentrations averaged over the nonattainment area was 4.5 $\mu$g m$^{-3}$ (November 13). The highest difference of 24 h–average PM$_{2.5}$ concentrations was 18 $\mu$g m$^{-3}$ at PS6 (January 27). This means that, on average, the point source emissions did not affect where the maxima of PM$_{2.5}$ concentrations occurred in the nonattainment area except around PS6.

The locations where PM$_{2.5}$ exceeded the NAAQS occurred at the same locations in the nonattainment area in both simulations except for those exceedances at PS6 and its adjacent grid cell that only occurred in REF. Five out of 39 exceedance days predicted by REF were avoided in NPE and the highest REF–NPE 24 h–difference on these avoided exceedance days was 5 $\mu$g m$^{-3}$. This value is only slightly higher than the highest 24 h–difference averaged over the nonattainment area. Out of all point sources in the nonattainment area, PS6 contributed the highest to the PM$_{2.5}$ concentrations at breathing level as it had the highest PM$_{2.5}$ emission and contributed to the exceedances in the grid cell holding it and in its adjacent grid cells 8 (0) times in REF (NPE).

In general, wind speed, temperature and mixing height were the main meteorological factors driving the PM$_{2.5}$ concentrations. Temperature strongly affected stability. Thus, these meteorological factors determined whether or not PM$_{2.5}$ was transported out of or accumulated in the nonattainment area. Typically PM$_{2.5}$ concentrations were high under calm wind, low temperature and shallow mixing height situations. All point sources had their highest impact on the PM$_{2.5}$ concentration at breathing level in the grid cells they fall into. The impact radius at breathing level was usually 10–12 km, but could reach up to 16 km downwind depending on the height of the emission levels, magnitude of wind speed and the presence of an inversion above the layer the point source emitted into.

The analysis showed that in the Fairbanks nonattainment area except at PS6 and its adjacent grid cells, the 24 h–average PM$_{2.5}$ concentrations depended mainly on non–point source emissions and the meteorological conditions, and were least sensitive to point source emissions. At PS6 and its adjacent grid cells, however, the 24 h–average PM$_{2.5}$ concentrations were sensitive to emissions from both the non–point source and point source sector as well as to meteorological conditions.

Based on the low average reduction (1.3 $\mu$g m$^{-3}$) and the low number of exceedance days avoided (5), one has to conclude that emissions from non–point sources are the main contributors to the PM$_{2.5}$ exceedances in the nonattainment area. The differences between the REF and NPE concentrations (up to 5 $\mu$g m$^{-3}$) on the
exceedance days that were avoided in NPE are small. They suggest that only a slight increase in non-point source emissions (e.g. from traffic, residential heating) is sufficient to exceed the NAAQS. Thus, tightening the filter requirements for point sources may only exclude some areas from experiencing an exceedance or avoid slight exceedances, if at all.

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