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Investigations on meteorological conditions for elevated PM_{2.5} in Fairbanks, Alaska

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ABSTRACT

The relationships between meteorological conditions (temperature, wind-speed and direction, relative humidity, surface-inversion depth and strength, and stability) and PM_{2.5} concentrations in Fairbanks, Alaska were investigated using ten years of observational data. The results show that during wintertime (November through February) PM_{2.5} concentrations exceeding the 24 h National Air Quality Standard (35 µg/m³) occurred under calm wind, extremely low temperature (\leq 20 °C) and moisture (water-vapor pressure <2 hPa) multiday surface-inversion conditions that trap the pollutants in the breathing level and inhibit transport of polluted air out of Fairbanks. PM_{2.5} concentrations tend to be higher under stable than other conditions, but are not sensitive to the degree of stability. The presence of a surface inversion and calm wind are necessary, but in combination with low temperatures and humidity, the conditions are sufficient for high PM_{2.5} concentrations. The low temperatures are required because they lead to increased emission rates from domestic heating and power production. During multiday inversions with temperatures above -20 °C, high relative humidity (>75%) partly caused by water-vapor emission reduces PM_{2.5} concentrations.

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1. Introduction

Concentrations of particulate matter of less than or equal to 2.5 μm in diameter (PM_{2.5}) are of concern for air-quality regulations since PM_{2.5} can affect human health (e.g. Godish, 2004; Dominici et al., 2006; Miller et al., 2007; Delfino et al., 2009). Adverse health effects of PM can be associated with both long-term and short-term exposure (e.g. Schwartz et al., 1997; Bernard et al., 2001; Kappos et al., 2004). To decrease health risks, in the United States of America, the 24 h National Ambient Air Quality Standard (NAAQS) for PM_{2.5} was tightened to 35 $\mu g/m^3$ in 2006. Communities, for which the last three years of PM_{2.5}-monitoring prior to 2006 showed violation of this new standard, were assigned PM_{2.5}-nonattainment areas. These communities have to develop strategies to

get into and remain in compliance. Such planning requires understanding of the meteorological and emission situations that lead to high $PM_{2.5}$ concentrations and exceedances.

In Fairbanks, Alaska, PM_{2.5} concentrations have exceeded frequently the new NAAQS in all winters (November to February) since the onset of monitoring in 1999 (Fig. 1). During winter, Fairbanks' high latitude location (64.838 N, 147.716 W) leads to a negative radiation balance as the outgoing radiation exceeds the incoming radiation. This fact and being enclosed by hills to three sides and being located about 800 km land-inwards lead to frequent winter inversions that are among the strongest anywhere and persist much longer than in midlatitudes (Wendler and Nicpon, 1975; Bourne et al., 2009). Daytime and nighttime surface inversions occur on about 82% of the days in December and January, and on about 68% of the days for November, and February to April during the episode 1957–2008 (Bourne et al., 2009).

During winter, PM_{2.5} exists in abundance from traffic and other combustion processes. The extremely cold weather and

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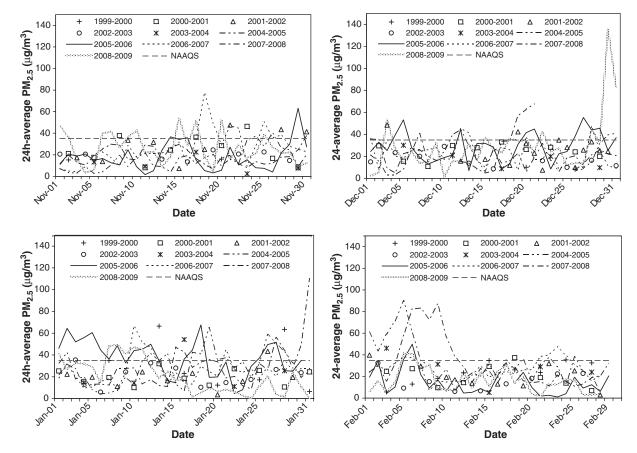


Fig. 1. Temporal evolution of 24 h-average PM_{2.5} concentrations for 1999 to 2009 for (upper left to lower right) November, December, January, and February.

long dark nights lead to huge fuel consumption for heating and power supply. Both fuel consumption for heating and power supply increase with decreasing temperature (e.g. Hart and de Dear, 2004). The power generation of the UAF power plant, for instance, was about 5%, 4% and 4% lower in November, December, and February 2008, respectively, than in January 2008 (Ward, 2009, pers. comm.). Cold start particle emissions from vehicles increase by an order of magnitude as temperature drops from 23 °C to -20 °C (e.g. Weilenmann et al., 2009). Furthermore, people are more likely to use their car or to idle their car as temperature decreases. Thus, during winter, traffic may be the cause for roughly 30% of the PM_{2.5} in downtown Fairbanks (Johnson et al., 2009).

Several statistical and modeling studies examined the relationship between $PM_{2.5}$ and meteorological conditions (Triantafyllou et al., 2002; Elminir, 2005; Wise and Comrie, 2005; Liao et al., 2006; Unger et al., 2006; Dawson et al., 2007). They illustrated the difficulty in identifying causal relationships between specific meteorological parameters and measured $PM_{2.5}$ concentration when the meteorological variables correlate strongly with one another. $PM_{2.5}$ concentrations were found to depend strongly on wind-speed, wind direction, temperature, humidity, mixing height, precipitation and cloud cover (Elminir, 2005; Wise and Comrie, 2005; Dawson et al., 2007). Stable conditions associated with temperature inversions, strongly correlate with high pollutant concentration since inversions hinder the upward transport

of polluted near-surface air (Chow et al., 1995; Triantafyllou et al., 2002).

These studies mainly focused on low and mid-latitude regions with quite different meteorological conditions than Fairbanks. The goal of our study is to examine the relationship between Fairbanks' wintertime inversions, high PM_{2.5} concentrations and meteorological conditions.

2. Data collection and analysis methods

PM_{2.5} concentrations have been monitored in downtown Fairbanks since 1972. The monitoring site (Fig. 2) is located on the roof of a building in the middle of the central business district. This site is equipped with two Thermo Electron Partisol 2000 samplers and a single Met-One Beta SASS Speciation Monitor running on a 1-in-3-day sampling schedule, and a single Met-One Beta Attenuation Monitor (BAM 1020) running on a real-time schedule. The inlets of all samplers are approximately 6 m above the ground (DEC, 2009). We used the SASS data since 1999, and the BAM 1020 available since June 2004 through 2009.

Since the NAAQS looks at the 24 h-average $PM_{2.5}$ concentrations, we calculated 24 h daily average concentrations using the BAM 1020 data for all days with complete datasets from June 2004 to February 2009. To examine whether the data from 1999 to 2004 provide valuable additional information for our study, we prepared three time-series from the



Fig. 2. View of the $PM_{2.5}$ monitoring site in downtown Fairbanks. Source: DEC (2009)

2004 to 2009 24 h-average $PM_{2.5}$ concentrations. These time-series start on June 1, 2, and 3, 2004 and only consider every third day's $PM_{2.5}$ data. Comparison of these time-series with the full 2004–2009 time-series shows that all three 1-in-3-day time-series of this episode well represent the relationships between $PM_{2.5}$ and meteorology. Therefore, we included the 1999–2004 $PM_{2.5}$ -monitoring data in our analysis.

Radiosonde data of temperature, dewpoint temperature, potential temperature, wind-speed and wind direction are available from daily soundings at the Fairbanks International Airport (FIA) at 0000 and 1200 UTC (1500 and 0300 Alaska Standard Time (AST)), respectively.

Inversion layer is defined as the layer wherein temperature increases with height and is associated with a positive temperature gradient. An algorithm was developed applying the technique introduced by Kahl (1990) to identify the first inversion layer (if any) from the sounding data at levels below 700 hPa. Surface inversions included those inversion layers starting at the ground or those starting at less than 100 m above ground. Inversion layers starting 100 m above the ground were accounted as elevated inversion layers.

At night (0300 AST) inversions are common phenomena due to the negative radiation balance (Wendler and Nicpon, 1975; Bourne et al., 2009). For the period considered here, 94% of the winter days had nighttime inversions with bases within 500 m height above the surface. Hereof, 93% had their base within the first 100 m above ground. In our study, we considered a day to be influenced by an inversion event when an inversion layer existed at 1500 AST. If several consecutive days had an inversion at 1500 AST as well as at 0300 AST, we would call this event a multiday inversion. In the analysis, we distinguished between single-day inversions lasting a day and multiday inversions lasting two or more days.

We defined the inversion depth (Δz) as the depth between the bottom and the top of the first inversion layer. Temperature gradients between the inversion base and the next 100 m, 200 m, 300 m, 400 m, 500 m, 600 m, 800 m, and the top layer were determined to represent the inversion strength. The temperatures for these critical levels were interpolated by the values of closest lower and upper levels recorded by the radiosonde. This method of looking at the temperature gradient of the inversion is called STRXs here-

after. Here X denotes the distance over which the gradient was determined (e.g. for STR100 the gradient is determined for 100 m). If an inversion ended below 200 m, for example, then just STR100 would be defined and no temperature gradients would be determined for the levels 200 m and above.

Potential temperature increases with height under stable conditions (positive gradient) whereas it decreases with height under unstable conditions (negative gradient). To examine the role of atmospheric stability on $PM_{2.5}$ concentrations we analyzed the vertical gradient of potential temperature. Similarly, to the method described above, the potential temperature gradient was determined from the ground to 100 m, 200 m, 300 m, 400 m, 500 m, 600 m and 800 m. These potential temperature gradients are denoted as PGXs. Again X denotes the distance over which the gradient was determined.

Additionally, our analysis considers surface observations at FIA of 2 m temperature (T) and dewpoint temperature (T_d), 10 m wind-speed (v) and direction (dir), sea-level pressure (SLP), and reported fog/mist conditions. To evaluate the atmospheric moisture, the water-vapor partial pressure (E_v) and relative humidity (RH) were determined by applying the Magnus–Tetens approximation (Lawrence, 2005).

Since the 24 h-average $PM_{2.5}$ concentrations were determined by averaging the hourly $PM_{2.5}$ data from 0000 AST to 0000 AST of the next day, we calculated daily averages for all meteorological data to investigate the correlation between meteorological conditions and $PM_{2.5}$ exceedances. The relative importance of thermal and mechanical turbulence in the local near-surface atmosphere was evaluated through the gradient Richardson number (Ri) which was calculated in accord with Rohli and Vega (2007)

$$Ri = \frac{g}{\bar{\theta}} \frac{\Delta \theta / \Delta z}{\left(\Delta u / \Delta z\right)^2}.$$
 (1)

Here Δ represents the difference of the potential temperature (θ) , wind-speed (u) and geometric height (z) at the first two sounding levels with valid data, g is the gravitational acceleration $(9.81~{\rm ms}^{-2})$ and θ is the mean potential temperature between these two levels. The calculation of the daily average Ri utilizes the radiosonde data of 0300 and 1500 AST.

In accord with our definition of "inversion days", we determined the correlation between the 24 h-average PM_{2.5} concentration and the inversion heights, inversion strengths, potential temperature gradient and *Ri* at 1500 AST for 1999 to 2009. The correlation between 24 h-average PM_{2.5} concentrations and daily average meteorological quantities (e.g., RH, e_v, T, v, dir, SLP, *Ri*) was calculated. For confidence in the correlations, we tested them for statistical significance at the 95% confidence level using a Student's t-test. In this study, the term "significant" will only be used if the correlation according to this test is significant at the 95% or higher confidence level.

We examined the individual influence of the various meteorological parameters on the 24 h-average PM_{2.5} concentrations and their importance with respect to the other parameters using a multi-regression method (Storch and

Zwiers, 1999). To evaluate the effect of inversion conditions on the exceedances, we calculated the ratio between the frequencies of inversions to the frequency of exceedances associated with inversion conditions. Analogously, we examined the effect of temperature below a certain threshold on exceedances.

3. Result and discussion

3.1. PM_{2.5} concentrations

Temporal variation of 24 h-average $PM_{2.5}$ concentrations in Fairbanks during the winters of 1999 to 2009 shows numerous exceedances of the NAAQS (Fig. 1). There were 128 exceedances during the winters of 2004 to 2009, and 17 (over 160 observed days) during the winters of 1999 to 2004. The variation of 24 h-average $PM_{2.5}$ concentration among these months is non-uniform from 2004 to 2009 (Fig. 1). In general, the number of exceedance days is highest in January followed by December. The highest and second highest exceedances occurred on December 29 and January 30, 2008 with over $135 \,\mu\text{g/m}^3$ and $110 \,\mu\text{g/m}^3$, respectively. On these days, the atmosphere was remarkably stable, extremely cold, and dry with temperatures of $-38 \,^{\circ}\text{C}$ and $-32 \,^{\circ}\text{C}$, and relative humidity of 57% and 71%, and there was no wind.

3.2. Inversions

In Fairbanks, a closed snow-cover exists continuously from mid October through early May (Shulski and Wendler, 2007). The high albedo of the snow-covered surface reflects incident shortwave radiations. The temperature-albedo feedback results in cooler temperature close to the ground than in the air layers aloft and therefore may contribute to the formation of surface inversions. The frequency of surface inversions during November, December, January and February in the years 1999–2009 was 65%, 82%, 80% and 72%, respectively. These frequencies are slightly higher than the average frequencies of 60%, 76%, 77%, and 60% reported by Bourne et al. (2009) for the winters of 1957-2008. January 2009 had the highest frequency with surface inversions on 94% of the days. The variance of frequency is highest in November, and lowest in December followed by January and February. For the winters considered in this study, 96% of the surface inversions have their base at the ground surface.

In the winters of 2004–2009, all 128 exceedances were associated with surface inversions. In the winters of 1999–2004, 17 out of 18 detected exceedances were associated with surface inversions. Of the 128 exceedances in the winters of 2004–2009 18%, 25%, 35% and 22% occurred in November, December, January and February, respectively (Fig. 3). This means that January has the highest exceedance occurrence and highest frequency of surface-inversion events.

No exceedances occurred when elevated inversions existed, even with those based within 100–200 m, which make up 15% of the total 153 elevated inversion events during the winters of 2004–2009. Based on these findings, one has to conclude that inversion layers having their base within the first 100 m above ground play a major role for the occurrence of PM_{2.5} exceedances. Therefore, in the following, all discussion focuses on surface inversions.

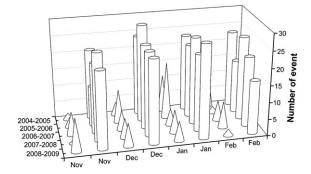


Fig. 3. Evolution of PM_{2.5} exceedance (cone) and surface-based inversion (cylinder) from November through February in 2004–2009.

Among the 440 surface inversions during the winters of 2004 to 2009, 18 were single-day inversions out of which only two days had PM_{2.5} exceedances. During these winters, 86% of the PM_{2.5} exceedances occurred under multidayinversion conditions (Fig. 4). For the winters of 1999-2003, PM_{2.5} data were only available every three days. Out of the total 473 surface inversions during these winters, 28% fall on a day with PM_{2.5} data. Out of these 13% were associated with PM_{2.5} exceedances that occurred under multiday-inversion conditions. All these results suggest that formation of a PM_{2.5} exceedance is an accumulated effect of continuous pollutant trapping over several days in the inversion above Fairbanks and the poor dispersion associated with multiday inversions. However, the number of PM_{2.5} exceedances is uncorrelated with the duration of multiday inversions (Fig. 4). A large number of single and multiday inversions had no exceedance and the temporal evolution of surface-inversion events differs from that of PM_{2.5} exceedance events. Therefore, one has to conclude that the presence of a surface inversion is not the only factor leading to PM_{2.5} exceedance.

Surface-inversion depth varies from less than 100 m up to more than 2000 m. However, although the PM_{2.5} exceedances occur at all scales of inversion depths, they are more likely to occur for inversion depths greater than 300 m. During the winters of 1999 to 2009, 41% of the surface inversions had depths less than 300 m, but they were just associated with 18.4% of total exceedances, resulting in a ratio of occurrencefrequency of 0.45. Surface-inversion layers having depths greater than 300 m had an occurrence-frequency ratio of 1.39 indicating that they were more likely associated with exceedances. The highest occurrence–frequency ratio (1.49) was found for inversion layers having depths in the range 300-350 m. They made up 7.9% of the total surface-inversion events and were associated with 11.8% exceedances. Nevertheless, inversion depth does not strongly influence the 24 haverage PM_{2.5} concentration as indicated by the generally low (0.272), but significant correlation between these quantities. Inversion layers having depths in the range of 0 to 200 m have weak and insignificant correlation with the PM_{2.5} concentration and make up 26% of the total inversion events. Correlation between inversion depth and 24 h-average PM_{2.5} concentration increases and becomes significant as the depth range increases. The highest correlation (0.382, significant) was found for the depth range 0-350 m that made up 49% of the total inversion events. At greater depth

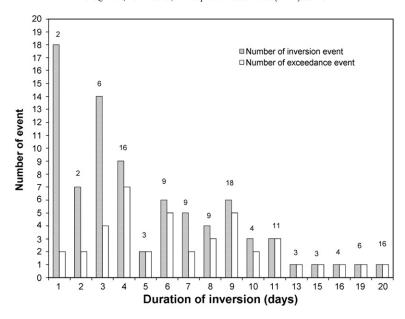


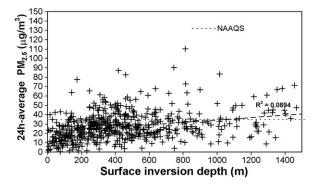
Fig. 4. Frequency of multiday-inversion occurrence and its associated PM_{2.5} exceedances. The grey shaded bar shows the occurrence–frequency of inversions with a given duration; the white bar represents the number of exceedance events that each duration is associated with; the number above each bar represents the total number of days having exceedance. For example, single-day inversions occurred 18 times and two times, they coincided with exceedances.

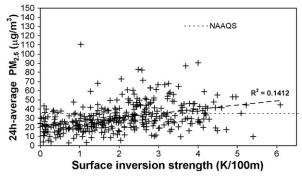
ranges (e.g. 0-400~m, 0-500~m and more), correlation decreases, but is still significant. This means that inversion layers having depths greater than 350 m do not influence the $PM_{2.5}$ concentration as effectively as those layers having depths less than this threshold.

Because of the interesting behavior of the highest correlation at 350 m, we included this height into the investigation of inversion strength as STR350. From a theoretical point of view, one has to expect that the 24 h-average PM_{2.5} concentrations will increase if the inversion strength increases. The strongest inversions usually occurred within the first 100 m above ground when for STR100 the frequency of inversions with strength > 8 K/100 m is 10.7% compared to less than 1% when strength is determined for levels 0-200 m or higher. PM_{2.5} exceedances may occur at all magnitudes of inversion strength, but they are more likely to occur when inversion strength exceeds 2 K/100 m. Inversions with strength less than 2 K/100 m made up for 39% of all cases, but they were only associated with 18% of all exceedances, resulting in a ratio of occurrence-frequency of 0.45. Inversions with strength greater than 2 K/100 m have occurrencefrequency ratios greater than 1. The ratio increases with inversion strength. The correlation behavior of inversion strength with 24 h-average PM_{2.5} concentration is similar to that of inversion depth with 24 h-average PM_{2.5} concentration. Correlations are 0.221, 0.293, and 0.325 for STR100, STR200, and STR300, respectively and reach 0.376 as the highest correlation for STR350 (all significant). Above 350 m, correlations decrease to 0.373, 0.369, and 0.230 for STR400, STR600 and STR800, respectively (all significant). Overallcorrelation of inversion strength with 24 h-average PM_{2.5} concentration is low (0.296), but significant. In general, neither inversion depth nor inversion strength correlates strongly with the 24 h-average PM_{2.5} concentration. Instead, PM_{2.5} exceedances occurred at different ranges of inversion strength and depth (Fig. 5).

3.3. Stability

In the Fairbanks' winter surface-inversion layers, the atmosphere is extremely stable and the potential gradient is largely positive (16 K/100 m at the highest). We included PG350 in the analysis of potential temperature-gradient impacts, and investigated the correlation of PGX with the 24 h-average PM_{2.5} concentrations on inversion days and on all winter days. Here again X denotes the distance over which the potential temperature gradient (PGX) or inversion strength (STRX) was determined (e.g. X equals 100 m, 200 m, etc.). PGX shows different correlation behaviors with 24 haverage PM_{2.5} concentrations on inversion days and on all winter days (Fig. 5). Under inversion conditions, PGX-PM_{2.5}behavior is similar to STRX-PM_{2.5}-behavior. The highest correlation between PM_{2.5} and potential temperature exists at 350 m (0.379, significant) with gradual decrease towards higher and lower levels. PM_{2.5} exceedances were found for PGX greater than 1 K/100 m which is the typical condition observed during surface-inversion events. The ratio of frequency of exceedances to PGX>3 K/100 m is higher than 1. These ratios indicate that more exceedances occurred when PGX exceeded this threshold. However, the highest potential temperature gradients as obtained with PG100 were not necessarily associated with the highest 24 h-average PM_{2.5} concentrations and exceedances. For all winter days, PGX correlates much stronger with the 24 h-average PM_{2.5} concentration than for inversion days and correlations increase when the potential temperature gradient was calculated over a thicker than thinner layer. Correlations were 0.418, 0.515, 0.561, 0.576, 0.586 and 0.595 for PG100, PG200, PG300,





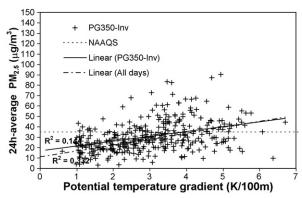


Fig. 5. Correlation between the inversion depth, inversion strength, or potential temperature (from top to bottom) and the 24 h-average $PM_{2.5}$ concentration.

PG350, PG400, and PG500, respectively (all significant). For PG600 and beyond, correlations decreased. The lower correlation on inversion days suggests that the degree of stability does not effectively influence the magnitude of the 24 h-average PM_{2.5} concentrations during an inversion event. Obviously, like inversion strength, the degree of stability plays a role, but it does not govern the PM_{2.5} exceedances alone.

3.4. Wind-speed and direction

Various studies showed that wind-speed plays an important role for dispersion of pollutants and thus 24 h-average PM_{2.5} concentrations (Elminir, 2005; Dawson et al., 2007). Wind direction may lead to PM_{2.5}-advection from upwind sources (Chu et al., 2009). During wintertime, calm winds (average wind-speed < 0.5 m/s) dominate in the Fairbanks area (Shulski and Wendler, 2007). During the winters of 1999 to 2009, wind was calm on 68.4% of the days. During these winters of 92.6% of the PM_{2.5} exceedances occurred under inversion, calm-wind conditions. Under inversion conditions, wind-speed correlates higher with the 24 h-average PM_{2.5} concentration than under non-inversion conditions (-0.347and -0.213, respectively), while for the entire winters the correlation is -0.330. All these correlations are significant. Low 24 h-average PM_{2.5} concentrations may nevertheless occur on days with calm-wind conditions. Thus, one has to conclude that calm wind is a critical pre-requisite for high 24 h-average PM_{2,5} concentrations, but it is not the key factor.

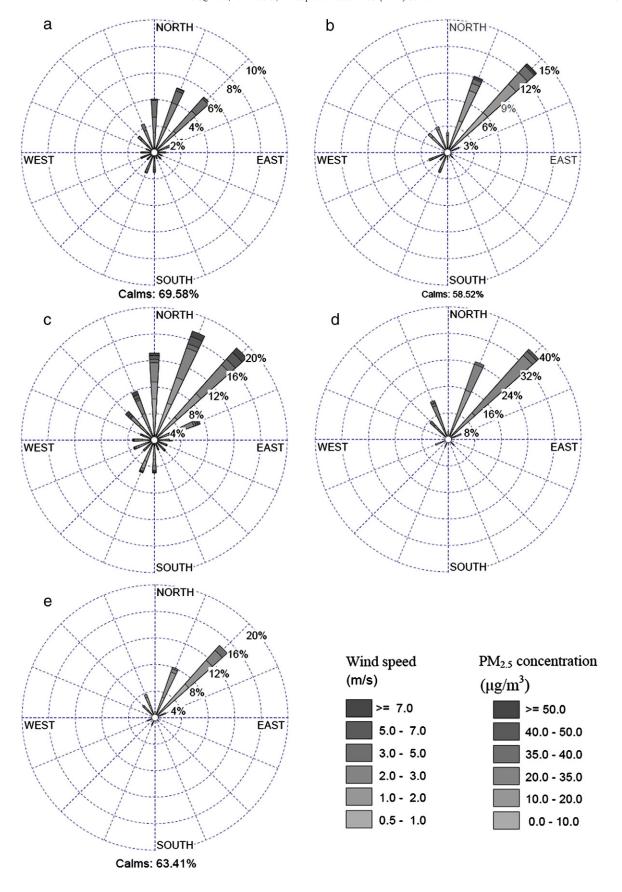
During the 1999–2009 winters, winds from North–Northeast dominated in Fairbanks. This wind direction also dominated during inversion events (Fig. 6). None of the wind directions favored accumulation of high 24 h-average PM_{2.5} concentrations. Most of the high 24 h-average PM_{2.5} concentrations occurred under calm-wind condition under which wind direction cannot be identified clearly. Thus, we have to conclude that none of the point sources (e.g. power plants, industrial facilities that emit into higher atmospheric layers than the breathing level) are the major cause for the 24 h-average PM_{2.5} concentrations measured at the Fairbanks downtown site.

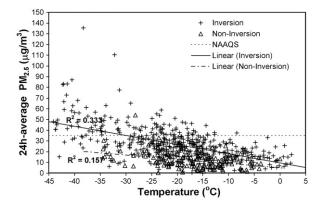
3.5. Temperature

In Fairbanks, monthly mean temperature is $-16\,^{\circ}\text{C}$, $-21\,^{\circ}\text{C}$, $-23\,^{\circ}\text{C}$ and $-20\,^{\circ}\text{C}$ in November, December, January and February, respectively; during these months temperatures can range from -51 to $10\,^{\circ}\text{C}$ (Shulski and Wendler, 2007). The winters of 1999 to 2009 fall into this typical range. In January, inter-annual variability shows frequently extreme temperature changes where temperature drops to below $-40\,^{\circ}\text{C}$ within 60 h at the longest and then goes up to values above freezing. Such rapid temperature changes occurred in 2005, 2008, and 2009. For the episode considered in our study, the lowest daily average temperature was $-47\,^{\circ}\text{C}$ in December 1999, which is 7 K higher than the lowest temperature observed and is the 8th lowest temperature since onset of record in 1930.

In the winters of 1999 to 2009, $PM_{2.5}$ exceedances occurred more likely at temperatures below $-15\,^{\circ}\text{C}$ and were intensively associated with temperatures below $-20\,^{\circ}\text{C}$ (Fig. 7). Higher 24 h-average $PM_{2.5}$ concentrations were associated with lower temperatures. No obvious relationship between temperature and inversion condition exists as inversions occur at various temperatures. During these winters 4.4% of the exceedances occurred at temperature above $-15\,^{\circ}\text{C}$. Such temperature conditions occurred on 30.5% of the days. Thus, we obtain a ratio of exceedance of $PM_{2.5}$ to temperature above this threshold of 0.14. The ratio for temperature in the range of $-20\,^{\circ}\text{C}$ to $-15\,^{\circ}\text{C}$ was 0.49 (10.5% vs.

Fig. 6. Wind-rose profile (a) on an hourly basis, (b) on daily average and (e) under inversion events during winters of 2004–2009. The PM_{2.5}-concentration rose for (c) hourly and (d) daily averages represents the relation between PM_{2.5} exceedances and wind direction on days having wind-speeds higher than 0.5 m/s, the threshold that allows the average wind direction to become important.





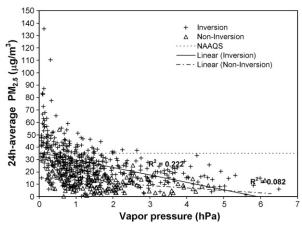


Fig. 7. Correlation of temperature (top) and partial water-vapor pressure (bottom) with $PM_{2.5}$ concentration with the trend line being superimposed.

21.3%) whereas the ratio for temperatures between $-35\,^{\circ}\mathrm{C}$ and $-20\,^{\circ}\mathrm{C}$ was 1.41 (54.4% vs. 38.5%). Temperatures $\leq 35\,^{\circ}\mathrm{C}$ have the highest ratio with 3.20 (30.7% vs. 9.6%). Correlations between temperature and 24 h-average PM_{2.5} concentration are -0.577, -0.396 and -0.568 (all significant) under inversion, non-inversion and for all winter days, respectively. This behavior of ratios demonstrates the strong influence of temperature on the PM_{2.5} concentration. The ratios explain why the greatest number of exceedance days and the highest PM_{2.5} concentrations typically occur in late December and in January when temperatures reach their lowest values during wintertime. Elminir (2005) and Dawson et al. (2007) found similar correlation and behavior for Cairo, Egypt and the Eastern US, respectively.

3.6. Partial water-vapor pressure and relative humidity

Both vapor partial pressure (e_v) and relative humidity (RH) represent atmospheric moisture. In a moist atmosphere, aerosol particles take up water vapor, swell and may coagulate. This change in size and density increases their sedimentation velocity (e.g. Donateo et al., 2006). Therefore, PM_{2.5} concentrations are reduced when e_v and RH are high. In our study, 97% of observed exceedances coincide with e_v less than 2 hPa, out of which 84% of the exceedances occurred at

 $e_{\rm v}$ less than 1 hPa (Fig. 7). Water-vapor pressure and PM $_{2.5}$ 24 h-average concentration correlate moderately ($-\,0.438$), but significantly.

Several studies performed for the Apulia, Italy and Los Angeles, CA revealed that RH exceeding 70% would affect PM_{2.5} characteristics (e.g. MIE, 1994; Shen et al., 2002; Donateo et al., 2006). In our study, a threshold of RH 75% was found to affect the PM_{2.5} concentration. The ratio of the occurrence-frequency of PM_{2.5} exceedances over RH is greatly reduced for RH>75% (1.53 and 0.58 for RH≤75% and RH>75%, respectively). No exceedances occurred at RH>90%. The 75% threshold for RH well correlates with the e_v-threshold of 1 hPa that indicates atmospheric conditions as being "dry" in Fairbanks if RH≤75%. This RHthreshold also correlates well with the threshold of -20 °C that temperature has to be below for PM_{2.5} exceedances to be likely to occur. Around this temperature, the atmosphere is still super-saturated with respect to ice. As temperatures fall below -20 °C, ice crystals form efficiently and fall out. This process reduces the atmospheric moisture load that would otherwise have been favorable for high PM25 concentrations and thus exceedances. At temperature above -20 °C, the atmosphere may be super-saturated with respect to water. The particles swell and may achieve diameters greater than 2.5 µm, for which the PM_{2.5} concentration goes down. These phenomena indicate indirect effects of temperature on PM_{2.5} concentrations. Note that no relationship between inversion conditions and ev as well as RH was found.

3.7. Gradient Richard number, sea-level pressure and ice fog

No evident relationship between gradient Richardson number (Ri) and 24 h-average PM_{2.5} concentration exists. Within a wide range of Ri, 78% of the Ri are larger than 2. This fact indicates that the thermal buoyancy is stronger than the wind shear. Large positive Ris were found for non-inversion days due to the close to zero wind shear and calm-wind conditions that are common during winter in Fairbanks. The 24 h-average PM_{2.5} concentration and Ri insignificantly correlate (0.059), i.e. no relationship between Ri and exceedances occurred at all ranges of Ri>1.

Sea-level pressure marginally (0.184, significant) correlates with the 24 h-average PM_{2.5} concentrations, but typically correlates slightly better on inversion days (0.222, significant) than non-inversion days (0.051, insignificant). No typical range of SLP on inversion days or exceedance events exists. Therefore, we conclude that SLP marginally affects PM_{2.5} concentration.

Theoretically, formation of ice fog weakens the strength of surface-based inversion, and hence, may help to reduce the PM_{2.5} concentrations. However, in Fairbanks, normally ice fog does not achieve a sufficient thickness to destroy the inversion in the lowest 16 m above ground where ice fog is formed (Wendler and Nicpon, 1975). In our study, we found that the PM_{2.5} exceedances occurred on days with and without ice fog suggesting that ice fog has no effect on exceedances in Fairbanks. This behavior well agrees with the finding discussed above that the inversion strength does not effectively govern the PM_{2.5} concentration.

3.8. Combined effects

Analysis of the correlation coefficients between the various meteorological quantities and the 24 h-average concentration suggests excluding wind direction, Ri and SLP from the multi-linear regression analysis due to their low correlations. Both RH and e_v represent atmospheric moisture for which only one of them should be considered to avoid redundant information. We selected e_v to avoid ambiguities related to saturation over ice and water that would require to break the multi-linear regression into two equations in relation to RH≤75% and RH>75%. Out of all X for PGX and STRX, we chose 350 m as here the strongest correlations were found. PG350 is included in the analysis as it can represent both the inversion strength and atmospheric stable conditions. We excluded STR350 as it provides slightly redundant information to PG350. All other parameters were included in the multi-regression analysis. We determined the multilinear regression equations for inversion days only (denoted INV) and the entire winters (denoted ALL). For INV, we investigated the relationship of 24 h-average PM_{2.5} concentration with the above-identified meteorological parameters $(\frac{\partial \theta}{\partial z}|_{0=350m}, \Delta z, v, T, \text{ and } e_v)$ for surface-inversion events only. For ALL, we determined the relationships between the 24 haverage PM_{2.5} concentrations and these quantities except inversion depth. Note that the multi-linear regression calculation requires data availability for all quantities at the time of calculation. This requirement reduced the number of days considered in the calculation of multi-regression coefficients from 601 to 458 for ALL and INV, respectively. The regression analysis provided

$$PM_{2.5} = -0.538 + 3.009 \frac{\partial \theta}{\partial z} \Big|_{0-350 \text{ m}} + 0.001\Delta z - 1.728v$$
$$-0.843T + 1.85232e_v \qquad (2)$$

for inversion days only (INV) and

$$PM_{2.5} = -6.181 + 4.081 \frac{\partial \theta}{\partial z} \Big|_{0-350m} - 0.936v - 0.888T$$
 (3)
+ 2.451e_v

for all winters (ALL) with R^2 of 0.435 and 0.509, respectively. Reasons for the relatively low values of R^2 can be unidentified parameters that affect the 24 h-average $PM_{2.5}$ concentrations, the limited data availability, measurement errors, the distance between the radiosonde and $PM_{2.5}$ -measurement sites (about 8 km), the impact of local $PM_{2.5}$ emission sources, or combination of those. The impact of low data availability is obvious, as adding more data increases the R^2 -value (all winters vs. inversion days only).

Nevertheless, the regression coefficients of Eqs. (2) and (3) still permit for evaluating the importance of the various quantities for the $PM_{2.5}$ concentration. The standardized coefficient (SC) indicates the role of each quantity for the 24 h-average $PM_{2.5}$ concentration. Positive (negative) SC implies a positive (negative) correlation of the quantity with the 24 h-average $PM_{2.5}$ concentration. The importance of a quantity was judged based on the magnitude of its SC that indicates the ratio of deviation of its value to the deviation of

24 h-average PM_{2.5} concentration. Parameters having large SC are more important to the 24 h-average PM_{2.5} concentration than those with low SC (Schroeder et al., 1986).

In further multi-linear regression tests, quantities were removed and included alternatively to examine their role and interaction effects with other quantities on PM_{2.5} concentrations. For INV, inversion depth has positive and the lowest SC (Table 1). This behavior well agrees with the finding that the 24 h-average PM_{2.5} concentration is not very sensitive to inversion depth. One has to conclude that out of the quantities examined here inversion depth is the least important for the 24 h-average PM_{2.5} concentration. For both INV and ALL, wind-speed is of second least importance to the 24 h-average PM_{2.5} concentration during winter. This finding may be misleading and results from the fact that on average over all winters considered, low wind-speeds dominate in Fairbanks and high as well as low 24 h-average PM_{2.5} concentration occurred at any wind-speed less than 1 m/s. However, investigation of individual cases shows that as wind-speed was high (>10 m/s), PM_{2.5} concentrations were low as the pollutants quickly leave the area. Exclusion of wind-speed from the analysis only slightly reduces R² (Table 1). However, the finding emphasizes that low windspeed is a mandatory condition for PM_{2.5} exceedances to occur. The third least important contributor is e_v. It has positive SC although it has negative correlation with the 24 haverage PM_{2.5} concentration. This phenomenon is due to the strong correlation between e_v and temperature. Removing temperature from the analyses results in e_v gaining negative SC, but a great decrease of R². This behavior indicates that temperature is more important than ev. Considering only $\frac{\partial \theta}{\partial z}|_{0-350m}$, Δz , v and T for inversion days only and $\frac{\partial \theta}{\partial z}|_{0-350m}$, v and T for all winters yields

$$PM_{2.5} = 5.966 + 3.052 \frac{\partial \theta}{\partial z} \Big|_{0-350 \text{ m}} + 0.001 \Delta z - 1.738 \nu \quad (4)$$

$$-0.645 \text{T}$$

$$PM_{2.5} = -2.566 + 4.189 \frac{\partial \theta}{\partial z} \Big|_{0-350 \,\mathrm{m}} -1.022 \nu - 0.616 \mathrm{T}$$
 (5)

with R^2 being reduced marginally to 0.430 and 0.501, respectively as compared to the full regression equations. This means that both the potential temperature gradient and temperature are the most important meteorological quantities that affect the $PM_{2.5}$ concentrations. Interestingly, their

Table 1 Standard coefficients of meteorological parameters in functional relations with $PM_{2.5}$ concentration. Blank cells indicate quantities that were excluded in the respective tests.

Case of analysis	$\frac{\partial \theta}{\partial z}\Big _{0-350\mathrm{m}}$	Δz	v	T	e _v	\mathbb{R}^2	Valid data
INV	0.280	0.019	-0.092	-0.609	0.157	0.435	458
	0.298	0.031	-	-0.632	0.158	0.428	
	0.316	0.072	-0.124	-	-0.355	0.369	
	0.284	0.022	-0.0922	-0.466	-	0.430	
ALL	0.418	-	-0.0582	-0.599	0.199	0.508	601
	0.431	-	-	-0.620	0.207	0.506	
	0.475	-	-0.102	-	-0.315	0.444	
	0.429	-	-0.064	-0.416	-	0.501	

roles in influencing the 24 h-average PM_{2.5} concentrations differ for inversion days as compared to all winter days. Under inversion conditions, temperature was more important than the potential temperature gradient, while for the entire winter their roles were almost equal. Thus, the findings of the multi-regression analysis further emphasize that the characteristics of surface inversions are not the key factors that determine the magnitude of PM_{2.5} exceedances. Instead, temperature is the determining factor for PM_{2.5} exceedances to occur, which in turn relates to the emission strength. Hart and de Dear (2004), for instance, reported an increase in emissions from heating with decreasing temperatures. Timmer and Lamb (2007) found a strong correlation (R>0.8) between natural gas consumption for heating and heating degree-days in the northern states of the US that on average experience colder winters than do the other states except Alaska. Weilenmann et al. (2009) reported that the cold-start emissions of passenger cars rise drastically at -20 °C as compared with those at -7 °C. Nam et al. (2010) found that regardless of vehicle model year, the emission of particulate matter doubles for every 20 °F-decrease of the ambient temperature.

Another potential reason for the relationship between temperature and $\rm PM_{2.5}$ concentration is the effect of temperature on the gas-to-particle conversion. As temperature decreases, the vapor pressure decreases accordingly and the gas-to-particle partitioning shifts towards the aerosol phase (Strader et al., 1999). A temperature decrease by 10 K leads to an increase of 20% to 150% in the secondary organic aerosol (SOA) concentrations (Sheehan and Bowman, 2001). These studies, however, were all carried out for mid-latitudes, and knowledge on the formation mechanisms of SOAs under extremely cold temperature and low solar radiation conditions like in Fairbanks during winter is still limited.

4. Conclusions

In Fairbanks, 24 h-average PM_{2.5} concentrations frequently exceeded the new NAAOS during the winters (November through February) of 1999 to 2009. During winter, surface inversions existed 75% of the time. The results of our study suggest that inversion layers with bases within the first 100 m above ground enhance the likelihood for PM_{2.5} exceedances. Based on the low correlations between 24 h-average $PM_{2.5}$ concentrations and inversion depth (R=0.272), potential temperature gradient (R = 0.379), wind-speed (R =-0.347) or inversion strength (R = 0.296), we conclude that the characteristics of the surface inversion are of marginal impact for high PM_{2.5} concentrations. The duration of an inversion event has no impact on the magnitude of PM2.5 concentrations. The results also lead to the conclusion that the presence of a surface inversion and calm wind are only necessary, but not sufficient conditions for high PM_{2.5} concentrations. In addition, the atmosphere must be sufficiently dry and cold. If the atmosphere becomes colder than -20 °C and drier than 1 hPa, PM_{2.5} exceedance will occur if a surface inversion exists. For water-vapor pressure less than 2 hPa, the likelihood for exceedances is already 97%. Under these cold and dry conditions, the PM_{2.5} concentrations become temperature-sensitive. On the contrary, if temperatures and water-vapor pressure exceed -20 °C and 2 hPa, respectively, both atmospheric stability and temperature will mainly influence the 24 h-average $PM_{2.5}$ concentration. Under these conditions, the microphysical processes related to fog reduce the $PM_{2.5}$ concentrations.

The fact that temperature is a sufficient condition for elevated PM_{2.5} concentrations suggests that the enhanced emissions as temperatures drop are the major cause for increased PM_{2.5} concentrations. Emissions increase at low temperatures as more energy is consumed for heating and production of electrical power. Emissions from traffic (cold starts, idling of cars, and increased use of cars for even short distances) also increase with decreasing temperature. The insensitivity of PM_{2.5} concentrations to wind direction leads to the conclusion that none of the point sources is a major cause for the 24 h-average PM_{2.5} concentrations measured at the Fairbanks downtown site. Based on our study we conclude that reducing the emissions of area sources and traffic could be an effective measure to reduce the frequency of PM_{2.5} exceedances during winter in Fairbanks.

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