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Impact of nocturnal planetary boundary layer on urban air pollutants: Measurements from a 250-m tower over Tianjin, China

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ABSTRACT

It is well known that nocturnal planetary boundary layer (NPBL) has important effects on urban air pollutants. However, the direct measurements of the interactions between the NPBL height and urban air pollutants are normally difficult, because such measurements require continuous vertical profiles of air pollutants and meteorological parameters. This paper provides an unique data, which temperature, NPBL, NO_x and O_3 concentrations are measured at a 250-m meteorological tower in the city of Tianjin, China (a much polluted city located in central-eastern China). The results are analyzed to study the trend of NPBL and the impacts of NPBL on air pollutants in the city. The results show that the measured NPBL height ranges from 100 m to 150 m. The measurement of 10-year trend of the NPBL height suggests that the averaged NPBL height increases by about 20% between 1995 and 2006. The results also show that the NPBL height has important effects on air pollutants. This study suggests that NO_x and O_3 concentrations are strongly anti-correlated inside of the NPBL height. During nighttime, NO_x is directly emitted from the surface and is limited to inside of NPBL (40 m), resulting in high NO_x concentrations near the surface. The high NO_x concentrations depress O_3, producing low O_3 concentrations near the surface. The measurements of vertical gradient of O_3 show that about 30–50 ppbv of O_3 concentrations are chemically destroyed due to the surface emission of NO_x during nighttime, suggesting that NPBL plays important roles in regulating the diurnal cycle of O_3 at the surface.

1. Introduction

Recent satellite observations show that there are high concentrations of air pollutants in eastern China (high NO_2, CO, and aerosols) [11,13], and the high concentrations of air pollutants are continuously increasing [12,16]. The satellite data also reveals that the high air pollutants are located in highly populated areas, including several mega cities (defined as having a population of 10 million or more), such as Beijing, Shanghai, Guangzhou, etc. Tianjin is one of the mega cities within eastern China (at longitude of 117°E and latitude of 39°N) with about 1 million populations. The economical development in this region is very rapid with an annual growth rate of GDP at 14.5%.

Planetary boundary layer (PBL) has an important impact on urban air pollutants [14]. The pollutants, such as aerosols, CO, and NO_x ($NO + NO_2$) are directly emitted from the surface, and are concentrated in the PBL, resulting in high concentrations inside of the surface. Lena and Desiato [7] suggested that the PBL has strong influences on vertical diffusivity (k_x) and surface concentrations of air pollutants. Qin and Kot [10] suggested an average vertical difference of approximately a factor of 2 for both CO and NO_x between street level and the 25-m level. Vakeva et al. [15] observed the dilution factor to be on average 5 between street and rooftop (22 m) levels for CO and NO_x; the formation of secondary pollutants was found to significantly change the gradient of NO, NO_2 and O_3 from that due to the dilution only. Costabile and Allegrini [3] suggested that the concentrations of air pollutants such as NO_x and O_3 characterized by very fast chemical reactions can significantly vary within urban street-canyon due to the short distances between sources and receptor. Tie et al. [14] suggested that CO and NO_x concentrations have maximum values in early morning in Mexico City due to a very shallow PBL height. In noontime, the PBL height rapidly increases, producing a rapid dilution of CO and NO_x. As a result, concentrations of CO and NO_x are significantly reduced during noontime.

Rapid economical development and expanding of mega cities have important effects on the PBL height. Urbanization leads to the replacement of natural surfaces with buildings and paved

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surfaces, and alternates the surface heat, moisture and momentum exchange processes in the atmospheric boundary layer. As a result, urbanization leads to changes in the PBL height and air quality [8]. Study by Civerolo et al. [5] showed that extensive urban growth in the NYC metropolitan area has a potential to increase afternoon near-surface temperatures and the PBL height.

The nocturnal PBL is difficult to be measured, especially for its long-term trend. In this paper, we will examine vertical profiles of chemical and meteorological data measured from a 250-m tower in the city of Tianjin. The measured NPBL height and its impacts on air pollutants will be studied. The paper will be organized in the following way. In Section 2, we will describe the measured data in the tower. In Section 3, the analysis of the trend of the NPBL, and the relationship between the NPBL height and air pollutants measured in the tower will be examined.

2. Methods

The nocturnal PBL can be either determined by vertical temperature profiles, or by air pollutant structures (such as lidar measurement of aerosol vertical distributions). Vertical temperature profiles can be measured by balloon soundings or meteorological towers. In 1984, a 250-m meteorological tower was established in the city of Tianjin, and meteorological parameters, including wind, temperature and humidity profiles, are measured at 5 m, 10 m, 20 m, 30 m, 40 m, 60 m, 80 m, 100 m, 120 m, 140 m, 160 m, 180 m, 200 m, 220 m, and 250 m in every 10 s. With this relatively detailed vertical temperature profile, the vertical gradient is calculated as

$$\frac{\Delta T}{\Delta Z} = \frac{T(z+1) - T(z)}{Z(z+1) - Z(z)}$$

where $T(z+1)$ and $T(z)$ represent the measured temperatures at levels $z+1$ and z, and $Z(z+1)$ and $Z(z)$ represent the altitudes at levels $z+1$ and z. In this study, the height of the NPBL is determined by the top of the positive temperature vertical level.

From late 2006 to the present, air pollutants, including O_3 and NO_x, are measured with four levels in the tower. The O_3 concentrations are measured using O_3 Analyzer (model 400E), and the NO_x concentrations are observed by nitrogen oxides analyzer (model 200E). All the air pollutants are measured in every 1 min, and hourly averaged data is presented in this study. The vertical levels of the instruments are placed in the following orders. The first level (level 1) is near the surface (2 m above the ground), and other 3 levels are located at 40 m, 120 m and 220 m above the surface, respectively.

3. Results and analysis

3.1. Characterizations of nocturnal PBL

The NPBL height is determined by the measured vertical profile of temperature, and the temperature is strongly influenced by the surface characterizations of the city. For example, the heat capacity and albedo of the solar radiation are very different between concrete (urban surface) and soil (rural surface) [8,2]. As a result, expanding of urban areas leads to increase in the surface temperature at center of large cities, which is often referred to "urban heat island" effect [6]. The increase of heat in urban areas could also produce changes in the vertical profiles of temperature and the NPBL height.

Fig. 1 shows the land surface changes in the city of Tianjin from 1993 to 2006. The red dot in the figure shows the location of the meteorological tower. The surface map indicates that the city has rapidly expanded during the last 14 years. The city area (as indicted by the purple color) is almost doubled. The location of the tower located at outside of the city in 1993. With the expanding of the city, it located at inside of the city in 2006. As a result, the surface temperature at the site of the tower was increased. Fig. 2 shows that the surface temperature was increased by about 2 °C between 1995 and 2005 at the tower location. When the temperature is increased due to the enhancement of "urban heat island", the NPBL height is also increased [5]. Fig. 3 shows a trend of the NPBL height from 1995 to 2006, and it suggests that the NPBL height was increased by about 20% during this period. The NPBL height was about 100 m in 1995, and increased to about 120 m in 2006. This increase in the NPBL could have important effects on the concentrations of air pollutants during nighttime.

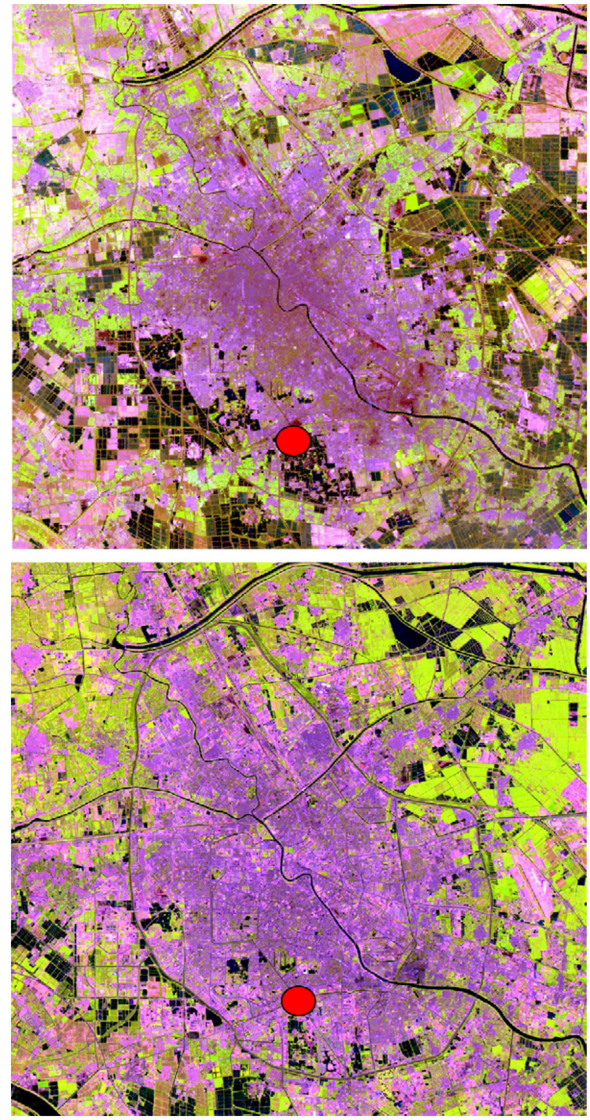

Fig. 1. Display of land surface changes in the city of Tianjin from 1993 to 2006. The upper and lower panels represent the land cover in 19xx and 20xx, respectively. The red dot shows the location of the meteorological tower. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

3.2. Impacts of the nocturnal PBL on air pollutants

At the present, air pollutant data from the tower is collected and analyzed from September 2006 to November 2006. In order to study the impact of the NPBL height on the concentrations of air pol-

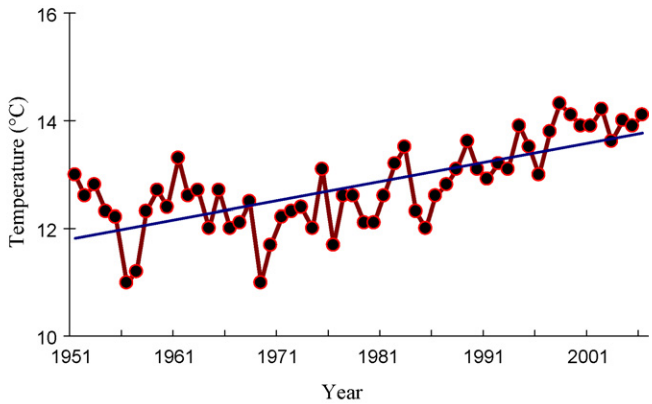

Fig. 2. Measured surface temperature from 1951 to 2005 in the city of Tianjin.

lutants, we first only analyze the structure of the NPBL height during this period, including its occurrence duration and diurnal variation. The NPBL heights in fall (Sept–Oct–Nov), winter (Dec–Jan–Feb), spring (Mar–Apr–May), and summer (Jun–Jul–Aug) are shown in Fig. 4. Fig. 5 shows the averaged NPBL height in different seasons. It shows that the seasonal variation of the NPBL height is generally small, ranging from 115 m to 142 m. There is an indication that the NPBL height is higher during morning period (1:00–7:00 am) than during evening period (19:00–24:00 pm). During the morning period, the NPBL height tends to be slightly higher in summer and lower in winter. However, during the evening period, this tendency disappears. Because the seasonal variations of the NPBL height are relatively small, we expect our analysis of the effect of NPBL on air pollutants in fall season can be applied in other seasons. However, because there is only data analysis in fall season, the seasonal variations of the impacts of NPBL on air pollutants due to other seasonal factors, such as the air pollutant emissions, the winds, and the strength of mixing processes, which cannot be taken into account in this analysis. The result also shows that the NPBL height has weak diurnal variation, ranging from 100 m to 150 m. The duration of the NPBL is about 12.5 h starting from 19:00 pm and ending at 7:30 am in the next day as indicated by red lines in Fig. 4. We note that there is also slight change in the duration of NPBL. In summer, the duration is smaller than in winter.

Because NPBL is a dynamical stable layer, the vertical mixing of pollutants between inside and outside of the NPBL is very weak [1], the measured air pollutants are distinguished by two different characterizations, i.e., air pollutants at inside and outside of NPBL. At the level 1 (surface) and the level 2 (40 m), the air pollutants are normally at inside of the NPBL. By contrast, the measured air pollutants at the level 4 (220 m) are generally at outside of the NPBL. The level 3 (120 m) is located at the top of the NPBL. Thus, air pollutants at this level are considered as at the transition zone between inside of the NPBL and outside of the NPBL. Due to the dynamical stability of the NPBL, the surface emitted air pollutants are normally trapped inside of the NPBL, and are not able to mix with the pollutants at outside of the NPBL. Very different characterizations of air pollutants between inside (levels 1 and 2) and outside (level 4) of NPBL are measured in the tower.

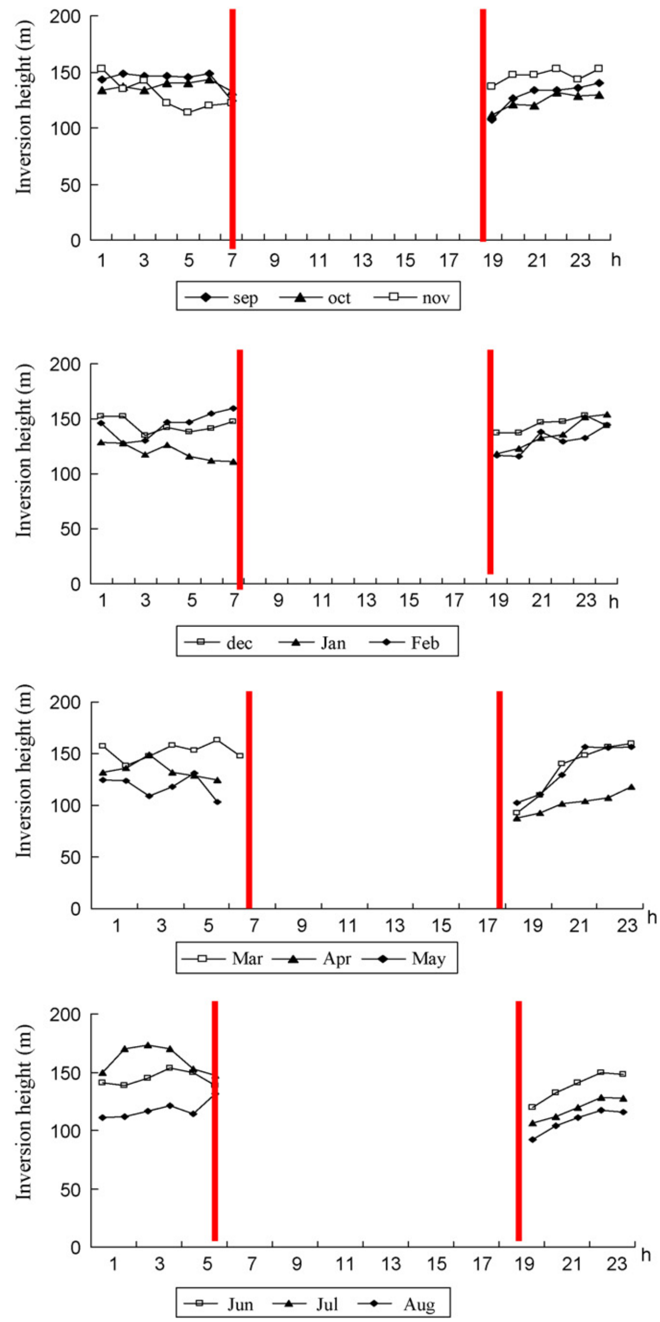

Fig. 4. Measured NPBL heights in fall (Sept–Oct–Nov), winter (Dec–Jan–Feb), spring (Mar–Apr–May) and summer (Jun–Jul–Aug) from the tower. The NPBL height has a slightly seasonal and diurnal variation, ranging from 100 m to 150 m. The duration of the NPBL is indicated by red lines. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

Fig. 6 shows O_3 and NO_x concentrations in September. It indicates that the NO_x concentrations (upper panel) at inside of the

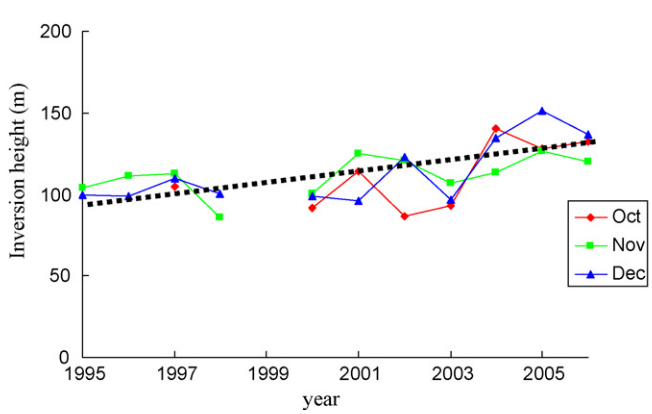

Fig. 3. Measured NPBL height trend from 2000 to 2006 in the city of Tianjin.

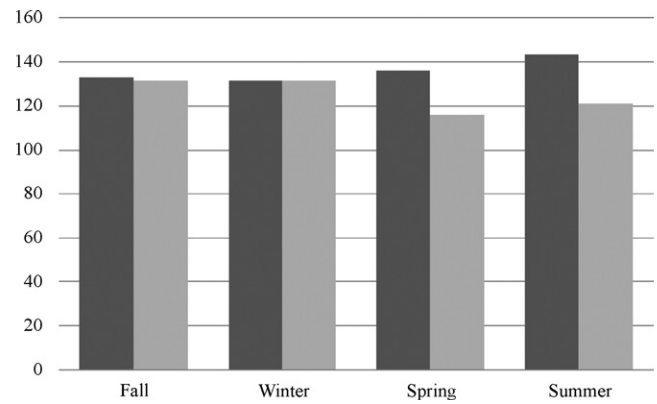

Fig. 5. The averaged NPBL height in different seasons. The dark bars represent the NPBL height during the period from 1:00 am to 7:00 am, and the grey bars represent the NPBL height during the period from 19:00 pm to 24:00 pm.

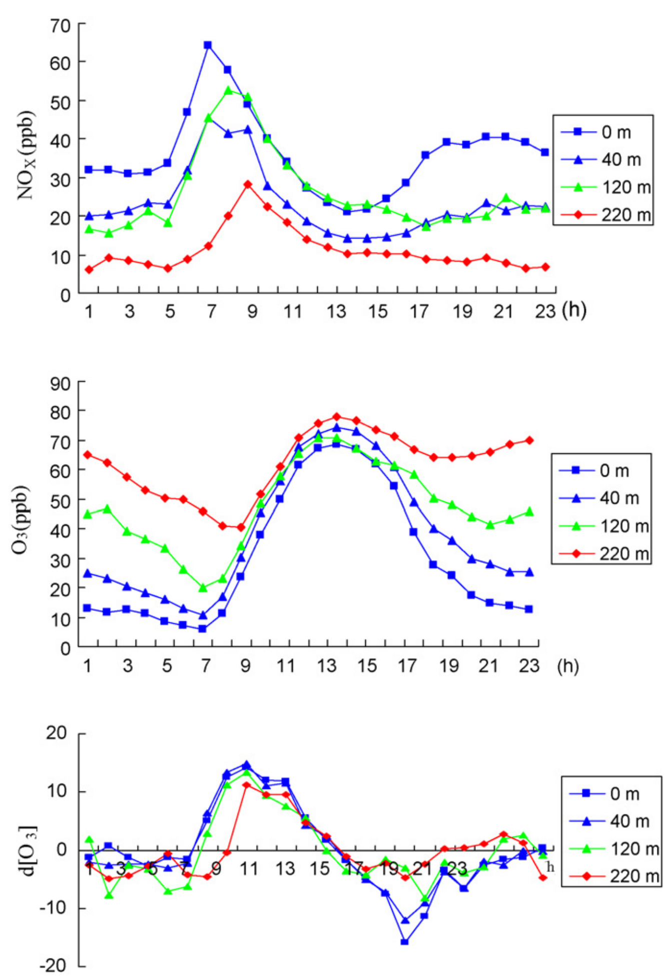

Fig. 6. Measured NO_x (upper panel) and O_3 (lower panel) concentrations in September at four levels from the tower. At the level 1 (surface) and level 2 (40 m), the measured air pollutants are below the NPBL (shown by blue lines). In contrast, the measured air pollutants at the level 4 (220 m) are located above the NPBL (shown by red line). The level 3 (120 m) is located at the top of the NPBL, and the air pollutants are in the transition zone between inside of the NPBL and outside of the NPBL (shown by green line). The bottom panel shows the O_3 tendency ($d[O_3] = O_3[t+1] - O_3[t]$) at different levels. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

NPBL are higher than the concentrations at outside of NPBL. At the surface level, the NO_x concentration is the higher than the values at other levels, and appears a strong temporal variation. The NO_x concentration is about 30 ppbv during nighttime, and increase to a maximum of 60 ppbv in early morning before the NPBL is transited to the daytime PBL. The rapid increase in the NO_x concentrations from nighttime to early morning is resulted from the increase in the city traffic emissions, while the NPBL height is relatively unchanged as indicated in Fig. 4. During daytime, the PBL height rapidly increases from about 120 m to about 1000–3000 m [9]. As a result, the pollutants at the surface are quickly diluted in the daytime PBL, and the surface concentration of NO_x is reduced due to the increase in vertical mixing. During evening (19:00 pm), the PBL height reduces to 100–150 m (the NPBL regime). With the significant reduction in the PBL height, the surface concentration of NO_x is increased from 20 ppbv at noontime to 40 ppbv at 19:00 pm.

Another unique aspect of the tower measurements is that it provides detailed vertical distributions of air pollutants. As indicated in Fig. 6, the vertical gradient of NO_x concentration is very strong near the surface. The concentration of NO_x at 40 m is rapidly reduced compared to the surface concentration in the NPBL, with a vertical gradient of 0.3–0.5 ppbv/m. By contrast, the vertical gradient of NO_x concentration between 40 m and 220 m is small, with the vertical gradient of 0.05–0.1 ppbv/m. This result indicates that during night the NO_x released from the surface is mostly limited to a narrow layer near ground (from surface to 40 m). Above the NPBL, the NO_x concentration is only 5–10 ppbv compared to 30 ppbv at the surface during night. This characterization of vertical gradient of the NO_x concentration suggests that there is a separation for air between inside and outside of the NPBL. In the following sections, we will name these two regimes as (a) the inside PBL air and (b) the outside PBL air. Because the strong diurnal variation of the PBL, air at the same level can either consider as the insider PBL air or the outside PBL air. For example, during daytime, the NO_x concentrations at the 220 m level is considered as the inside PBL air, and during nighttime, it is referred as the outside PBL air.

The measured diurnal variation of NO_x at different levels shows that there is a maximum value in morning at all levels. However, there is an important characterization that the maximum of NO_x concentration at 220 m occurs at 9:00 am, while the surface maximum occurs at 7:00 am. This time lag between the surface and 220 m of the NO_x maximum suggests that the NO_x is originated from the surface (or directly emitted from the surface), and has been propagated in the upper levels.

The measured O_3 vertical gradient shows that O_3 concentration is strongly affected by NPBL (see lower panel of Fig. 6). Fig. 6 indicates that during nighttime and early morning O_3 concentration is low with a minimum of 10 ppbv at the surface. The low surface O_3 concentration during nighttime and early morning is also observed in other large cities, such as Mexico City and Houston ([14]; Zhang et al. [17]). Their studies suggest that O_3 is reacted with NO resulted from primary NO_x emission during night, leading to a very low O_3 concentration during nighttime. As suggested by Emmerson et al. [4], the reaction of O_3 with alkenes plays important roles in depressing O_3 and is also an important radical source during nighttime. However, due to lack of measurement of alkenes, we will not discuss this reaction in detail. The measured O_3 concentrations at different levels show that O_3 concentration increases with altitude, reaching to highest values of 50–70 ppbv at 220 m, indicating that the vertical distributions of NO_x and O_3 concentrations are anti-correlated. This anti-correlation between O_3 and NO_x vertical profiles during night supports the idea that the nighttime O_3 is strongly depressed by the high NO_x emitted at the surface.

The measured O_3 diurnal variations at different levels of the tower show that O_3 concentrations are highest at 220 m and lowest in the surface. By contrast, the diurnal variation of O_3 concentrations is lowest at 220 m and highest at the surface. In order to analyze the difference of the O_3 variations at different levels, O_3 tendency is analyzed. The O_3 tendency is expressed as

$$d[O_3] = O_3[t+1] - O_3[t]$$

where $O_3[t]$ represents O_3 concentration at hour t, and $O_3[t+1]$ represents O_3 concentration at next hour $t+1$. The bottom panel of Fig. 6 shows the O_3 tendency during September. The result shows that during nighttime, there are no photochemical activities, and the chemical ozone production is close to be zero. As a result, the tendency of ozone is in negative value during nighttime. The negative O_3 tendency during nighttime suggests that the O_3 chemical destruction due to the reaction of O_3 + NO plays important roles in controlling O_3 concentrations during nighttime. During daytime, however, O_3 is chemically produced, and the O_3 tendency becomes positive value at morning, suggesting that the O_3 chemical production becomes a dominated factor. We note that there are several interesting aspects regarding the O_3 tendency. (1) The magnate of the O_3 tendency value is generally smaller during nighttime than daytime. (2) The transition time (Tt) from negative value to positive value for the O_3 tendency at different altitudes occurs at different time. For example, the Tt occurs at 6:00 am, 7:00 am and 8:00 am at 0–40 m, 120 m and 220 m, respectively. The time-lag of the Tt further suggests that the effect of O_3 destruction in NPBL is originated at the surface, and is propagated from the surface to 220 m. (3) During daytime, the O_3 tendency is rapidly increased during morning, and reaches a maximum value of 10–15 ppbv/h. (4) During afternoon, the O_3 tendency changes to negative value around 13:00 pm, suggesting that the rate of O_3 production is smaller than the rate of O_3 destruction at that time. In addition to the O_3 tendency, we also calculate the difference between the maximum of O_3 concentrations and the minimum of O_3 concentration ($\Delta O_3 = O_3(\max) - O_3(\min)$). The result shows that the ΔO_3 is about 35 ppbv at 220 m and is about 65 ppbv at the surface. At the surface, the ΔO_3 is largest. Because the maximum O_3 concentration at the surface in noontime is similar to the value at 220 m, the large ΔO_3 at the surface is mainly due to the fact that the O_3 concentration at the surface is about 30 ppbv lower than the value at 220 m in the early morning. This result suggests that about 30–40 ppbv of O_3 during

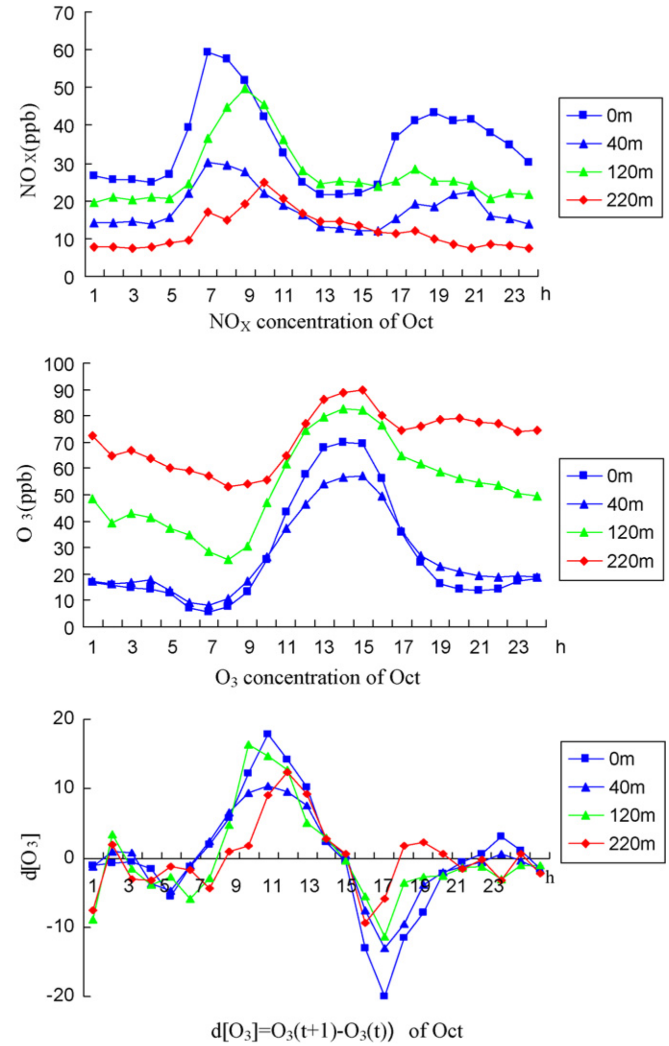

Fig. 7. Same as Fig. 6, except for October. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

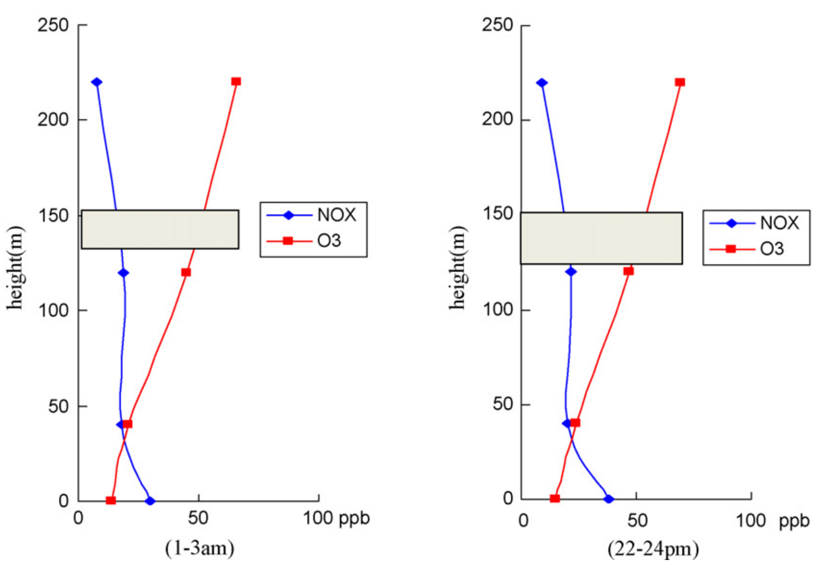

Fig. 8. Measured averaged (from September to November of 2006) vertical structures of NO_x and O_3 at inside and outside of the NPBL.

nighttime and early morning are reduced inside the NPBL (due to the surface NO_x emissions), and the reduction of the O_3 concentration during nighttime and early morning plays important roles in regulating the O_3 diurnal variation at the surface.

Fig. 7 shows O_3 and NO_x concentrations in October. Although the figures show generally a similar characterization of NO_x and O_3 to the results shown in Fig. 6, some differences between Figs. 6 and 7 are also shown. For example, the secondary maximum of NO_x concentrations is generally weaker during October than September. The ozone production during daytime is higher at 40 m than the value at the surface.

Fig. 8 shows averaged (from September to November of 2006) vertical profiles of NO_x and O_3 at inside and outside of the NPBL. The results clearly indicate that (1) NO_x and O_3 concentrations are strongly anti-correlated. This strong anti-correlation gives more supportive evidences that O_3 in the NPBL is strongly depressed by the high NO_x concentrations: (b) there is a strong vertical NO_x gradient between the surface and 40 m, which is much lower than the NPBL height (100–150 m), indicating that the vertical mixing inside of the NPBL is considerably weak inside the NPBL. We note that the vertical resolutions of measured temperature and air pollutants are different. The temperature profile is measured in more levels which can be more accurately used to determine the NPBL height. The pollutants, however, measured at only four levels. The low resolution in vertical levels of the air pollutants can give insights of the vertical distributions of air pollutants, but cannot give detailed vertical profiles.

4. Summary

Temperature, nocturnal planetary boundary layer (NPBL), NO_x and O_3 concentrations measured from a 250-m meteorological tower (located in Tianjin, China) are analyzed to study the characterizations of the NPBL and the impact of the NPBL on air pollutants. Tianjin is one of the mega cities within eastern China, with population of 1 million. The economical development in this region is very rapid with annual growth of GDP at 14.5%. Due to rapidly economical development and expanding of the city, the NPBL height increases from 100 m to 120 m between 2000 and 2006. Because the NPBL has significant impacts on air pollutants, the changes of the NPBL lead to important effect on air pollutants. The results show that during nighttime the NO_x emitted from the surface is mostly limited to below 40 m (inside of the NPBL). During nighttime, the NO_x concentration is only 5–10 ppbv above the NPBL compared to 30 ppbv at the surface, indicating a separation of NO_x between inside and outside of the NPBL. The O_3 concentration is also strongly affected by the NPBL. During nighttime and early morning, the O_3 concentration is low inside of the NPBL with a minimum of 10 ppbv at the surface, and the concentration increases with altitudes, reaching a maximum of 50–60 ppbv at 220 m level, suggesting that O_3 is reacted with NO during nighttime, which leads low O_3 concentration during nighttime. The strong anti-correlation of vertical distributions of NO_x and O_3 provides a clear evidence that the high NO_x concentration inside the NPBL reduces about 30–50 ppbv of O_3 during nighttime and early morning. As a result, the surface O_3 diurnal variation is strongly regulates by the NPBL.

Acknowledgements

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