

Letter of Responses

Reply to anonymous referee 1's comments (C3934)

We thank the reviewer (Anonymous Referee #1) for their helpful comments and suggestions which will help improving the original paper. Below are our itemized responses.

General comments

1) The authors conclude, that “NH₃ concentrations show regular seasonal variations, having significantly higher summertime concentrations”. While the second half of this sentence is undisputable, the first claim is quite far reaching, taking into account that it is based on two (!) field campaigns within one winter and one summer period only (23 resp. 30 cases). To underpin this conclusion, either more campaign data across different years, or evidence from previous measurements conducted in the same area with supporting data would be required. The references provided and discussed, however, are only used to underpin correlations of different parameters or providing support for individual patterns observed, not for this overall conclusion. It is clear, that the availability of such measurement data for China/Beijing is all but great and it may be difficult to obtain. Yet, to conclude on regular seasonal variations, two periods in one year is hardly justifiable at this point.

Reply:

According to studies of the ammonia level in China, the biggest contributors are livestock (which contributes about 30%–60%) and the application of nitrogenous fertilizer (which contributes about 17%–47%). The characteristics of these sources determine the temporal and spatial NH₃ distribution. Seasonality of NH₃ emissions in China has also been determined. The ammonia concentration in northern cities, as Beijing, is relatively high during the spring and summer farming period, while in southern cities no obvious seasonal variation occurs because the farmland is worked during all four seasons. Other studies have carried out that NH₃ concentrations showed distinct seasonal variations, with highest concentrations in summer and lowest concentrations in winter. Source strength and removal efficiency can explain the seasonal variations of NH₃ concentrations. In summer, high temperatures will favor ammonia volatilization from urea and/or ammonium bicarbonate applied to crops. High temperatures in summer will also favor NH₃ emission from other sources, such as animal housing, landfill, laystalls and farmers' toilets, animal

manure, natural and fertilized soils, and vegetation. Therefore, atmospheric ammonia concentrations were highest in summer at Beijing site. In winter, no fertilizer was applied and the temperature was very low at Beijing site. So less ammonia volatilized and hence the atmospheric NH₃ concentrations were low.

Thus, in this paper we cited all available references for ammonia measurements and the majority of them use two sampling periods (one winter and one summer period) to determine seasonal variations of NH₃ at Beijing. Instead, the remaining available references show previous NH₃ measurements across different years or across different months of year carried out in the China or in Beijing to determine seasonal variations of NH₃.

Yet, if the references cited and discussed in this paper don't provide support for our measurements or don't demonstrate this seasonality for NH₃ although they also refer to only two periods, we will agree to replace the sentence (p.14216, 3.1, 9/11) "NH₃ exhibited a distinct and significant ($p < 0.001$) seasonal variation with higher concentrations in summer than in winter (Fig. 1)" with "NH₃ exhibited a distinct and significant ($p < 0.001$) temporal variation with higher concentrations in summer than in winter (Fig. 1)". Thus, we will replace the words "seasonality" or "seasonal" with "temporal variation". Thus, we will modify the section 3.1 (after line 5) as follows:

"The temporal variations are evaluated applying the paired t-test to determine the significance at the 0.05 level ($p \leq 0.05$) of differences among the mean of components by examining the amount of variation between the samples. NH₃ exhibited a distinct and significant ($p < 0.001$) variation with higher concentrations in summer than in winter (Fig. 1). This difference in NH₃ concentrations has also been observed by other papers and is in agreement with the temperature dependence of NH₃ emissions from animal waste, natural and fertilized soils, and vegetations (Langford and Fehsenfeld, 1992; Langford et al., 1992; Yamamoto et al., 1995, Asman et al., 1998; Lefer et al., 1999; Aneja et al., 2000; Robarge et al., 2002; Pryor et al., 2001), which are at low values during winter. The characteristics of these sources determine the temporal and spatial NH₃ distribution. Source strength and removal efficiency can explain the seasonal variations of NH₃ concentrations. In summer, high temperatures will favor ammonia volatilization from urea and/or ammonium bicarbonate applied to crops. High temperatures in summer will also favor NH₃ emission from other sources, such as animal housing, landfill, laystalls and farmers' toilets, animal manure, natural and fertilized soils, and vegetation. Therefore, atmospheric ammonia concentrations were highest in summer at Beijing site. Thus, in agreement with past studies, higher ammonia emissions occurred when air temperature increased, suggesting intense local sources for NH₃ during the summer season at Beijing."

2) The conclusion, that no diurnal variation could be observed at all in either the winter or summer period is surprising and contradicts the temperature effect discussed elsewhere in the paper. The influence of temperature on observed NH₃ levels is missing in the conclusions, by the way. Unfortunately, only Fig. 3 enables the reader to assess this claim, and only for the winter period. At minimum, one figure for winter and one for summer, over several days (a week?) should be displayed. A closer look then at Fig. 3 then does suggest a peak at noon, likely dependent on temperature increase and human activity (NO_x peak is similar) on day 1 and a less pronounced peak on day 2, somewhat earlier. A quick calendar check reveals that Feb 9, 2007 was a Friday (day 1), Feb 10, 2007 a Saturday (day 2), which may explain the lack of a dominant peak on the second day due to less traffic, but a comparison with hourly temperatures would be prudent as well to investigate the reason for this missing diurnal pattern. It can be safely assumed that the diurnal pattern will be even more pronounced in the summer period. In addition, there is likely a weekly cycle due to the influence of e.g. traffic emissions.

Reply:

We followed your suggestions discussing more detailed the results and we will modify and add relevant sentences in sections 3.2.1 and 3.2.2 such that the conclusions on the NH₃ diurnal pattern are better supported by analysis of our data. We will also modify old Figures, we add new Figures, and, thus, we will necessarily change the numbers of all Figures, such as the old Fig. 9 which will become Fig. 10. They will be showed to the end of this point. In addition, we will delete the old Fig. 5. Thus, we will modify section 3.2.1 and 3.2.2 as follows:

Section 3.2.1 (after the sentence p. 14218, 3.2.1, 4/6)

“However, a weak but significant linear correlation ($R^2 = 0.32$, $p = 0.01$) can be obtained between NH₃ and air temperature considering ammonia concentrations measured from 12:00 on 9 February, when the air temperature reached maxima values (12:00 – 16:00 on two days), to the end of intensive measurements on 10 February. This weak temperature dependence of NH₃ reflect the small influence of the emission sources from agricultural activity which increase as the temperature rises. Ammonia emissions from animal manure, natural and fertilized soils, and vegetation will increase with temperature owing to the temperature dependency of aqueous-phase partitioning between NH₃ and NH₄⁺ in these systems, as well as the atmospheric equilibrium between NH₃ with

volatile ammonium nitrate. In fact, during this period the winds came principally from northwest direction (73%) (Fig. 2b) coinciding with the direction of agricultural areas such as Xibeiwang and Changping District which are located at the northwest suburb of sampling site (Shen et al., 2009; Zhang et al., 2010). The temperature dependence of NH_3 disappeared from 00:00 to 12:00 on 9 February with prevailing winds from southeast and southwest (60%) (Fig. 2b) and lower temperatures. This different behaviour may be explained from prevalence of local sources, such as traffic emissions, in determining the atmospheric NH_3 concentrations during this period.

While agriculture is the main source of atmospheric ammonia in Beijing, the contribution of vehicles, equipped with catalytic converters, especially since the introduction of three-way-catalysts, to non-agricultural NH_3 emissions has recently been considered and might be the most important factor influencing ammonia concentrations at urban locations and near roads (Sutton et al., 2000; Kean et al., 2000, Heeb et al., 2008). Also Beijing city is configured such that it is served by several ring roads with heavy traffic (Fig. 3b). Fig. 3a and 3b show Beijing city, measurement site and surrounding regions, where are the major NH_3 emission sources. Therefore, in order to examine the contribution of traffic to NH_3 concentrations it may be useful to compare the ammonia concentrations with those of primary non-reactive pollutants mainly emitted by motor-vehicle exhausts (Perrino et al., 2002), such as nitrogen oxides (NO_x) and carbon monoxide (CO), throughout this winter period. The time series of NH_3 , NO_x , CO, wind direction, wind speed, temperature and relative humidity are shown in Figure 3. Both CO and NO_x are commonly used as traffic emission indicators (He et al., 2002; Meng et al., 2008, Chak and Yao, 2008). Hao et al. (2005) estimated that the emissions in Beijing from vehicles, power plants, and industries in 1999 accounted for 35%, 27% and 26% in the total local NO_x emissions, respectively. They found that 74% of the ground NO_x was due to vehicular emissions while power plants and industrial sources only contributed 2% and 13%, respectively. In addition, in Beijing and Guangzhou, automobile pollution contribution in terms of CO was estimated to be more than 80% with two peak vehicle pollution levels occurring during each day, one from about 8:00–10:00 and the other from 15:00–17:00 during the rush hours (Hao et al., 2000).

NO_x peaks were observed between 08:00 and 12:00 on 9 February (Fig. 4). Wind from the southeast and southwest was dominant at the sampling site on 9 February, while wind from the northwest dominated on 10 February. Wind speeds on 9 and 10 February ranged from 0.02 to 8.85 m/s and from 0.06 to 4.10 m/s, respectively. However, on 9 February the overall NO_x level was significantly higher (from 11.60 to 243.55 ppb) than that on 10 February (from 5.85 to 73.40 ppb). The locations of the major NO_x sources, including local roads (motor vehicles), expressways, and power plants, which are six and principally coal-fired in Beijing, are located from the east to the southwest of the

sampling site (Lee et al., 2009). Thus, the weak wind speeds between 00:00 and 12:00 (0.02 – 1.53 m/s) and southeasterly and southwesterly wind directions on 9 February, in combination with the locations of the sources of NO_x emissions, resulted in higher NO_x concentrations between 08:00 and 12:00 due to emissions from Friday traffic during this daytime. Instead, the high wind speeds (0.30 – 3.83 m/s) and northwesterly wind direction on 10 February resulted in lower NO_x concentrations at the sampling site. In fact, NO_x concentration was about 31.55 ppb at 08:00 on 10 February, which is about eight times lower than that recorded at the same time of day on 9 February (243.55 ppb). These differences in NO_x concentrations between two days might reflect the characteristic of Friday traffic (on 9 February) and the wind conditions, which were marked by low winds and prevailing wind direction from the center of Beijing toward the sampling site (60% from southeast and southwest). The scatter plot of NH₃ concentration vs. NO_x concentrations during this two days (Fig. 5) showed a good and significant (at the 99.9% confidence level, $p < 0.001$) linear correlation of the two data sets ($R^2 = 0.65$), supporting the hypothesis that the traffic is also an important source of NH₃ in this season within the city. However, for the NH₃ and NO_x data, the best correlation ($R^2 = 0.80$, $p < 0.001$) was obtained considering concentrations measured on 9 February during the formation of the highest peaks of ammonia and nitrogen oxides due to rush-hour traffic. However, the amount of scatter about these regression lines indicates that other ammonia sources not linked to NO_x are also significant. In a city centre location, these will largely be human sources (Whitehead et al., 2007). In contrast, the correlation between NH₃ and NO_x did not occur considering concentrations measured only on 10 February during high wind speeds and northwesterly wind direction from where no major sources of NO_x are located. These results, in combination with weak temperature dependence of NH₃ during on 10 February, as said above, suggest that NH₃ concentrations were influenced mainly by non traffic-sources such as agricultural emissions.

In addition, the same findings were also observed comparing the ammonia concentrations with those of CO (Fig. 4). CO peaks were observed between 08:00 and 10:00 on 9 February. However, on 9 February the overall CO level was significantly higher (from 0.16 to 6.17 ppm) than that on 10 February (from 0.05 to 1.62 ppm) due to time period and wind conditions, as said above for NO_x. The scatter plot of NH₃ concentration vs. CO concentrations during this two days (Fig. 6) showed a good and significant (at the 99.9% confidence level, $p < 0.001$) linear correlation of the two data sets ($R^2 = 0.67$), supporting the hypothesis that the traffic is also an important source of NH₃ in this season within the city. However, for the NH₃ and NO_x data, the best correlation ($R^2 = 0.84$, $p < 0.001$) was obtained considering concentrations measured on 9 February during the formation of the highest peaks of ammonia and carbon monoxide due to rush-hour traffic. In contrast, the correlation

between NH_3 and CO did not occur considering concentrations measured only on 10 February during high wind speeds and northwesterly wind direction. These results, in combination with weak temperature dependence of NH_3 during on 10 February, further show that NH_3 concentrations were influenced mainly by non traffic-sources such as agricultural emissions.

Thus, the presence of local sources, such as traffic emissions, on 9 February and non-traffic sources on 10 February probably explains why the temperature didn't determine NH_3 concentrations in Beijing and why a diurnal cycle of NH_3 has not been observed because it showed high variability due to variation in emissions during two days of winter intensive measurements.

To help interpret further in general terms the source regions affecting the sampled ammonia and, thus, to identify the origin and transport pathway of large-scale air masses, 24-h backward trajectories arriving at the sampling site were calculated for two days of winter intensive measurements (Fig. 7). The trajectories at 100 m arrival height above ground level were computed every 2 h (from 01:00 on 9 February to 23:00 on 10 February, local time) using the NOAA ARL HYSPLIT trajectory model (<http://ready.arl.noaa.gov/HYSPLIT.php>). The Fig. 7 shows dominant transport of air masses from northwest (83%) of Beijing coinciding with the direction of Inner Mongolia and agricultural areas such as Hebei province, which completely surrounds Beijing and Tianjin municipalities. Actually, intensive agriculture is concentrated in the North China Plain which includes five provinces (Hebei, Henan, Shandong, Jiangsu and Anhui) and two municipalities (Beijing and Tianjin). Of the total agricultural ammonia emissions in the North China Plain, the Hebei, Henan and Shandong provinces take the larger part (Zhang et al., 2010). Contributions of NH_3 emissions from livestock and fertilizer activities were also found in Inner Mongolia (Klimont, 2001; Ju et al., 2004). Beijing is downwind of Shanxi, which is one of the largest coal mining and coal-fired power generation provinces in China, which can emit large amounts of NH_3 (Meng et al., 2010). In addition, these air masses passing the northwestern regions arrived faster (8.17 – 15.70 m/s) and, thus, they had less time to accumulate ammonia. Instead, as said above, on 9 February local wind speeds, especially between 00:00 and 12:00, coming principally from southeast and southwest of Beijing arrived slower and, thus, local air masses had much time to accumulate pollutants. This suggests that NH_3 received the largest impacts from local emissions in the city of Beijing, where morning peaks of NH_3 , which correlated with morning traffic emissions, were observed.

In contrast, on 10 February local high wind speeds from the northwest dominated in Beijing, where the correlation between NH_3 and both CO and NO_x did not occur, in combination with temperature dependence of NH_3 . This suggests a possible and further contribution of regional and agricultural

sources, located to the northwest of Beijing, to the ammonia concentrations observed in Beijing during on 10 February.

These results reveal that during the winter NH₃ concentrations are influenced by meteorological conditions (wind direction, wind speed and, sometimes, temperature), local and regional sources in the winter 2008 in Beijing. Further modeling studies are needed to quantify the contribution of local versus regional sources to the atmospheric ammonia variations.”

Section 3.2.2 ((after the sentence p. 14219, 3.2.2, 18/21)

“The highest ammonia concentrations were principally observed when the wind was from the northwest during low wind conditions (Fig. 8), reflecting the large contribution due to agricultural activity and fertilizer use. A moderate but a significant linear correlation ($R^2 = 0.29$, $p < 0.001$) was detected between natural-log transformed ammonia concentrations and wind speeds (Robarge et al., 2002).

However, the diurnal pattern of ammonia did not show a clear and well-defined temperature dependence during the summer field campaign, as it was in the winter. The temperature dependence of NH₃ can be examined further by filtering all the data by hours of day and for only particular hours during all days of intensive summer measurements (from 06:00 to 12:00 on 17 August, from 00:00 to 10:00 on 18 August, from 02:00 to 10:00 on 19 August, from 02:00 to 12:00 on 20 August, from 02:00 to 06:00 on 21 August) there is a weak but significant correlation between temperature and NH₃ concentration (at the 99.9% confidence level, $R^2 = 0.22$, $p = 0.03$) with prevailing winds from northwest. As said for winter period, this weak temperature dependence of NH₃ reflect the influence of the emission sources from agricultural activity coming from northwest suburb of Beijing (Shen et al., 2009; Zhang et al., 2010). The temperature dependence of NH₃ disappeared for the other hours of every day with prevailing winds from south and southwest. This different behaviour may be explained from prevalence of local sources, such as traffic emissions, in determining the atmospheric NH₃ concentrations during this summer period.

The data of Fig. 8 show that NH₃, CO and NO_x had similar temporal patterns, but the correlations between these species were not good. A weak but significant linear correlation occurred between NH₃ and CO ($R^2 = 0.18$, $p < 0.001$) (Figure 6) for all sampling period (17-21 August 2007). For the NH₃ and NO_x data, this correlation did not occur (Figure 5). However, for the CO data, the best relationships with correlation coefficients of 0.73 ($p < 0.001$), 0.69 ($p = 0.03$) and 0.33 ($p = 0.03$) were obtained considering concentrations measured only on 18, 19 and 20-21 August 2007, respectively, during the formation of the higher peaks of NH₃ and CO supporting the hypothesis

that the traffic is also an important mobile source of NH_3 . In addition, for the NO_x data, the best relationships with correlation coefficients of 0.39 ($p < 0.001$), 0.32 ($p = 0.03$) and 0.70 ($p = 0.001$) were also obtained considering concentrations measured for the same days, respectively, in correspondence with the highest concentrations of NH_3 and NO_x . NO_x and CO peaks were observed between 06:00 and 10:00 during all days, in combination with northwesterly wind direction and low wind speeds (0.3 – 1.2 m/s). Winds from the south were dominant at the sampling site for the other hours of day, in combination with the locations of the sources of NO_x and CO emissions. However, the amount of scatter about these regression lines indicates that other ammonia sources not linked to NO_x and CO are also significant. In addition, the correlations between NH_3 and both NO_x and CO were weaker than that in the winter, suggesting that other, non-traffic sources, became significant. The higher temperatures in the summer will increase ammonia emission from agricultural sources, resulting in the breakdown in the relationship between these gases.

The same findings were also observed comparing the ammonia concentrations with those of $\text{PM}_{2.5}$ which accounts for 90% of total PM emissions from on-road vehicles in Beijing (Zheng et al., 2005). $\text{PM}_{2.5}$ peaks were observed between 08:00 and 10:00 during all days (Fig. 8). A weak but significant linear correlation occurred between NH_3 and $\text{PM}_{2.5}$ ($R^2 = 0.17$, $p = 0.004$) (Fig. 9) for all sampling period. However, for the $\text{PM}_{2.5}$ data, the best relationships with correlation coefficients of 0.80 ($p < 0.001$), 0.60 ($p = 0.009$) and 0.30 ($p = 0.03$) were obtained considering concentrations measured only on 18, 19 and 20-21 August 2007, respectively, during the formation of the higher peaks of NH_3 and $\text{PM}_{2.5}$ supporting the hypothesis that the traffic is also an important mobile source of NH_3 .

The highest ammonia values during the day can be also attributed to stable atmosphere conditions. The analysis of the temporal pattern of natural radioactivity (Fig. 8) shows that the convective mixing of the atmosphere occurs between the late morning (12:00-13:00 a.m.) of the previous day and the early morning of the subsequent day (03:00-04:00 a.m.). Subsequently, a rapid increase of natural radioactivity determines atmospheric stability with high values at 6:00 and at 08:00 in the morning. In these conditions, pollution events are generally favoured. This implies that most of the traffic emission is injected into a stagnant atmosphere causing the sharp increase in the concentration of ammonia, which reached the remarkable value of $105.67 \mu\text{g}/\text{m}^3$ (18 August 2007), one of the highest values during the intensive measurements. During the same hours CO and NO_x concentrations reached the levels of $2.15 \text{ mg}/\text{m}^3$ and $112.6 \mu\text{g}/\text{m}^3$ (Figure 6), respectively, and this is an indication of a common origin of these pollutants, i.e., traffic emission (Perrino et al., 2002; Edgerton et al., 2007).

In some studies, increased NH_3 concentrations have also been attributed to the dissociation of particulate ammonium nitrate (Langford et al., 1992; Lee H. S. et al. 1999; Possanzini et al., 1999). Volatilization of NH_3 from the aerosol phase may be significant enough to dominate over traffic emissions during the summer (Whitehead et al., 2007) and mask any correlations with traffic related pollutants. It is known that volatile ammonium salts collected on the Teflon filter (NH_4Cl and NH_4NO_3) dissociate to HCl and HNO_3 , a phenomenon which depends mainly upon meteorological conditions (air temperature and relative humidity), on the aerosol composition, and on the acidity of the particles. In this work the gases evolved from the front Teflon filters were recovered on the back-up Nylon and phosphorous acid-impregnated filters. Data from 2h sampling over the 17-21 August period showed that Teflon filters exhibited a nearly complete depletion of nitrate and chloride (> 90%) which were recovered on Nylon filters. This indicated that nitrate and chloride were almost entirely associated to ammonium. Indeed, a comparison of μmol amount of NH_4^+ with μmol amounts of anions ($\text{NO}_3^- + \text{Cl}^-$) nitrate determined on the back-up filters showed a good correlation ($R^2 = 0.80$, $p < 0.001$). However, the diurnal variation in NH_3 concentrations had a similar trend as for NH_4^+ , with one exception of 17 August. During this day, the formation of aerosol NH_4^+ leads to the decrease of NH_3 concentrations and vice versa. This behaviour could be explained with the displacement of the thermodynamic equilibrium between ammonium salts and their gaseous precursors. While for the other days (18-21 August), the fact that the NH_3 and NH_4^+ exhibited similar patterns is due to dissolution of a significant fraction of NH_3 in humid aerosols under high relative humidity conditions (Hesterberg et al., 1996; Krupa, 2003; Trebs et al., 2004 and 2005; Hu et al., 2008). In fact, the concentrations of ammonia and ammonium reach the maximum values at the same time (between 06:00 and 10:00). Possible evaporation of NH_3 from wet surfaces due to temperature increase just after sunrise, when relative humidities were still high might have caused a significant fraction of gaseous NH_3 to dissolve in still deliquescent aerosols, therefore enhancing aerosol NH_4^+ .

A more detailed study of the behaviour of ammonia and ammonium salts in the atmosphere may be carried out considering the mass balance on back-up filters. In fact, in principle we should find equimolecular amounts of anions ($\text{Cl}^- + \text{NO}_3^-$) and of evaporated ammonia ($\text{NH}_{3\text{ev}}$) determined on the back-up filters. On the contrary, the results reported in Fig. 10 show that during the intensive measurements in the summer period, ammonia frequently exceeded the sum of chloride and nitrate. This unbalance can be regarded as an additional phenomenon which adds ammonia to the dissociation of ammonium nitrate and chloride; a possible explanation is the presence of other anions (e.g. organic matter) or of gaseous ammonia adsorbed or dissolved in deliquescent aerosol ($\text{NH}_{3\text{ads}}$), as said before, which is desorbed from the collected fine particulate matter during the

sampling and is then recovered on the back-up acid-coated filters, as already seen in previous studies (Perrino and Gherardi, 1999). This unbalanced amount of ammonia, ranged from 0,06 $\mu\text{mol}/\text{m}^3$ (1.15 $\mu\text{g}/\text{m}^3$) to 0.72 $\mu\text{mol}/\text{m}^3$ (13 $\mu\text{g}/\text{m}^3$) with an average value of about 0.26 $\mu\text{mol}/\text{m}^3$ (4.75 $\mu\text{g}/\text{m}^3$) during the summer sampling period. The value of 0 indicated the ammonia is in balance on back-up filters (Fig. 10). The unbalanced ammonia, clearly adsorbed on particles, can be a further source of atmospheric ammonia during specific meteorological conditions such as dry and warm and windy environments.

As in winter, to help interpret further in general terms the source regions affecting the sampled ammonia and, thus, to identify the origin and transport pathway of large-scale air masses, 24-h backward trajectories arriving at the sampling site were also calculated for five days of summer intensive measurements (Fig. 11). The trajectories were computed every 2 h (from 07:00 on 17 August 2007 to 05:00 on 21 August, local time). The Figure 11 shows dominant and regional transport of air masses from south (53%) and southeast (15%) of Beijing, which are the most frequent in summer time, coinciding with the direction of Hebei province and Tianjin municipalities, which are not only agricultural areas but also quite polluted by industrial, vehicular, coal mining and power generation, and biomass burning emissions (Xia et al., 2007; Street et al., 2007; Chen et al., 2009; Meng et al., 2009; Zhang 2010). In addition, these air masses passing the southern regions were relatively slow (1.63 – 3.85 m/s) and, thus, they had much time to accumulate ammonia in the Beijing area, contributing greatly to the air pollution. The urban area itself is a major source for traffic emissions. This suggests that the atmosphere of Beijing received transported polluted air on locally produced NH_3 .

Thus, the presence of local and regional sources during summer intensive measurements probably explains why the temperature didn't determine NH_3 concentrations and a diurnal cycle of this gas in Beijing was not observed due to variation in emissions.

These results reveal that during the summer NH_3 concentrations are influenced by meteorological conditions (wind direction, wind speed and, sometimes, temperature), atmospheric mixing, local and regional sources in the winter 2008 in Beijing. Further modeling studies are needed to quantify the contribution of local versus regional sources to the atmospheric ammonia variations.”

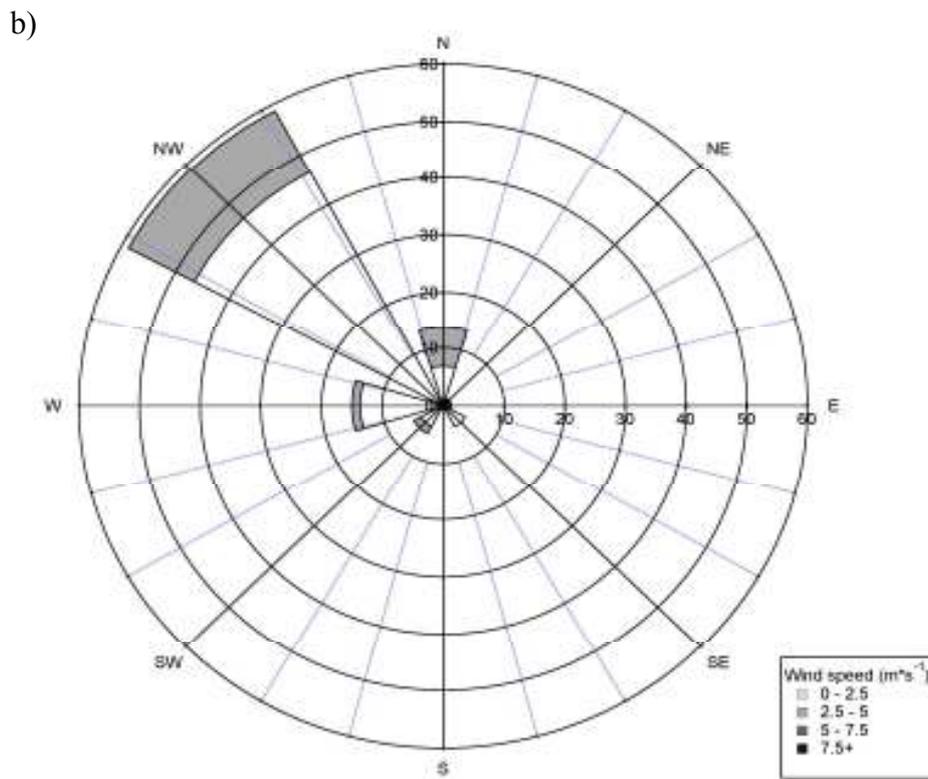
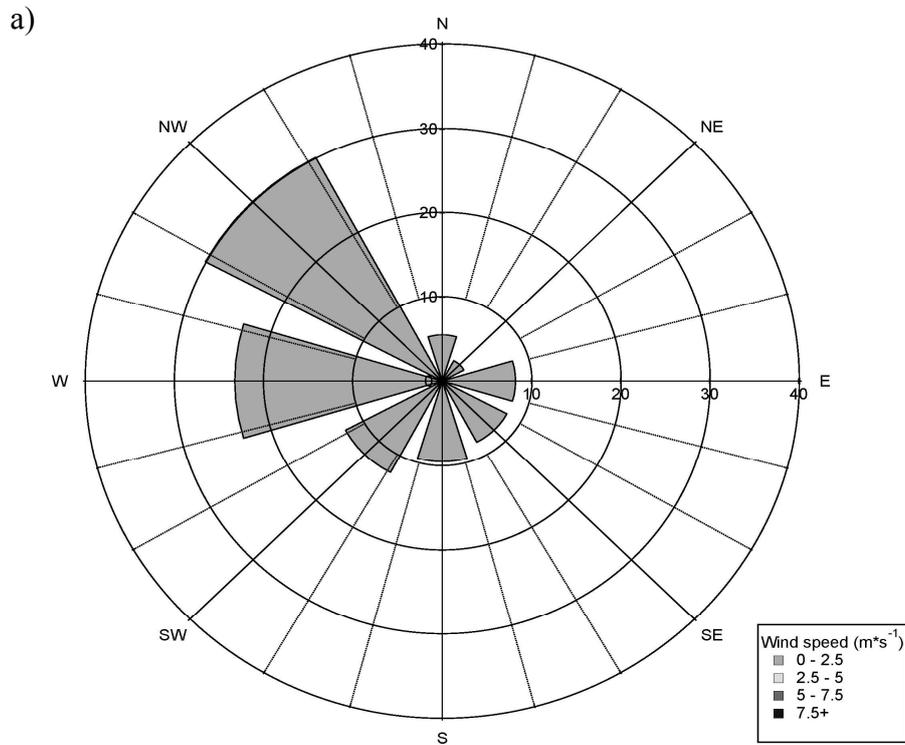


Fig. 2. Wind plots showing the frequency distributions of wind directions and speeds (grey scale) of the intensive winter measurements for two days: on 9 February (a) and 10 February (b). The radius axis represents the occurrence from 0% to 40% and from 0% to 60%, respectively.

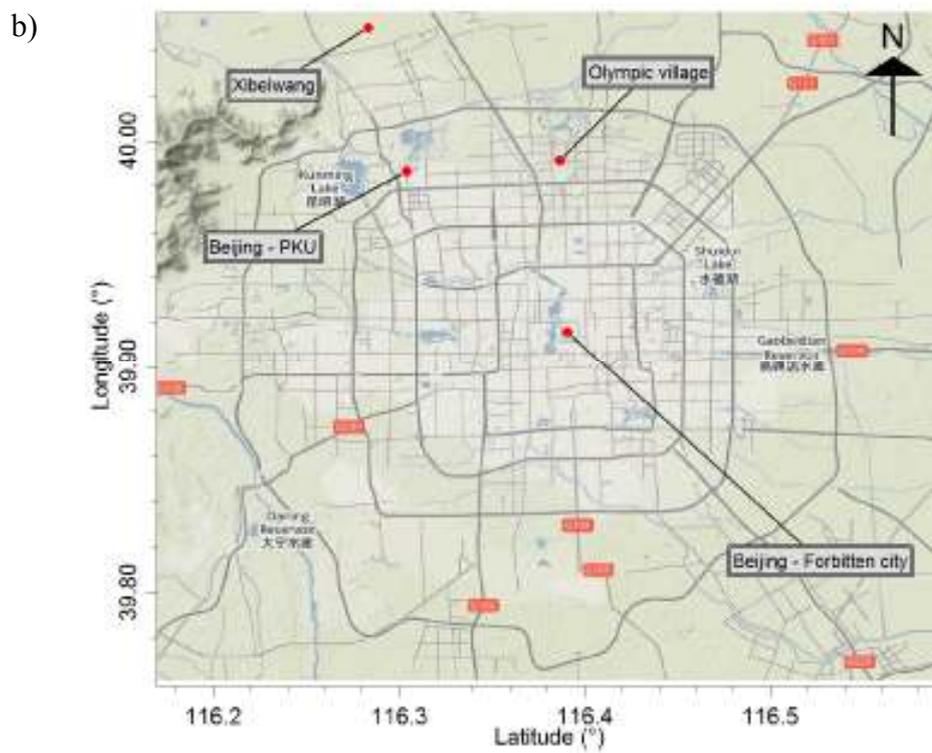
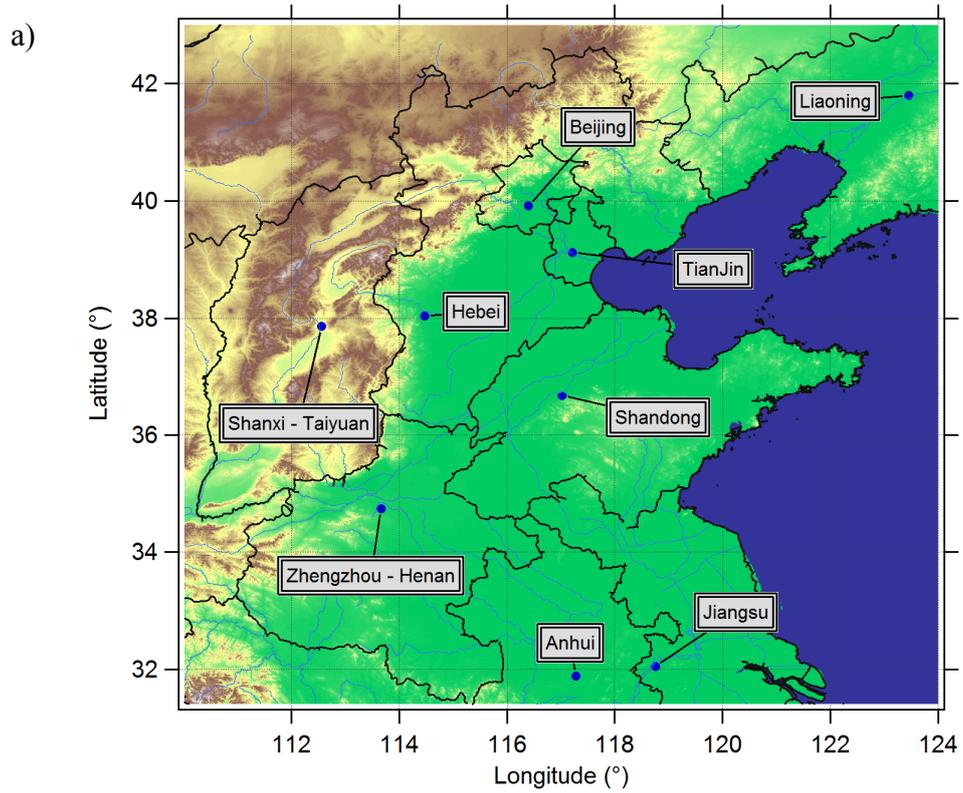


Fig. 3. Beijing city and measurement site (b) and surrounding regions (a).

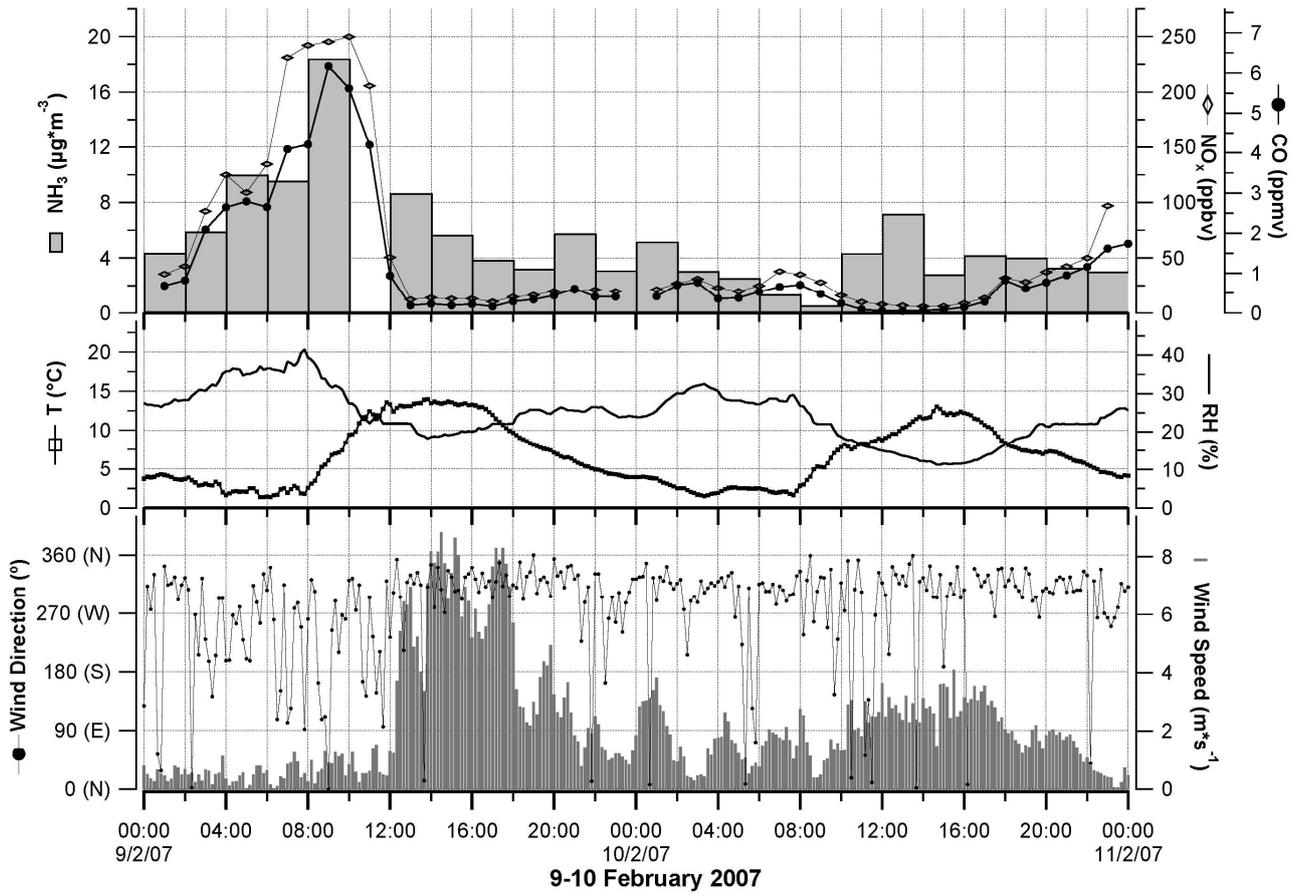


Fig. 4. Diurnal trends of NH₃, NO_x, CO, temperature (T), relative humidity (RH), wind speed and direction during the intensive winter measurements.

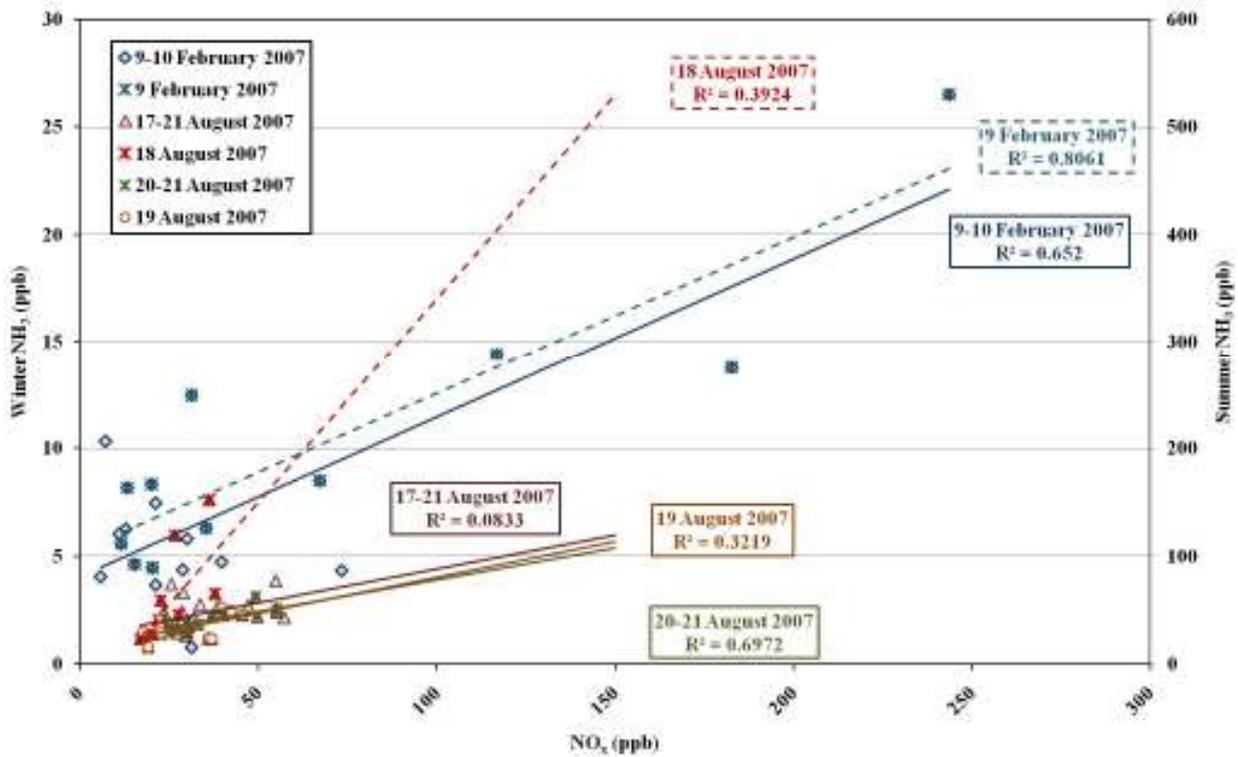


Fig. 5. Relationship between NH_3 and NO_x during the intensive winter and summer measurements.

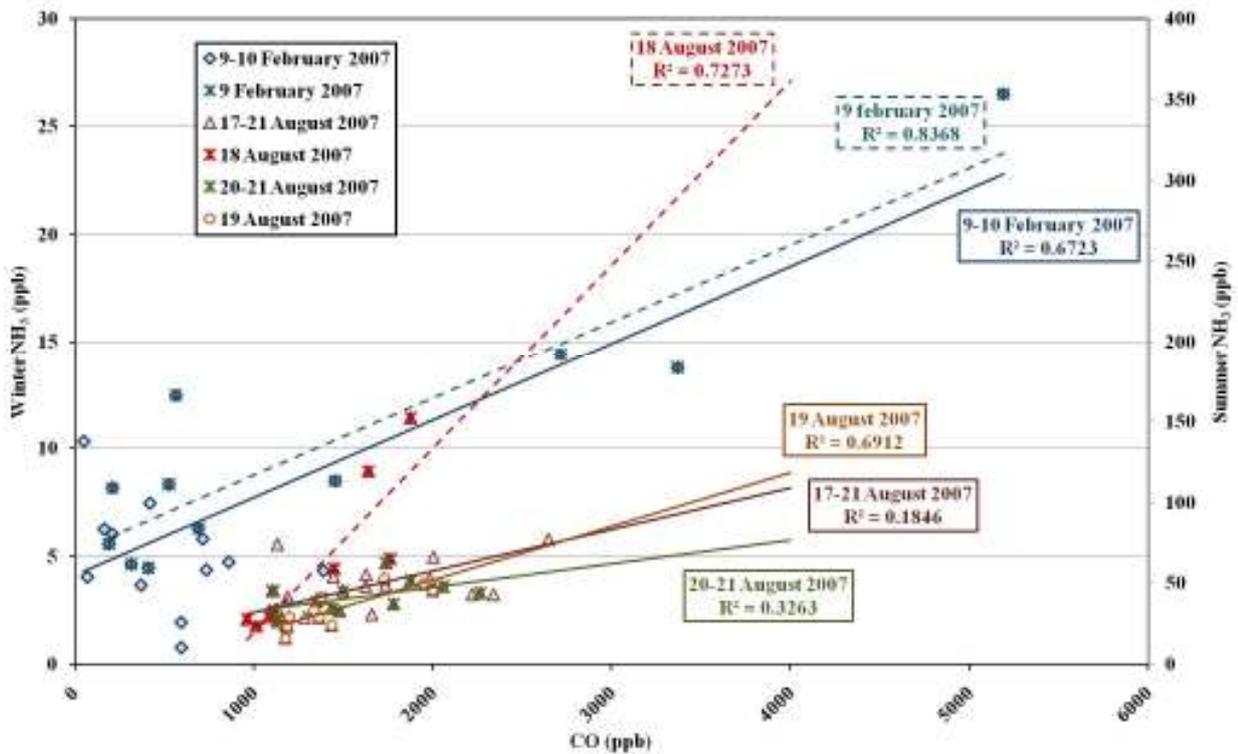


Fig. 6. Relationship between NH_3 and CO during the intensive winter and summer measurements.

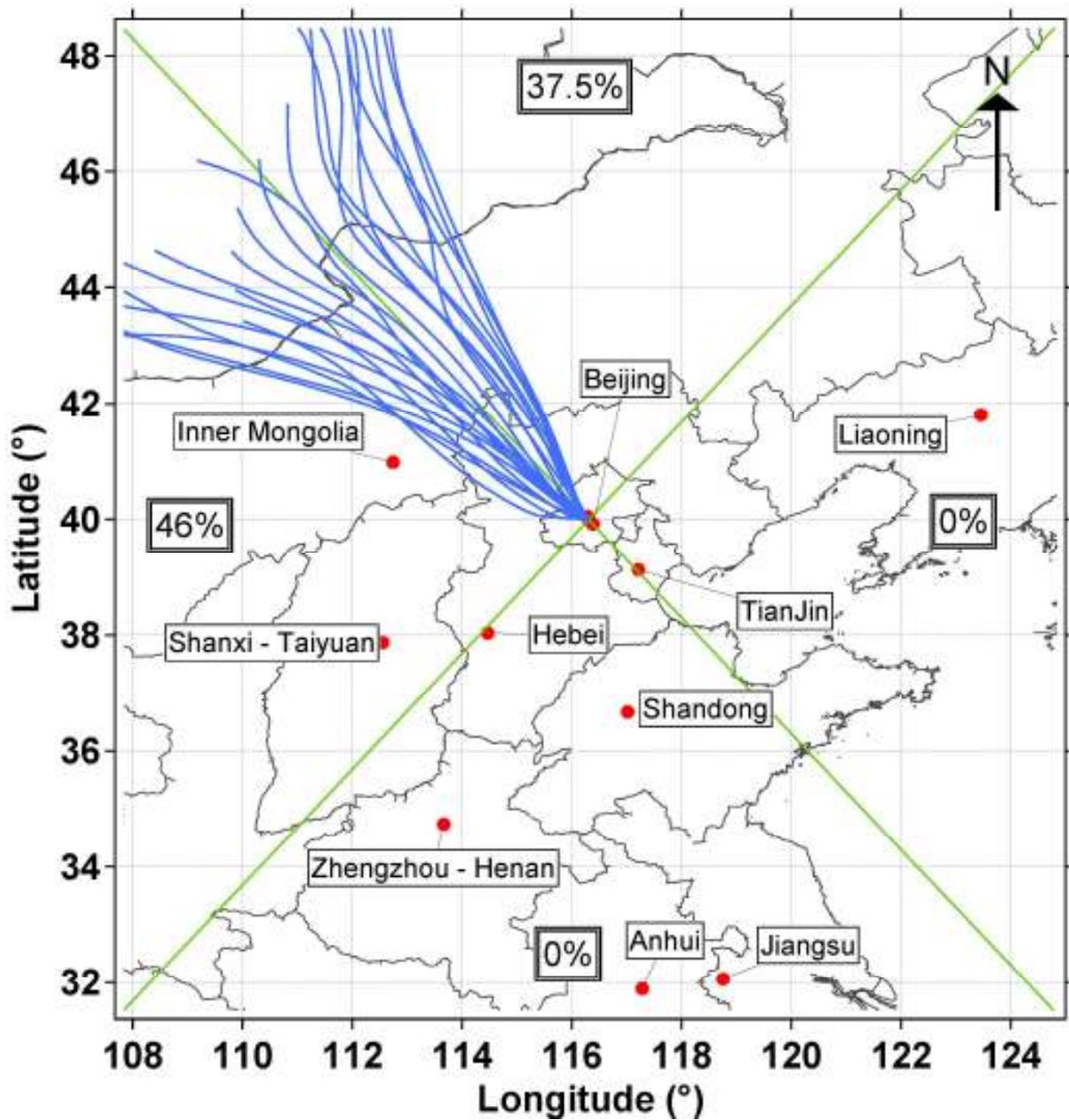


Fig. 7. Geographical map showing the 24-h backward trajectories arriving at Beijing during the intensive winter measurements, reconstructed using the NOAA ARL HYSPLIT trajectory model (<http://ready.arl.noaa.gov/HYSPLIT.php>).

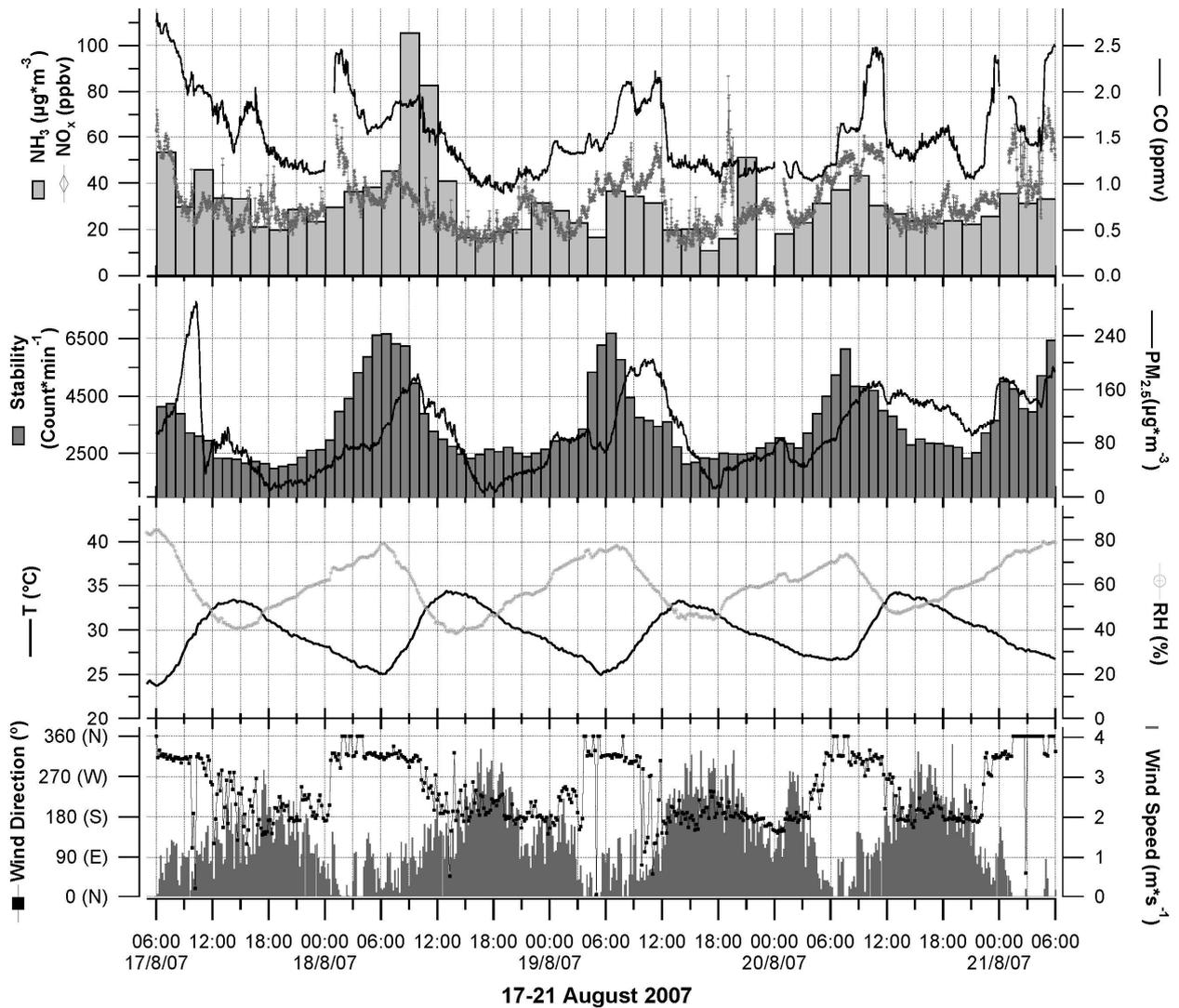


Fig. 8. Diurnal trends of NH_3 , NO_x , CO, $\text{PM}_{2.5}$, temperature (T), relative humidity (RH), wind speed and direction during the intensive summer measurements.

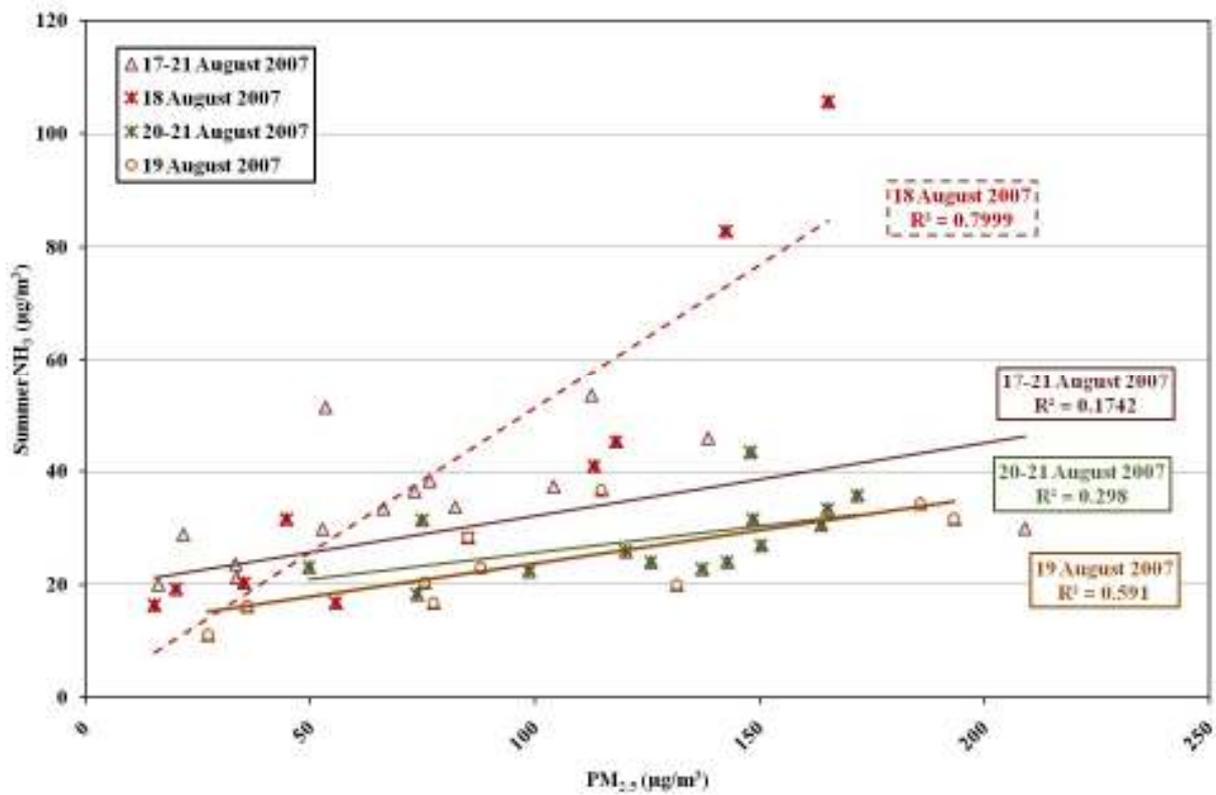


Fig. 9. Relationship between NH_3 and $\text{PM}_{2.5}$ during the intensive summer measurements.

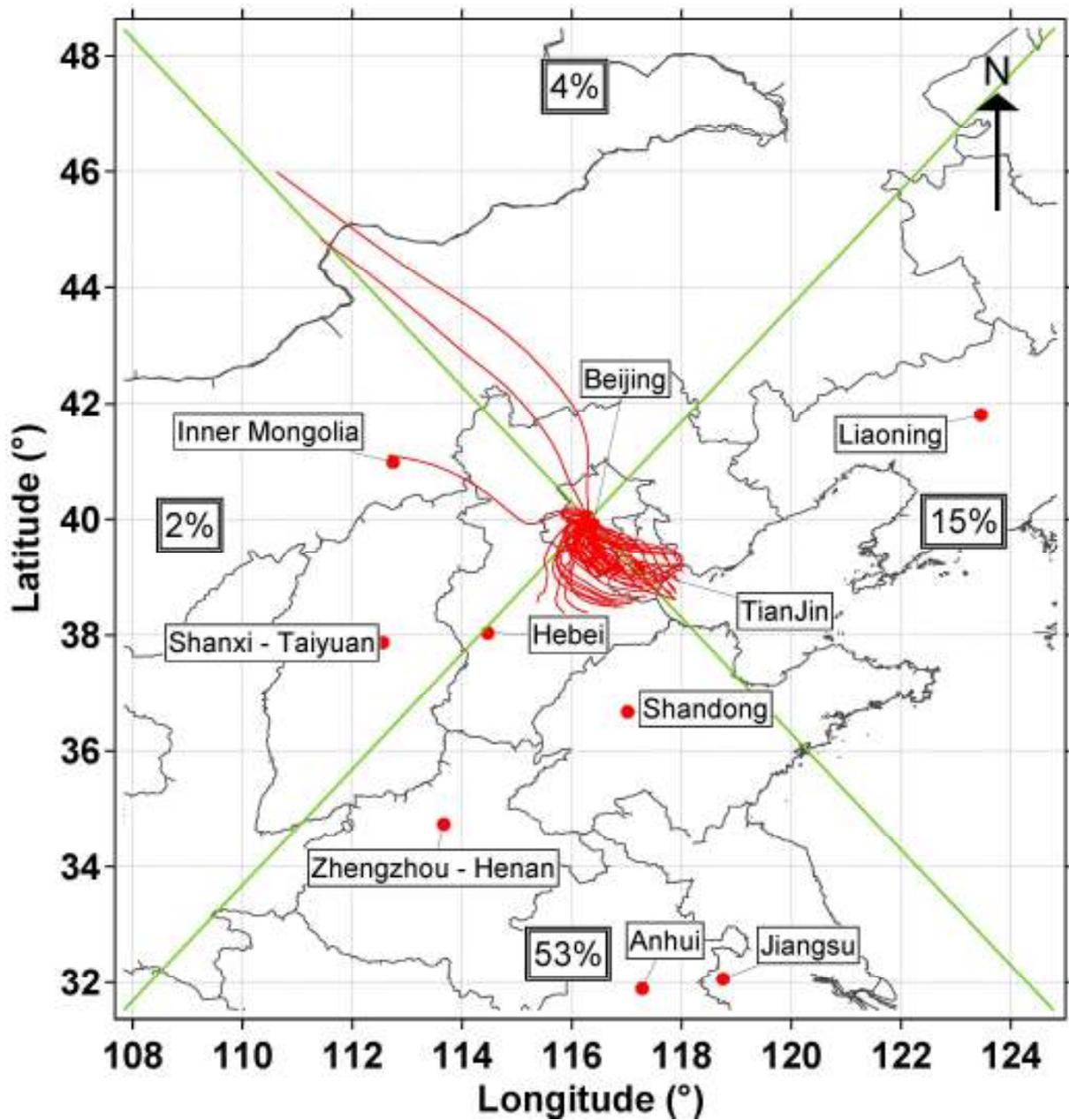


Fig. 11. Geographical map showing the 24-h backward trajectories arriving at Beijing during the intensive summer measurements, reconstructed using the NOAA ARL HYSPLIT trajectory model (<http://ready.arl.noaa.gov/HYSPLIT.php>).

3) The influence of different sources to observed levels is highlighted at several points in the paper and features as well in the conclusions (correlation with other trace gas emissions from transport). But while specific events during the campaigns are attributed to the influence of e.g. agricultural sources, depending on wind direction/speed, resp. road transport or other local sources (boundary layer conditions), even a short discussion of the spatial domain in which the measurements have been taken is missing. A map of the domain within which the measurements have taken place, locating the measurement site and – overlaying the wind roses presented in Figs. 2 and 5 – the location of potential strong sources of NH₃ (agricultural fields/farms, major roads/highways, power plants etc.) would be essential to underpin these assumptions now only based on peaks/events observed in the data.

Reply:

We followed your suggestion including two maps (Fig. 3a and 3b) which show Beijing city, measurement site and surrounding regions, where are the major NH₃ emission sources. Regions with high NH₃ concentrations are mainly located in eastern China. Actually, intensive agriculture is concentrated in the North China Plain which includes five provinces (Hebei, Henan, Shandong, Jiangsu and Anhui) and two municipalities (Beijing and Tianjin). Of the total agricultural ammonia emissions in the North China Plain, the Hebei, Henan and Shandong provinces take the larger part (Zhang et al., 2010). Contributions of NH₃ emissions from livestock and fertilizer activities were also found in Inner Mongolia (Klimont, 2001; Ju et al., 2004). Beijing is downwind of Shanxi, which is one of the largest coal mining and coal-fired power generation provinces in China, which can emit large amounts of NH₃ (Meng et al., 2010).

4) Analysing other trace gases to detect correlations between NH₃, NO_x and CO is a viable and sensible approach to allow for interpretations regarding source-attribution of observed events. The discussion of the results of these comparisons in this paper could be more detailed and thorough. In addition to that, literature quoted for nontransport sources (Sutton et al. 2000 for instance) is valid, but not the primary literature one would expect when looking at NH₃ emissions from for instance road transport sources. There are plenty of publications discussing NH₃ emission factors from power generation and mobile sources that has not been cited and seems not to have been reviewed (e.g. Heeb et al., 2007, doi:10.1016/j.atmosenv.2007.12.008, or COPERT, <http://lat.eng.auth.gr/copert/>). As an aside, the comparison with CO emissions may not be

straightforward at all, esp. with regard to difference in summer/winter (cold start emissions from vehicles, small combustion sources).

In addition to that, it does not become clear from the description how the authors account for the specific situation of the source composition in China, resp. Beijing. The technologies used in road transport vehicles (e.g. non-catalytic converter equipped, early simple and advanced catalytic converters) and in stationary sources (SCR/SNCR, primary measures etc.) will substantially affect the ratio of different pollutant emissions and hence the conclusions that can be drawn from correlations observed or not observed. A table with an (even coarse) emission inventory for China/Beijing with the relative contributions to each of the major source groups and how this would affect the expected concentrations to be observed would be needed to put the currently unfounded assumptions explaining observed events on a stronger footing. In this context, Figs 7 and 8 do not offer a substantial contribution to the discussions and could be omitted/combined.

Reply:

We followed your suggestion discussing more detailed the results about correlations between NH_3 , NO_x , and also CO and $\text{PM}_{2.5}$. Indeed, we added new data on the concentrations of CO in the winter and $\text{PM}_{2.5}$ in the summer, which support further our interpretations regarding NH_3 emission sources. During the winter $\text{PM}_{2.5}$ data were not good and significant because there were instrumental troubles.

We followed your suggestion adding new publications about NH_3 emissions from power generation and mobile sources such as Huai et al., 2003; 2005; Wang X. et al., 2005; Heeb et al., 2008, Reis et al., 2009). However, we already cited many publications in our paper (Zhao and Wang, 1994; Bouwman 1997; Olivier et al., 1998; Kean et al., 2000; Sutton et al., 2000; Kirchner et al., 2002; Battye et al, 2003; Streets 2003; Heeb et al., 2006) regarding these topics.

We followed your suggestion adding a coarse table which shows the mean emission factories of NH_3 for transport vehicles and agricultural activities believed to contribute mainly to the concentrations of NH_3 in Beijing. Hence, corresponding description will be added into the revised manuscript as follows:

“The Table 2 shows the mean emission factors of NH_3 for transport vehicles and agricultural activities believed to contribute mainly to the concentrations of NH_3 in Beijing. This investigation of emission inventory within Beijing was conducted with the base year being taken 2005, by using atmospheric NH_3 emission information by current literatures. There are not many published data available for the NH_3 emissions in China and, thus, only limited data are shown in the table.

Vehicle emission inventory for regional or national scale are usually developed using a macro-scale approach in China. Wang H. et al. (2009) and Wang H. et al. (2010) provided a bottom-up approach by combining vehicle emission factors and vehicle activity data from a travel demand model estimated at the grid level to generate vehicle emissions data for the Beijing urban area in 2005. Average vehicle activity data, such as Vehicle Kilometers Traveled (VKT), are estimated by investigation and/or a statistical method for each fleet. The emissions inventory is estimated as the product of emission factors and vehicle activity:

$$Q^P = \sum_{i=1}^m \sum_{j=1}^n EF_{i,j}^P VKT_{i,j}$$

where, Q^P is the total emissions for pollutant P, g; $EF_{i,j}^P$ is the emission factor of pollutant P for vehicle type i in grid cell j, g/km; $VKT_{i,j}$ is the vehicle kilometers traveled for vehicle type i in grid cell j, km.

For these applications, emission factors are assumed to represent long-term vehicle population averages for a given vehicle class, and are often based on default or average inputs. In this study, the vehicles driving in the urban area of Beijing were summarized into 6 classes: passenger car (PC), shuttle bus (SB), taxi, heavy duty truck (HDT), light duty truck (LDT) and bus. Because the light duty vehicles (PCs and taxis) are the main sources of traffic emissions in the city comprising over 90% of the total VKT in the urban area of Beijing, we considered only these vehicles for the NH_3 emission inventory.

Heeb et al. (2006) reported mean NO and NH_3 emission factors ($mg\ km^{-1}$) and average vehicle speed ($km\ h^{-1}$). In an urban area they calculated NH_3 emission factor of $48.8\ mg\ km^{-1}$ at $19\ km\ h^{-1}$. Taking into account that since 2008 the total number of vehicles registered in Beijing city had increased to about 3.3 million and is still increasing by more than 10% per year (Wang B. et al., 2010), we calculated the total NH_3 emission from traffic sources (Table 2).

Furthermore, Zhang et al. (2010) developed an agricultural NH_3 emission inventory in the North China Plain (NCP), calculating contributions of NH_3 emissions from different sources including Beijing and Tianjing municipalities. However, the largest sector contributor to NH_3 emissions in the NCP is agriculture (99%). Mineral fertilizer use contributed 54% to the total NH_3 emission in the NCP, while livestock sources contributed the remaining 46%. Thus, we also considered these contributions to agricultural NH_3 emission from Beijing and Taijing municipalities, and Hebei province.

Table 2. NH₃ emission estimates in Beijing and surrounding areas in 2005.

Provinces	Transport (Kt NH ₃ -N yr ⁻¹)	Agriculture (Kt NH ₃ -N yr ⁻¹)
Beijing	1.8	3.6
Taijing		87.6
Hebei		706.8

These results show that NH₃ emissions from agriculture are high as 3.6 Kt NH₃-N yr⁻¹ in Beijing in 2005 corresponding to two times that from traffic sources (1.8 Kt NH₃-N yr⁻¹). Thus, even if NH₃ emissions from road traffic mainly affect the air quality in urban environments as Beijing, however, the agricultural emissions can be also of major importance affecting substantially the observed NH₃ concentrations in Beijing, in combination with meteorological conditions and regional sources, as said above.

However, it is indeed very difficult to isolate the effects of local emission, regional sources and changes in meteorology. Our measurement data clearly illustrate the impact and importance of meteorology and regional sources on the observed NH₃ concentrations in Beijing.”

Specific comments

These 4 points raised above lead to the following specific questions that should be thoroughly addressed before publication is considered:

1) Are the 2 periods of measurements sufficient to conclude on the seasonality of NH₃ concentrations in Beijing? If not, what other evidence can be used to underpin this claim.

Response:

See the reply to point 1 in general comments.

However, in this paper the majority of cited references for ammonia measurements used two sampling periods (one winter and one summer period) to determine seasonal variations of NH₃ at Beijing because the seasonality of NH₃ emissions in China was already established.

Yet, if the references cited and discussed in this paper don't provide support for our measurements or don't demonstrate this seasonality for NH₃, we agree to change the sentence “NH₃ concentration show regular seasonal variations, having significantly higher summertime concentrations” in “NH₃ concentration show temporal variations, having significantly higher summertime concentrations”.

2) What is the underpinning evidence to conclude that no diurnal variation has been observed? Provide figures to either prove this claim, or reassess and discuss this in more detail.

Response:

See the reply to point 2 in general comments.

We followed your suggestions including a more detailed discussion of our results during the winter and summer periods.

We also modified the Figures 3 and 5 adding temperatures and other species such as CO and PM_{2.5}.

We also added two Figures () which show 24-h backward trajectories arriving at the sampling site for winter and summer intensive measurements to help interpret further in general terms the local and regional sources affecting the observed ammonia concentrations in Beijing.

3) Where are major sources (source groups) located in relation to the measurement site and with regard to wind direction/speed and distance from the source? Provide (a) map(s) which indicate source regions to underpin the claims regarding source attributions.

Response:

See the reply to point 3 in general comments.

We followed your suggestions including two maps (**Figures 1a and 1b**) which show Beijing city, measurement site and surrounding regions, where are the major NH₃ emission sources. We also calculated the back trajectories, as said above.

4) What are the relative contributions to the different sources of other trace gases used in comparison to NH₃ concentrations observed? Take a look at inventory data to underpin the source attribution and hence improve the interpretation of correlations observed/not observed with e.g. NO_x and CO.

Response:

See the reply to point 4 in general comments.

We followed your suggestions including a more detailed discussion on the correlations between NH₃ and NO_x, CO and PM_{2.5} during the winter and summer periods and deleting the Figures 4, 7

and 8. In fact, we made single Figures for NO_x and CO during winter and summer and we added a new Figure for PM_{2.5} during summer period.

In addition, we added new publications such as Heeb et al., 2008.

We tried to calculate a coarse emission inventory of NH₃ in Beijing and surrounding regions.

Technical corrections

The following technical corrections/points should be addressed:

- p.14211, 1.8/9: “is concerning with climate change” – rephrase/reword, not clear what is meant

Response:

We will replace the sentence (p.14211, 1.8/9) “is concerning with climate change.”, with “is linked to climate change based on its ability to form PM_{2.5}, specifically ammonium sulphates. These aerosols can possibly increase the earth’s albedo. Particles can either backscatter UV and visible radiation directly, reducing the amount that reaches the earth’s surface, or indirectly by increasing cloud cover due to increased numbers of cloud condensation nuclei, particles that give rise to cloud formation (Sutton et al., 2004)”.

- p.14211, 1.12/13: “causing consequences to remote sensitive ecosystems”, ambiguous, could be phrased better and more concrete

Response:

We will replace the sentence (p.14211, 1.12/13) “causing consequences to remote sensitive ecosystems” with “causing effects to sensitive ecosystems with consequent changes in plant and animal communities (Sutton et al., 1993; Fangmeier et al., 1994)”.

- p.14211, 1.24: “is different from developed countries”, in what sense is it different? The way it is phrased suggests that in China, the share of NH₃ is higher than in developed countries, while e.g. in Europe in 2005 (based on EMEP data), agriculture contributes more than 90% to total NH₃ emissions. Reconsider/rephrase.

Response:

We will replace the sentence (p.14212, 1.21/22) “The total NH₃ emission from China exceeds that from Europe by at least 27% (Zhao and Wang, 1994)” with “The total NH₃ emission from China are

estimated to be about two to three times higher than European and US emissions over the period of 1990 to 2005 (Klimont, 2001; Zhao and Wang, 1994; EMEP, 2009; NEI, US EPA, 2009, Reis et al., 2009)".

Thus, we will delete the sentence (p.14212, 1.23/24) "This is the distinguishing feature which is different from developed countries".

- p.14212, 1.6: add "emissions" after NH₃ and "insolation" should be "insulation"?

Response:

We will add "emissions" after NH₃ and we will replace "insolation" with "insulation".

- p.14212, 1.8/9: "But the interactions between these factors are so complex that no uniform result is forthcoming" – this sentence is hard to understand, what does it mean? In a scientific paper, one would expect a thorough discussion of parameters that are included in the analysis and those that are not, for obvious reasons; hence, this sentence should be omitted and instead replace with a short paragraph on what is within the scope of the analysis, and what beyond.

Response:

We will delete the sentence (p.14212, 1.8/9) "But the interactions between these factors are so complex that no uniform result is forthcoming".

- p.14212, 1.20: "control systems" better as "emission abatement technology"

Response:

We will replace "control systems" with "emission abatement technology".

- p.14213, 1.3: "winds" does it refer to "wind speed/direction?" please clarify

Response:

The word "winds" refers to wind speed and thus, we will replace "winds" with "wind speed".

- p.14214, 1.9: add "(PKU)" after "Peking University" as you use the abbreviation later, without introducing it

Response:

We will add "(PKU)" after "Peking University".

- p.14215, 1.5: Rephrase the start of this sentence to read "Although the focus of this paper is on ..."

Response:

We will replace the sentence (p.14215, 1.5) "Although this paper was focused on atmospheric ammonia," with "Although the focus of this paper is on atmospheric ammonia".

- p.14217, 1.8-10: How have day/night hours been grouped on 18:00-6:00/6:00-18:00, or on sunrise/sunset, as indicated in the next sentence? Unclear, please clarify

Response:

We will replace the sentence (p.14217, 1.8/10) "Data were grouped into day (between 06:00 and 18:00) and night (between 18:00 and 06:00) periods. Day and night hours were based on sunrise and sunset times within each sampling period." with "Data were grouped into sunrise (between 06:00 and 18:00) and sunset (between 18:00 and 06:00) times."

- p.14217, 1.20: "The source of NH₃..." which source is referred to? Explain and elaborate a bit more, the following sentence is making quite strong assumptions based on literature.

Response:

We will replace the sentence (p.14217, 1.20/21) "The source of NH₃ is at ground level, thus NH₃ concentrations might be generally lower at higher wind speeds because of turbulent diffusion" with "Since NH₃ is either readily converted to NH₄⁺ or subjected to dry deposition, high concentrations are found only close to the surface and near to emission sources (Ferm, 1998; Krupa, 2003). Thus, NH₃ concentrations might be generally lower at higher wind speeds because of turbulent diffusion."

- p.14219, 1.4/5: "solvent use"? The contribution of solvent use to total NH₃ emissions should be marginal (less than 2%), in addition, solvent use has not been mentioned in any discussion of NH₃ sources before, so how does it enter the picture here? Explain, or rephrase, please.

Response:

We will delete the words “solvent use” in all paper.

- p.14222, 1.17/18: the sentence starting “These results confirm ...” does not mention agricultural emissions at all, which is surprising for summer observations of NH₃. Explain why agricultural emissions are not among the parameters driving NH₃? In addition, the use of “evolution” here is ambiguous, should be replaced by a more concrete reference to e.g. variability, concentration levels etc.

Response:

See the reply to point 2 in general comments. In fact, we will replace the sentence (p.14222, 1.17/18) with “These results reveal that during the summer NH₃ concentrations are influenced by meteorological conditions (wind direction, wind speed and, sometimes, temperature), atmospheric mixing, local and regional sources in the winter 2008 in Beijing.”

Thus, we will delete the word “evolution”.

- p.14223, 1.2: “didn’t” should be replaced by “did not”

Response:

We will replace “didn’t” with “did not”.

- p.14234: Fig. 4 does not add much evidence/enlightenment to the paper, could be well combined with Figs. 7 and 8 into one set of 3 or 4 correlation plots in one figure.

Response:

We made a single Figure 4 (including the old Fig.4 and 8) for the correlations between NH₃ and NO_x during winter and summer periods and thus, we will delete the Figure 8.

We made a single Fig.5 (including the Fig.7) for the correlations between NH₃ and CO with Fig.7 during winter and summer periods and thus, we will delete the Figure 7.

In addition, it will be necessary to change the numeration of all Figures in the paper.

- p.14236: Fig. 6. is quite cluttered and at its current size makes is hard to read or distinguish the relevant details for the reader. If not possible to increase the size, consider splitting

Response:

We will increase the size of Fig.6.

- p. 14237/8: Figs. 7 and 8 could be omitted/combined with Fig. 4 (see above)

See above.

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