Pollution plumes observed by aircraft over North China during the IPAC-NC field campaign

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Received January 15, 2013; accepted May 21, 2013; published online July 16, 2013

Strong pollution emissions from large urban and industrial centres in eastern China are thought to greatly impact regional air quality and global atmospheric chemistry. In this study, we investigate extensive pollution plumes over North China, observed by aircraft during the IPAC-NC (Influence of Pollution on Aerosols and Cloud Microphysics in North China) field campaign in spring 2006. Several pollution plumes were encountered, characterized by sharp and strong increases of SO₂ and NO_x (NO plus NO₂) as well as remarkable O₃ variability. During a flight performed downwind of the megacity Tianjin on 9 April, O₃ was found to be depleted at NO_x>15 ppbv and enhanced at NO_x mixing ratios of 6–7 ppbv. Our analysis indicates that the plumes had recently been released by industrial plants, and partly mixed with urban emissions, giving rise to the large-scale transport of air pollution and substantial degradation of air quality.

pollutant emissions, urban/industrial plumes, photochemical smog, tropospheric ozone, aircraft measurements

Citation: Ma J Z, Wang W, Liu H J, et al. Pollution plumes observed by aircraft over North China during the IPAC-NC field campaign. *Chin Sci Bull*, 2013, 58: 4329–4336, doi: 10.1007/s11434-013-5978-9

Pollutants freshly emitted from major urban and industrial centres, including e.g. power plants and petrochemical facilities, tend to undergo substantial chemical evolution during plume transport [1,2]. The chemical characteristics of pollution plumes and their variation as a function of transport distance are diverse, depending on the type and strength of emission sources, chemical features of the background atmosphere and meteorological conditions. Understanding the intricate physical, chemical and dynamical processes occurring in pollution plumes is essential to assess the influence of pollution outflow from megacities on air quality and climate from local to global scales [3–6]. Aircraft measurements can be very helpful in providing three-dimensional spatial and temporal insights into the transport and chemical evolution of pollution plumes [7–11]. The O₃ variability, in combination with other reactive species in the plumes, has

been a main focus of many studies that investigated the non-linear response of ozone to its precursors and its influence on tropospheric chemistry. For example, pollution plumes are typically found to shift from being O₃-depleted to O₃-enriched during transport, and differences of a factor of 2 or more in ozone formation rates and yields as a function of NO_x ($\equiv$ NO+NO₂) and volatile organic compound (VOC) concentrations were often observed in U.S. power plant plumes [1].

North China (namely Huabei in Chinese Pinyin), located between 32°–42°N in eastern China, encompasses two megacities (Beijing and Tianjin) and several major industrial centres (e.g. Tangshan, Shijiazhuang and Taiyuan cities), which therefore represents a main pollution hot spot in China [12]. Satellite observations have shown an increasing trend in tropospheric NO₂ and O₃ over North China in the past decade [13–16]. Outflow of the emissions from North China, together with that of other severely polluted regions

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of eastern China, may have large impacts on background levels of pollutants over the Pacific Rim region, North America and even Europe [17–20]. Therefore, it is important to study the chemical characteristics of pollution plumes freshly emitted from urban and industrial centres in North China, not only to support the control of regional air pollution, but also for the understanding of tropospheric chemistry in the entire northern hemisphere. Recent studies based on surface measurements have shown that pollutants emitted from these urban and industrial centres can greatly impact air quality in the region [21–26]. However, measurements of pollution plumes aloft over North China have not yet been reported; though the levels and vertical distributions of air pollutants over this region were reported previously [27–30].

Here we present the results of comprehensive aircraft observations of pollution plumes over North China during the IPAC-NC (Influence of Pollution on Aerosols and Cloud Microphysics in North China) campaign in spring 2006. The IPAC-NC campaign was the first intensive field experiment that included simultaneous measurements of atmospheric trace gases, aerosols and clouds by aircraft over this part of China. The most important findings of IPAC-NC include the following. (1) Widespread haze clouds, characterized by very high concentrations of gaseous pollutants and haze particles mixed with low clouds, were observed, and it has been demonstrated that these haze-clouds exert a strong cooling forcing on regional climate [31]. (2)

Enhanced levels of OH and HO₂ radicals were shown to be associated with the very high pollution concentrations, and it was indicated that the lower atmosphere over North China acts as a pollution and oxidation pool over China [29]. In this study, we focus on pollution plumes observed during the IPAC-NC campaign. We investigate some of the high levels and the variability of the reactive gases SO₂, NO, NO_x and O₃ as well as CO and non-methane hydrocarbons (NMHCs) in pollution plumes encountered over North China. Potential sources and aging processes of the plumes and implications for regional chemistry are discussed.

1 IPAC-NC field campaign

The IPAC-NC field campaign was carried out between 2 April and 16 May, 2006, with a twin-engine YUN-12 aircraft operated from Tianjin International Airport. During the campaign, aircraft measurements were performed in the larger Beijing, Tianjin and Tangshan area with some flight tracks extending towards the Bohai Gulf. Figure 1 presents the spatial distributions of emission sources of the primary air pollutants SO₂, NO_x, CO and NMHCs from central North China, based on an emission inventory developed by our group [32]. The major emission centers coincide with megacities (Beijing 39.92°N, 116.46°E and Tianjin 39.02°N, 117.02°E) and other large industrial centers (Tangshan

Figure 1 Annual emission rates of the primary pollutants (a) SO₂, (b) NO_x, (c) CO and (d) NMHCs in the central North China. Strong emissions from Beijing (BJ), Tianjin (TJ), Tangshan (TS) and Shijiazhuang (SJZ) cities are clearly visible.

39.36°N, 118.11°E, Shijiazhuang 38.02°N, 114.30°E). The strong air pollution emissions, plus the converging wind flow pattern, tend to result in the accumulation of pollutants in the lower atmosphere over the region [29].

Instruments for chemistry measurements were mounted on board the aircraft to detect the trace gases SO₂, NO, NO_x, O₃ and CO [29,31,33]. A FEP-Teflon tube was used to introduce the ambient air from below the fuselage into the aircraft cabin for gas analysis. SO₂ was monitored by a UV pulse fluorescence analyzer (TECO Model 43C-TL, USA) with a response time of about 80 s. Nitrogen oxides were monitored by a chemiluminescence NO-NO_x analyzer (TECO Model 42C-TL, USA). The analyzer measures the chemiluminescence of NO₂ formed by the reaction between NO and O₃, of which the intensity is proportional to the NO concentration. NO_x measurements are approximated using the thermal reduction of NO₂ to NO by a heated (320°C) molybdenum converter. Although it is expected that the signal of NO₂ was dominant, other reactive nitrogen (NO_y) compounds may be converted to NO by this system so that the NO₂ data presented here should be considered as an upper limit. The 10 s duty cycle of the TE42C-TL alternates between NO and NO_x measurements. The difference between the two measurements allows the quasi-continuous signals of NO and NO_x. Data were produced at 1 Hz, although the readings of NO and NO_x were only updated every 10 s. O₃ was measured by a UV-absorption analyzer (TECO Model 49, USA) with a response time of 20 s. CO was monitored by a gas filter correlation analyzer (TECO Model 48C, USA) with a response time of about 60 s. Since the CO analyzer on board the aircraft needed a relatively long period for stabilization during each flight, only part of the CO data could be used for analyses. NMHC sampling was done using 2–4 canisters per flight and each canister was typically filled in 10–20 min.

Seventeen research flights were performed during the IPAC-NC campaign [29]. Flight patterns were designed and implemented based on a combination of research objectives, weather conditions, and air traffic control regulations. The flight altitude ranged up to 3500 m, the cruising velocity was generally ~200 km h⁻¹, and the flight duration was typically around 4–5 h. Detailed descriptions of the IPAC-NC campaign, including the instruments, aircraft flight information and weather conditions, can be found in [29].

2 Results and discussion

2.1 General features of the plumes

Several pollution plumes, characterized by a sharp increase in mixing ratios of the primary pollutants SO₂ and NO_x as well as remarkable O₃ variability (enhanced or depleted) during extended flight tracks (>10 km), were encountered during IPAC-NC. The geographical positions of typical plumes, as observed on 9 April, 15 April and 7 May of 2006,

are depicted in Figure 2. Also shown in Figure 2 are the wind fields and spatial tracer distributions over the region concurrent with the plume observations, which were simulated by the regional meteorological forecast model GRAPES with passive tracers added [29,34,35]. To show the regional transport of urban plumes, the tracers were released in the lowest layer of the model from the locations of the five major cities Beijing, Tianjin, Tangshan, Shijiazhuang and Taiyuan [29].

According to our model results, the plumes observed on 9 April had been directly influenced by the emissions from the Tianjin megacity. However, the plumes on 15 April and 7 May were not predicted by our urban tracer calculations, indicating that they had originated from other pollution sources than the cities included. Detailed features of the plumes observed by aircraft on these days are discussed below.

On 9 April, the airplane took off from Tianjin Airport at 01:54 UTC (09:54 Beijing Time), flew towards the north, and reached Huangzhuang Village (39.48°N, 117.52°E) near northern Tianjin at about 02:18 UTC. Over Huangzhuang it performed several consecutive circle flights from the top downward until 05:35 UTC, with a radius of 15 km at the altitudes of 2.9, 2.5, 2.1, 1.7, 1.2, 0.8, and 0.4 km, respectively. Aircraft measurements as well as satellite observations showed that widespread warm stratiform clouds prevailed over much of North China during the flight, covering an altitude range of about 750–1200 m [31]. Figure 3(a) shows the vertical profiles of SO₂, NO, NO_x, and O₃ measured during the circle flights on 9 April. Two pollution plumes were encountered, one above the clouds at an altitude of 1.7 km, and another below the clouds at 0.4 km.

On 15 April, when the weather was clear, a flight was performed at night, from 11:51 UTC to 14:43 UTC. The airplane flew across the Bohai Gulf, cruising from Gujuzi (38.02°N, 118.37°E) to Haitiancun (39.28°N, 119.03°E) at a constant altitude of 2 km and returning back to Gujuzi at a constant altitude of 1 km. A pollution plume was observed when the airplane was approaching Gujuzi. On 7 May, also a clear day, the airplane took off from Tianjin Airport at 04:45 UTC, flew towards the east, and reached the border area between Tianjin Municipality and Hebei Province (39.08°N, 118.07°E, south of Tangshan City) at about 05:20 UTC. There, it performed sequential vertical cross section flights from the flight top downward until 06:33 UTC, with a horizontal width of about 60 km at altitudes of 2.6, 2.1, 1.6, 1.1, and 0.6 km. As shown in Figure 3(b), a plume was observed at an altitude of 0.6 km. It appears that the vertical extent of the plumes on these days was limited to less than ~500 m depth.

2.2 SO₂ and NO_x in the plumes

Figure 4 shows the changes in gaseous pollutant concentrations as a function of flight time in the plumes observed on

Figure 2 Geographical locations (red lines) of the plumes observed during flights and coincident wind fields and spatial distributions of pollution tracers emitted from major urban centers of North China, as simulated by the model for (a) 850 hPa on 9 April, 2006; (b) 925 hPa on 9 April, 2006; (c) 925 hPa on 15 April, 2006; (d) 925 hPa on 7 May, 2006. Mountainous areas are masked (white).

Figure 3 Vertical profiles of trace gases around the time when the plumes were observed on 9 April, 2006 (a) and 7 May, 2006 (b).

Figure 4 Changes in mixing ratios of SO_2 , NO , NO_x , and O_3 as a function of flight time in the plumes, as observed at (a) 1.7 km altitude on 9 April, 2006, (b) 0.4 km altitude on 9 April, 2006, (c) 1.0 km altitude on 15 April, 2006 and (d) 0.6 km altitude on 7 May, 2006. Also shown are NMHC mixing ratios and the compound categories as well as the peak CO mixing ratios (available only for cases (b) and (d)).

9 April, 15 April and 7 May, 2006. Remarkably higher primary gaseous pollutants were observed persistently for flight times of 10–20 min (33–66 km along the flight track), characterizing the extent of the plumes. For example, the mixing ratio of SO_2 reached up to 41–54 ppbv in the plume

cores, 10–20 ppbv higher than outside the plumes. The mixing ratio of NO_x in the plumes was up to 7–27 ppbv, about 2–10 ppbv higher than outside the plumes. CO data were missing for some plumes due to instrumental limitations. Nevertheless, a peak value of 2.4 ppmv for CO was observed

in the 0.4 km altitude plume of 9 April, 0.8–1.0 ppmv higher than in the adjacent air masses.

The concentrations of primary pollutants in a plume are affected by various processes, including dispersion, mixing and chemical reactions. Dispersion is expected to have the same impact on all pollutants in a plume. The effects of mixing processes depend on the concentrations of pollutants in the plume relative to those in the ambient atmosphere as well as in other plumes. The influence of chemical reactions is typically different for the various species in the plumes. Therefore, the relative changes of the mixing ratios of primary pollutants with different chemical lifetimes can be used as an indicator of the plume aging process. The lifetimes of SO₂ and NO_x against gas-phase oxidation by OH in the planetary boundary layer of North China are estimated to be about a day and 4 h, respectively, based on our box model simulations constrained by measurements during IPAC-NC [29]. Therefore, it can be expected that with constant emissions the SO₂/NO_x ratio increases with plume age, assuming that no mixing occurred.

Table 1 shows the molar ratios of the primary pollutants measured in the plume cores by our aircraft, as well as those emitted from various pollution sources, according to the emission inventory for North China [32]. The SO₂/NO_x ratios were measured to be ~2.0 ppbv/ppbv in the 0.4 km-altitude plume of 9 April and ~1.8 ppbv/ppbv in the 0.6 km-altitude plume of 7 May, comparable to the emission ratios for power generation and industrial activities; they were ~3.2 ppbv/ppbv in the 1.0 km-altitude night-time plume of 15 April and ~7.3 ppbv/ppbv in the 1.7 km altitude plume of 9 April, remarkably larger than the emission ratios for power generation and industrial activities. The SO₂/NO_x ratios measured in the plumes were much higher than the emission ratios for traffic and biomass burning. This indicates that the observed plumes had recently originated from power gener-

ation and/or industrial plants, though the influence of emissions from traffic and biomass burning (especially NO_x) on the plumes cannot be excluded. Zhou et al. [11] showed that within a limited range of plume ages (0.3–1.3 h), the SO₂/NO_x ratios measured in the Texas power plant plumes (which varied between 1.2 and 11 ppbv/ppbv) were predominantly determined by the emission ratios of the corresponding plants (which varied between 1.1 and 8.9 ppbv/ppbv). The plumes that we observed most likely originated from different plants, with SO₂/NO_x emission ratios smaller or larger than the average emission ratios shown in Table 1. Therefore, the ages of these plumes cannot be inferred exactly by merely using measured SO₂/NO_x ratios in a plume without detailed information of individual emission sources.

2.3 O₃ in the plumes

The variability of the secondary photochemical pollutant O₃ was large in the different plumes, acting either divergent from (Figure 4(b)) or in parallel (Figure 4(a), (c) and (d)) to that of its precursors. For example, the O₃ mixing ratio was depleted by ~5 ppbv in the 0.4 km altitude plume of 9 April at NO_x>15 ppbv (NO>2 ppbv) (Figure 4(b)), but was enhanced by up to ~20 ppbv in the 1.7 km altitude plume on the same day at NO_x mixing ratios of 6–7 ppbv (NO≈0.5 ppbv) (Figure 4(a)). An increase of O₃ by up to ~10 ppbv was also found in the night-time plume of 15 April, at an NO_x level of 10–12 ppbv (NO≈1–2 ppbv) (Figure 4(c)). Note that CO as well as total NMHCs and their composition contributions also varied greatly from one plume to another. Ozone production is often nonlinear with respect to its precursors [36], being confirmed by numerous field experiments including aircraft measurements of urban and industrial plumes [1,37–39]. Ryerson et al. [1] reported that at 36 km downwind of a power plant in the United States, O₃ concentrations were enhanced at NO_x<10 ppbv toward the edge of the plume, whereas O₃ was still suppressed through titration by NO at NO_x>15 ppbv within the plume core. During IPAC-NC, the O₃ mixing ratio was enhanced by up to ~20 ppbv in the 0.6 km altitude plume of 7 May at NO_x>20 ppbv (Figure 4(d)), in contrast to the cases above. This can be explained by the addition of freshly emitted NO_x to an aged plume. In such a plume, ozone formation probably occurred for some time and the accumulated O₃ can then be titrated by freshly released NO. As a result, the observed decrease of O₃ is not significant and the level of NO was not very high (<2 ppbv) in the plume. It is interesting to see that for the daytime observations in IPAC-NC, ozone mixing ratios in the plumes could reach 60–80 ppbv in the O₃-enhancement cases (Figure 4(a) and (d)), much higher than the 21–25 ppbv in the O₃-depletion case (Figure 4(b)). Such differences between the pollution plumes can significantly influence the O₃ distributions at regional scales.

O₃ in the plumes can be changed by two types of chemical mechanisms, the titration by NO, which does not change

Table 1 Molar ratios of the primary pollutants SO₂ and NO_x, based on an emission inventory for North China [32] and aircraft measurements in the plume cores during the IPAC-NC field campaign

	SO ₂ /NO _x (ppbv/ppbv)
Emissions	
Power generation	1.6
Industrial activities	2.0
Traffic	0.1
Biomass burning	0.3
Beijing megacity	0.3
Tianjin megacity	1.0
Plumes	
9 April 2006 (1.7 km altitude)	7.3
9 April 2006 (0.4 km altitude)	2.0
15 April 2006 (1.0 km altitude)	3.2
7 May 2006 (0.6 km altitude)	1.8

the amount of total oxidant O_x , and the net production (destruction) of O_3 , which results in an increase (decrease) of O_x . We calculated O_x using measured O_3 , NO and NO_x assuming $[NO_2]=[NO_x]-[NO]$, though detected NO_x might probably include some other NO_y compounds [29]. It turns out that O_x concentrations are higher within the plumes than outside in all cases (not shown in the figure) and they covary with NO_x in the O_3 depletion cases (Figure 4(b)) while the correlation is much less in the O_3 enhancement cases (Figure 4(a), (c) and (d)). Higher plume O_x in the O_3 depletion case is accompanied with enhanced NO_2 resulting from the titration of O_3 by NO within the plume; however, it cannot be quantitatively determined to what extent photochemical source and sink processes played a role due to limited measurement data.

For comparison, net O_3 destruction was observed in an urban plume (from Manaus) over the heart of Amazon Basin as reported by Kuhn et al. [39]. Under high NO_2 conditions in the plumes, depletion of OH radicals, needed for NMHC oxidation, may occur, resulting in the decreased availability of peroxy radicals for O_3 production. Higher O_x in the O_3 enhancement case indicates net photochemical O_3 production in the plume during the sampling period or earlier. In North China there are many large industrial complexes with power plants, steel and iron smelters, cement factories and chemical plants, i.e. upwind of the area of investigation [32]. The mix of gaseous pollutants, especially NMHCs, emitted from these various industrial sectors can be very diverse, as indicated by the fractional contributions of alkanes, alkenes and aromatics to the total NMHCs measured in the plumes, with sometimes very high mixing ratios in excess of 12 ppbv (Figure 4). Plumes from these pollution sources could be merged and mixed into air masses or plumes transported from urban areas such as Tianjin, which hitherto photochemically aged with enhanced O_3 concentrations. Therefore, the plumes encountered by our instrumented aircraft during IPAC-NC represent a wide diversity of chemical characteristics.

3 Conclusions

Several pollution plumes were observed by aircraft over North China during the IPAC-NC field campaign in spring 2006. The spatial scales of these plumes were typically limited to less than 500 m depth and 30–60 km width. While we typically found sharp increases in SO_2 and NO_x , remarkable O_3 variability (either relative increases or decreases) were also observed in the plumes. The mixing ratios of SO_2 reached up to ~50 ppbv in the plumes, with increases of ~20 ppbv relative to the plume environmental air. The peak mixing ratio of NO_x in the plumes was observed to be nearly 30 ppbv, about 10 ppbv higher than outside the plumes. For a similar vertical profile downwind of the megacity Tianjin on 9 April, O_3 was found to be depleted at

$NO_x > 15$ ppbv in the 0.4 km altitude plume and enhanced at NO_x mixing ratios of 6–7 ppbv in the 1.7 km altitude plume. While the decrease of O_3 in the plume can at least partly be attributed to the titration of O_3 by NO, the enhancement of O_3 in the plumes indicates net photochemical production of O_3 , with O_3 mixing ratios in the plumes reaching 60–80 ppbv. The molar ratios of SO_2 to NO_x measured in the plumes were typically ~2.0 ppbv/ppbv, indicating that they had recently been emitted from power generation or industrial plants. Plumes from different pollution sources can merge and mix with plumes transported from urban areas. Further source identification and chemical characterization of the plumes in eastern China needs to be performed in dedicated studies with an extended suite of instrumentation.

This work was supported by the National Natural Science Foundation of China (40433008), the National Basic Research Program of China (2011CB706903), and the China Special Fund for Meteorological Research in the Public Interest (GYHY201206015). The research leading to these results has also received funding from the European Research Council under the European Union's Seventh Framework Programme (FP7/2007-2013)/ERC grant agreement no. 226144. We are grateful to Xu Mingzhi and Yin Baohui of CRAES and Wang Xiaobin of CAMS for their assistance with the aircraft measurements.

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