

Interactive comment on “Aircraft measurements of gases pollutants and particles during CAREBeijing-2008: distributions, characteristics and influencing factors” by W. Zhang et al.

W. Zhang

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Dear Dr. Prof. Baumgardner,

Dr. Baumgardner has brought out many good points that were not adequately addressed in the manuscript. We appreciate it very much for your useful comment and kind help on improving English in the version. These points will be addressed in the order in which they appear in your review and a revised manuscript will include all of the responses. In the new version, the dispersion forecast results were also considered during the grouping for all the flights, and the box and whisker plots, and the independent t-test results for gas and particle pollutants were also added. Correspondingly, the manuscript results and discussion were almost re-written following the new results. We have corrected and added the tables, figures and new text following your suggestions (see below).

All the best,

On behalf of all the authors,

Wenjie Zhang

Answers to specific comments:

Comment 1

The first problem, of course, is that it was not thoroughly edited by someone with a better grasp of English. Normally this would not cause me problems, but there are too many errors in general to allow easy reading and this is an obstacle that should not be placed immediately in front of the reviewers.

Thank you very much for it, and you have really helped us for the English editing, as well as the revision suggestions on some small errors. The detailed

information are as follows.

Comment 2

The second problem is one of how the results are presented. A lot of numbers are presented in the text and in tables but the reader is forced to try to understand these numbers with no easy way to put them into context. I will make a few suggestions below, but in general only the most important numbers should be discussed in the text with everything else in readable Tables (Table 2 is not. Either it needs to be rotated and enlarged or split into several tables with notable values highlighted). All the flights should be given designations that are in Table 1 but that are easier to refer to than dates. For example all flights in a line could be referred to as flights I-1, I-2: : I-9; II-1..II-3, for example. The dates are only relevant when talking about the back trajectories and meteorology.

Following your suggestions, the new tables have done with the flights I-1, I-2: : I-9; II-1..II-3 and so on, and only the back trajectories and dispersion results were shown as dates.

The new table 1 and table 2 are as following.

Table 1 The flight time, range, pattern, area and related information for different flight line

Line	Flight	Date	Time	Altitude Range	Pattern	Flight Area
L1	I-1	2008/8/27	10:35~14:25	2100~900~600	Linear,	ZJ(N39°17',
	I-2	2008/9/2	14:13~17:53	2100~900~600	back	E117°27')~AC(N39°31',
	I-3	2008/9/3	9:41~13:33	2100~900~600	and	E116°42')
	I-4	2008/9/11	9:09~13:04	2100~900~600	forth	
	I-5	2008/9/12	8:50~12:43	2100~900~600		
	I-6	2008/9/25	14:00~18:04	2100~900~600		
	I-7	2008/9/27	9:07~12:16	900~600		
	I-8	2008/10/11	9:15~13:20	2100~900~600		
	I-9		13:46~17:40	2100~900~600		
	I-10	2008/10/13	12:50~16:42	2100~900~600		
L2	II-1	2008/8/29	9:04~12:54	2100~900~600	Linear,	AC(N39°31',
	II-2	2008/9/1	8:44~12:37	2100~900~600	back	E116°42')~ZZ(N39°29',
	II-3	2008/9/20	10:06~13:20	2100~900	and	E115°58')
	II-4	2008/9/21	9:09~14:30	2100~900~600	forth	
L3	III-1	2008/8/28	8:57~13:06	2100~600	Linear,	ZJ~AC~ZZ~BD(N38°52',
	III-2	2008/9/1	15:04~18:25	2100~600	back	E115°28')~SJ(N38°3',
	III-3	2008/9/8	13:55~17:27	900~600	and	E114°31')
	III-4	2008/9/15	13:25~17:18	2100~600	forth	

L1 Total	10 flights, 2283 min (38 h), 3 heights
L2 Total	4 flights, 970 min (16.2 h), 3 heights
L3 Total	4 flights, 887 min (14.8 h), 3 heights

Table 2 The average concentration of gases pollutants, condensable nuclei and PM_{0.5} at different heights of each flight (The unit of CN, PM_{0.5} are N/cm³, CO is ppmV, and other gases are ppbV)

Height	Flight	Group 1										Group 2										Group 3			Group 4			
		I-1	II-1	II-2	III-1	III-2	I-2	I-3	I-6	I-8	I-9	II-4	I-4	I-5	I-7	II-3	I-10	III-3	III-4									
2100	NO ₂	16.36	9.42	8.93	11.36	6.09	4.42	3.88	2.20	1.20	2.54	4.60	8.37	6.44	-	4.63	2.43	-	7.68									
	NO	0.52	0.35	0.35	0.42	0.39	0.37	0.44	0.39	0.35	0.44	0.43	0.39	0.41	-	0.47	0.76	-	0.44									
	NOx	16.88	9.77	9.28	11.78	6.48	4.79	4.31	2.59	1.55	2.98	4.88	8.76	6.84	-	5.10	3.20	-	8.12									
	SO ₂	4.85	6.73	0.21	2.50	0.34	0.38	0.13	0.42	0.69	0.03	1.05	0.34	0.33	-	2.72	3.86	-	1.22									
	O ₃	58.64	66.85	46.60	58.87	49.36	51.82	47.15	39.66	43.39	45.47	51.63	42.15	44.14	-	59.00	55.25	-	53.65									
	CO	0.12	0.05	-	0.13	-	0.33	-	0.70	0.52	0.15	0.33	0.53	0.24	-	0.46	0.83	-	0.35									
	CN	3144	-	629	-	8507	8518	-	1112	-	-	734	1381	-	-	3763	3900	-	8496									
	PM _{0.5}	14691	4141	2692	3621	5381	5073	-	2877	1435	1849	3481	2527	1999	-	6574	5771	-	5561									
	NO ₂	18.54	10.81	8.47	11.48	-	9.62	12.42	4.04	0.63	5.86	13.07	16.18	13.01	3.12	6.67	6.62	11.08	-									
	NO	1.49	0.56	0.47	0.45	-	0.46	1.28	0.54	1.95	1.23	0.84	0.48	0.65	0.45	0.47	0.50	0.58	-									
NOx	20.03	11.37	8.73	11.93	-	10.08	13.70	4.58	2.58	7.10	13.91	16.66	13.66	3.56	7.14	7.12	11.65	-										
SO ₂	11.10	14.54	0.83	7.64	-	3.37	17.93	0.65	0.62	0.99	9.62	1.19	1.33	0.54	4.82	7.41	8.62	-										
O ₃	67.03	73.59	44.88	63.69	-	69.31	88.26	32.89	35.61	39.44	69.77	48.69	46.69	34.76	67.77	74.73	74.80	-										
CO	0.48	0.09	-	0.06	-	0.18	0.35	0.49	0.37	0.14	0.56	0.30	0.26	0.43	0.65	0.53	0.76	-										
CN	8288	-	2778	-	-	16353	11508	20770	8918	5941	9384	14880	8916	964	1805	9193	15141	-										
PM _{0.5}	12123	10663	11277	7100	-	16114	-	23321	9654	10671	-	12963	8695	6267	4936	12080	18609	-										
600	NO ₂	22.15	20.56	14.30	17.33	12.17	14.17	15.61	5.02	9.31	10.35	7.83	17.06	16.91	8.68	17.49	16.06	18.92										
	NO	1.13	0.78	1.58	0.74	0.56	0.45	0.90	0.39	3.68	0.74	0.48	0.51	0.49	3.27	-	0.63	0.80	0.82									
	NOx	23.28	21.34	15.88	18.07	12.73	14.62	16.51	5.41	12.99	11.09	8.31	17.58	17.40	11.96	-	18.12	16.86	19.74									
	SO ₂	11.68	11.83	2.29	12.88	3.56	5.19	21.99	0.78	2.14	1.75	7.18	2.49	1.05	13.60	-	23.11	7.68	20.30									
	O ₃	73.25	82.77	49.98	68.80	60.59	81.13	95.65	32.19	34.81	41.72	66.97	55.16	48.91	29.21	-	102.69	67.48	104.68									
	CO	0.61	0.19	-	0.62	-	0.11	0.52	0.38	0.42	0.18	0.63	0.21	0.15	0.89	-	1.15	0.90	1.00									
	CN	8829	-	6936	-	6453	13034	12280	13547	35725	10954	5402	25835	7662	16439	-	17016	15508	20945									
	PM _{0.5}	14653	14356	17764	12805	13642	20147	-	17127	29343	12925	10883	15924	10183	70722	-	19824	17466	30670									

Correspondingly, the text parts have been revised in the new version especially

on section 3.2, as following.

The flights that correspond to the four groups are: 1) Flight I-1 (FI-1), FII-1, FII-2, FIII-1 and FIII-2 for group 1 (G1), 2) FI-2, FI-3, FI-6, FI-8, FI-9 and FII-4 for group 2 (G2), 3) FI-4, FI-5, FI-7 and FII-3 for group 3(G3), and 4) FI-10, FIII-3 and FIII-4 for group 4(G4).

The other parts in the text have been revised for the flights and groups.

Comment 3

The third problem is that there is no analysis in this paper. Results are presented with no discussion at all that justify the conjectures. For example, at the end of presenting results for each of the four cases of air coming from various directions, statements are made like “The gases pollutants showed some remarkable features, particularly in the air with high concentration of SO₂ and O₃, as shown in Table 2. This may be due to the stagnation with low wind speeds and highly active photochemistry, the urban emissions of both primary compounds and precursors for secondary ions lead to an additional pollutant on top of the already elevated regional level”. This is completely hypothetical and no attempt is made to support this conjecture. Every section has statements like this that are completely irrelevant with no solid analysis that backs them up.

Thanks you very much for your nice suggestions, and we have tried to revise it, and have deleted some comments without proofs. After having added the statistical t-test results and some references, the text above has been revised as “The gases pollutants showed significantly higher concentrations between G1 and other groups, particularly in the air with high concentration of SO₂ and O₃, as shown in Table 2. This may be due to air stagnation under conditions of low wind speeds and highly active photochemistry; the urban emissions of both primary compounds and precursors for secondary ions lead to an additional pollutant on top of the already elevated regional level, as other researcher reported (Van Pinxteren et al., 2009; Streets et al., 2007). This may be shown from the higher concentration of particles, especially for PM_{0.5} in G1.”

However, only the gases and number concentration of particles were made

during the flights. It is very difficult to make too deep analysis for the changes and variations during the transport, and get the exact proofs for that. In addition, there seems not too much aircraft measurements especially not so many field campaigns during Beijing around areas in China have been made before, it is the most interesting results of this work. Considering this, the averages of the gas and particle pollutants for the same group were also statistically analyzed to discuss, and changes and variations were discussed and compared too. Too detailed information for each flight have not been included too much, and other researches for detailed flight will be discussed specially.

Comment 4

The fourth problem is that virtually none of the aircraft data is shown graphically in a way that highlights the important features. A lot is said about the gases but only CO is shown graphically and that is not very informative. Time series along the flight track provide a useful start for the analysis.

Considering the wide information and many flights of this work, it is impossible and not necessary to give too many details for each flight. Thus only the averages of the gas and particle pollutants for each group were made in Figure 2, and the average information for each flight of different groups were listed in Table 2 in the new version. In addition, there seems not too much aircraft measurements especially not so many field campaigns during Beijing around areas in China have been made before, it is too difficult to make too deep discussion on variations without the chemical species information. Considering this, the same group were also statistically analyzed to discuss, and changes and variations were discussed and compared in Table 3.

As only CO is relatively longer lived and less reactive, it showed different characteristics with other reactive gases, NO_x, SO₂, and O₃, the detailed information for CO were specially discussed in Figure 7. For other gas and particle pollutants, other researches for special flight will be discussed elsewhere.

The new Figure 2 and Table 3 are as following.

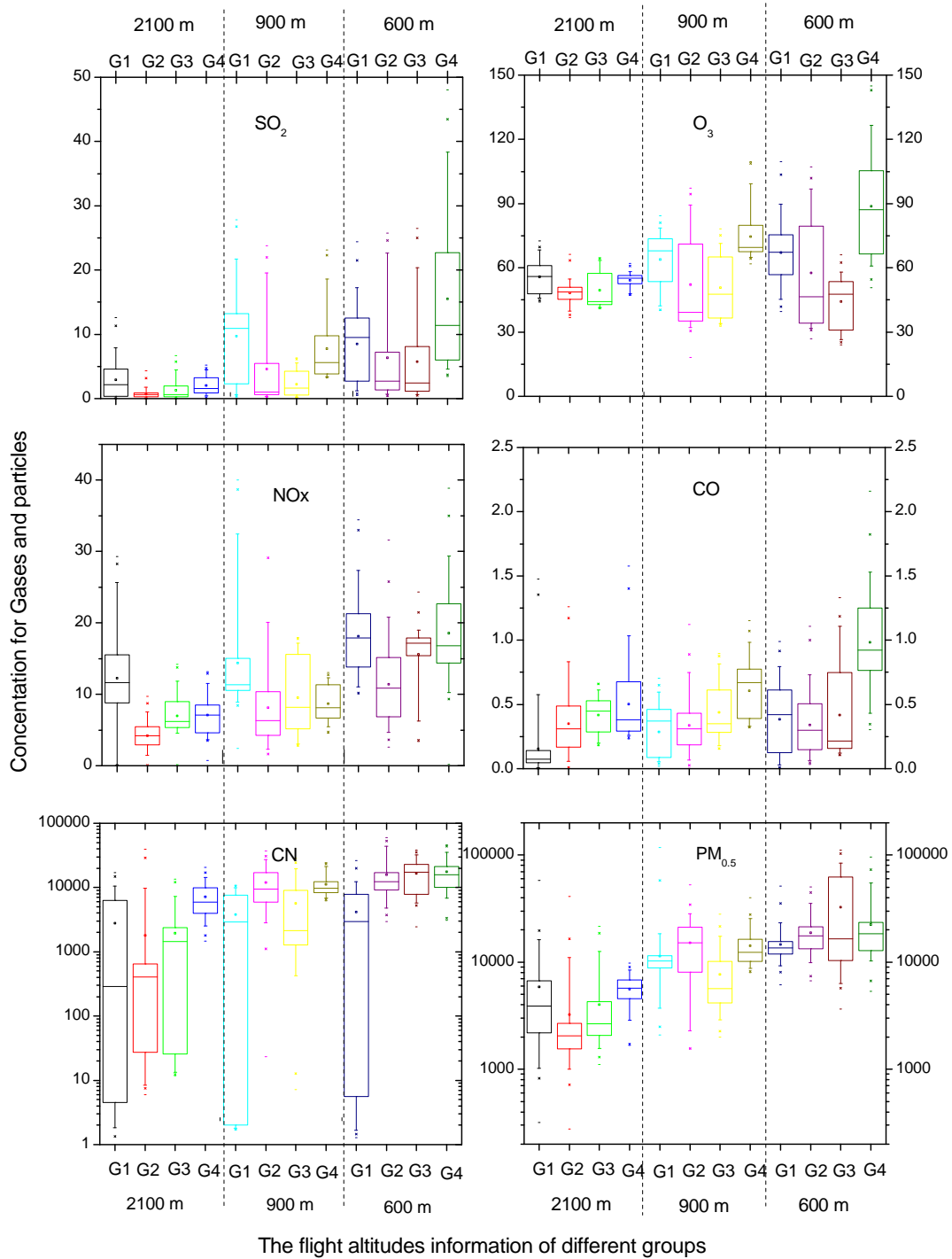


Fig. 2 The box and whisker plots for gases pollutants, condensable nuclei and PM_{0.5} at different altitudes of each group (The unit of CN, PM_{0.5} are N/cm³, CO is ppmV, and other gases are ppbV)

Comment 5

General suggestions: Summarize all the statistics in bar charts that compare average, standard deviations and maximums. Maybe put these on the map of Figure 1, or stratify by cases.

Select the most interesting features and analyze with respect to the physics of what is actually happening, backed by solid evidence, not just conjecture.

I started to do a detailed, point by point review, but realized that I was going to have a review longer than the current manuscript.

In the end, all I can do is summarize as I have the deficiencies of this paper and offer the authors my assistance, if they wish to make use of it, to tidy up and improve upon this potentially useful paper.

Thank you so much for this, and we have done the new Figure 2 on the statistics in bar and whisker plots, as shown above. Also, we have revised the new version following your detailed, point by point review suggestions. We really appreciate your kind help very much for all.

As most of the parts were re-written, only the big revisions and question comments on your detailed revision suggestions and responses are listed as following.

Page 1, we have revised the topic as “Airborne measurements of gases gas and particle pollutants and particles during CAREBeijing-2008”

Page 2, Following your suggestions, the new abstract has been revised and re-written as follows. “Measurements of gaseous pollutants, including ozone (O₃), sulfur dioxide (SO₂), nitrogen oxides (NO_x = NO + NO₂), carbon monoxide (CO), particle number concentrations (5.6-560 nm and 0.47-30 μm), and meteorological parameters (T, RH, P) were conducted during the Program of Campaigns of Air Quality Research in Beijing and Surrounding Region (CAREBeijing) from August 27 through October 13, 2008. The data from a total 18 flights (70 h flight time) from near the surface to 2100 m were obtained with a Yun-12 aircraft in the southern surrounded areas of Beijing (38°N-40°N, 114°E-118°E). The objective of these measurements were to characterize the regional variation of air pollution during and after the Olympics of 2008, the importance of air mass trajectories and evaluation of other factors that influence the

pollution characteristics. The results suggest that there are primarily four distinct sources that influenced the magnitude and properties of the pollutants in the measured region based on back trajectory analysis: (1) southerly transport of air masses from regions with high pollutant emissions, (2) northerly and northeasterly transport of less pollutant air from further away, (3) easterly transport from maritime sources where emissions of gaseous pollutant are less than the south but still high in particle concentrations and (4) the transport of air that is a mixture from different regions, i.e. the air at all altitudes measured by the aircraft was not all from the same sources. The relatively long-lived CO concentration is shown to be a possible transport tracer of long-range transport from the northwesterly direction, especially at the higher altitudes. Three factors influenced the size distribution of particles, i.e. air mass transport direction, ground source emissions and meteorological influences were also discussed.”

Page 3, Line 1-2, *Need extensive references here*

Following your suggestions, we have added the references in the text, and revised it as “Many studies have been conducted to study the air pollution in Beijing and surrounding areas (Dickerson et al., 2007; Garland et al., 2009; Guinot et al., 2007; Huang et al., 2010; Jung et al., 2009; Streets et al., 2007)”.

Related references are as follows.

Dickerson, R.R., Li, C., Li, Z., Marufu, L.T., Stehr, J.W., McClure, B., Krotkov, N., Chen, H., Wang, P., Xia, X., Ban, X., Gong, F., Yuan, J., Yang, J., Aircraft observations of dust and pollutants over northeast China: Insight into the meteorological mechanisms of transport, *Journal of Geophysical Research*, 112, D24S90, doi:10.1029/2007JD008999, 2007.

Garland, R. M., Schmid, O., Nowak, A., Achtert, P., Wiedensohler, A., Gunthe, S. S., Takegawa, N., Kita, K., Kondo, Y., Hu, M., Shao, M., Zeng, L. M., Zhu, T., Andreae, M. O., Pöschl, U., Aerosol optical properties observed during Campaign of Air Quality Research in Beijing 2006 (CAREBeijing-2006): Characteristic differences between the inflow and outflow of Beijing city air, *Journal of Geophysical Research*, 114, D00G04, doi:10.1029/2008JD010780, 2009.

Guinot, B., Cachier, H., Sciare, J., Tong, Y., Xin, W., Jianhua, Y., Beijing aerosol: Atmospheric interactions and new trends, *Journal of Geophysical Research*, 112, D14314, doi:10.1029/2006JD008195, 2007.

Huang, X. F., He, L. Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L. W., Liu, X. G., Zhang, Y. H., Jayne, J. T., Ng, N. L., Worsnop, D. R., Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer, *Atmospheric Chemistry and Physics*, 10, 8933–8945, doi:10.5194/acp-10-8933-2010, 2010.

Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Hu, M., Sugimoto, N., Optical properties of atmospheric aerosols obtained by in situ and remote measurements during 2006 Campaign of Air Quality Research in Beijing (CAREBeijing-2006), *Journal of Geophysical Research*, 114, D00G02, doi:10.1029/2008JD010337, 2009.

Streets, D.G., Fu, J.S., Jang, C.J., Hao, J., He, K., Tang, X., Zhang, Y., Wang, Z., Li, Z., Zhang, Q., Wang, L., Wang, B., Yu, C., Air quality during the 2008 Beijing Olympic Games, *Atmospheric Environment*, 41, 480-492, 2007.

Page 3, Line 3, *What does this mean? All sources are local but when they are taken together they form a regional problem*

Thank you very much, and we have revised it, and have added the related references, as “While, the air pollution in Beijing is a regional problem due to different sources mixed together from local and surrounding areas (Garland et al., 2009; Jung et al., 2009; Matsui et al., 2009)”

Page 3, Line 16-30, Following your suggestions, this parts have been rewritten and revised as “Aircraft measurements provide the means to study the vertical structure of the pollution over large horizontal distance and relatively short time scales. An airborne measurement platforms allows rapid deployment to multiple areas of interest and the vertical mobility provides insight into boundary layer dynamics, vertical layering of pollutants, vertical stability and provides measurements of a more statistically relevant area [Taubman et al., 2006]. A number of aircraft field campaigns have been carried out in or downwind of China. The Asia Pacific Regional Aerosol

Characterization Experiment (ACE-Asia, Kawamura et al., 2003; Huebert et al., 2004; Simoneit et al., 2004) made aircraft measurements over the Yellow and East China Seas, an outflow region pollutants from China, as well as the spatial and vertical distributions of pollutants over coastal and inland China (Wang, et al., 2007). An international project, the Atmospheric Brown Clouds-East Asian Regional Experiment (ABC, Wang et al., 2005, 2008a), has been conducted in China since the early 1990s, and many domestic projects supported by the Chinese science foundation have also collaborated in many aircraft field campaigns.”

Page 5, Line 18, *Explain how you selected the three altitudes. What are there significance? What about L3? Were the same three altitudes flown?*

As the Yun-12 aircraft is non-pressurized, 2100 m is the proper higher altitude considering the pressure and temperature variation for the instruments, while 900 m and 600m is considered for the larger changes and variations below 1000m. Thus the three different altitudes, 2100m, 900m and 600m were selected in this research. It is the same for the L1~3, except that only two altitudes of them were conducted in L3 due to the limit of the maximum distance for each flight.

Thus the new text were added after Line 18, as “The three altitudes were selected based upon the average depth of the boundary layer in the morning and afternoon. The 2100 m flight level puts the aircraft in the free troposphere but below the maximum altitude where the flight crew would require oxygen masks (the aircraft is unpressurized) and also at a low enough pressure altitude such that the instruments would function properly. The flight levels at 600 m and 900 m placed the aircraft at two different altitudes, well within the mixed layer where more than 90% of the pollutants are contained. Comparison of the 600 and 900 m concentrations allowed an estimate of the rate of mixing and dilution between the mixed layer and free troposphere.”

Page 8, Line 4-8, *Why not use data from the airports or if you are downloading data for the gases from the government and EPA, don't they also record meteorology? What about surface wind data? Aren't these also important?*

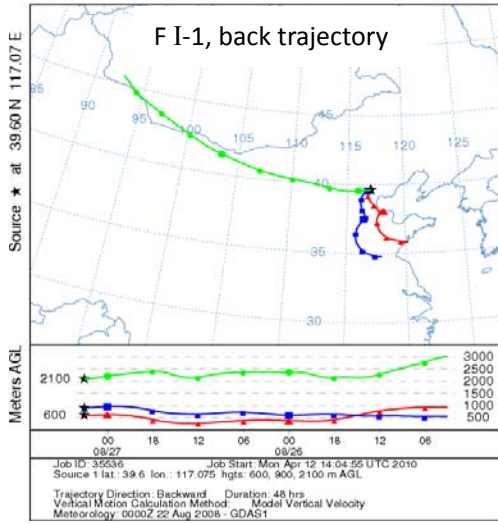
As it is convenient for me to download the meteorological data from the website,

it is nowhere for me to download it in China.

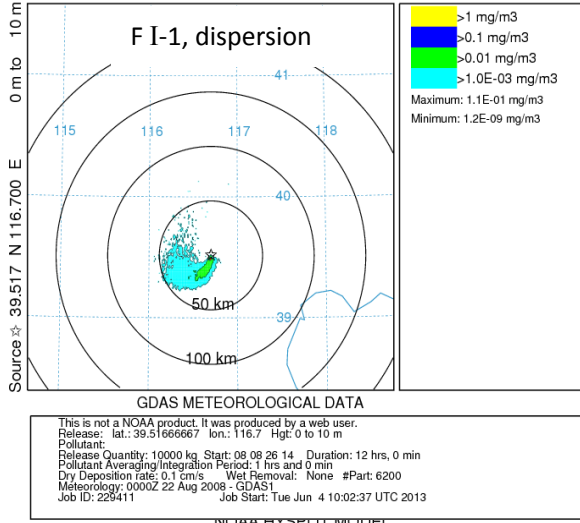
Page 8, Line 14, As the dispersion model results were also discussed and added, the description of them are added here, as “In addition, the forecast dispersions of both the gases and particles are calculated by the dispersion model of HYSPLIT (Draxler and Rolph, 2013, <http://ready.arl.noaa.gov/HYSPLIT.php>) assuming either puff or particle dispersion. In the puff model, puffs expand until they exceed the size of the meteorological grid cell (either horizontally or vertically) and then split into several new puffs, each with it's share of the pollutant mass. In the particle model, a fixed number of particles are advected through the model domain by the mean wind field and spread by a turbulent component. The model's default configuration assumes a 3-dimensional particle distribution (horizontal and vertical) (<http://ready.arl.noaa.gov/HYSPLIT.php>).”

The new figures 3-6 have added the results from dispersion model results, in addition to the back trajectory results. The following Figure 3 are listed as an representatives.

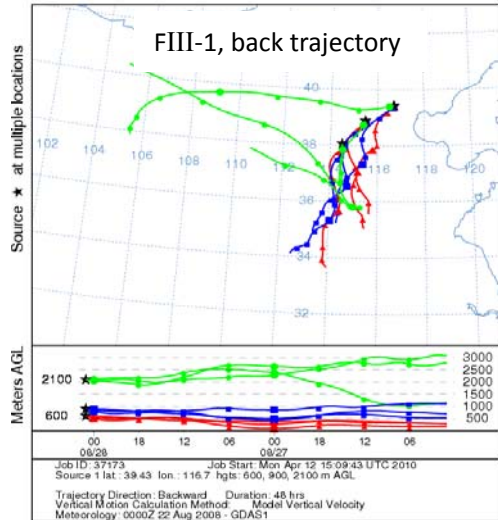
NOAA HYSPLIT MODEL
Backward trajectories ending at 0300 UTC 27 Aug 08
GDAS Meteorological Data



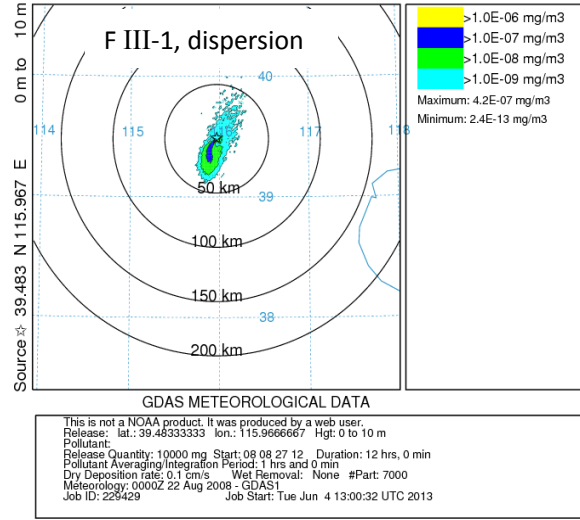
NOAA HYSPLIT MODEL
Concentration (mg/m3) averaged between 0 m and 900 m
Integrated from 0200 27 Aug to 0300 27 Aug 08 (UTC)
Release started at 1400 26 Aug 08 (UTC)



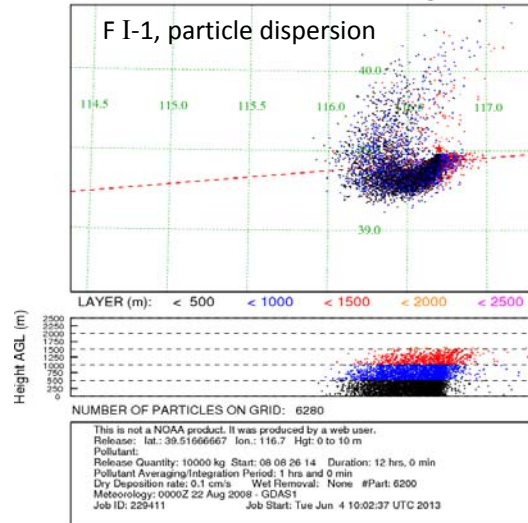
NOAA HYSPLIT MODEL
Backward trajectories ending at 0100 UTC 28 Aug 08
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Concentration (mg/m3) averaged between 0 m and 100 m
Integrated from 0000 28 Aug to 0100 28 Aug 08 (UTC)
Release started at 1200 27 Aug 08 (UTC)



NOAA HYSPLIT MODEL
PARTICLE CROSS-SECTIONS
PARTICLE POSITIONS AT 03 UTC 27 Aug 08



NOAA HYSPLIT MODEL
PARTICLE CROSS-SECTIONS
PARTICLE POSITIONS AT 01 UTC 28 Aug 08

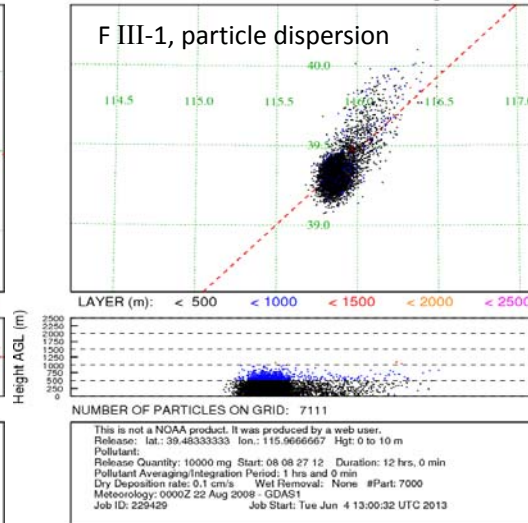


Fig. 3 A 48 h back trajectories and 12 h forecast dispersions for Flight I-1 and III-1 of G1.

Page 9, Line 2, *What is GDAS?*

We have added it, and revised it as “Global Data Assimilation System (GDAS) archived meteorological data have been used as input for both the back trajectory and dispersion model (Draxler and Rolph, 2013, <http://ready.arl.noaa.gov/HYSPLIT.php>).”

Page 9, Line 4-15, *What is the purpose of showing these data? Unless you can link them to the aircraft measurements, they convey no relevant information.*

Thanks, and we have deleted this part following your suggestions.

Page 9, Line 16-22, *This isn't relevant. Measurements at the surface cannot be easily related to the measurements from the aircraft. Remove all references to the ground*

based measurements unless you flew directly over them.

Thanks, and we have deleted this part following your suggestions.

Page 9, Line 24-27 to Page 10, Line 1-4, Big revisions have been made on this part, as the re-numbering of different flights, and the deletion of the ground parts. They are revised as “Based on both the back trajectory and the archived dispersion model analysis, the flight routes were classified into four groups related to the origin of the air masses in the region where flights were conducted, these four origins were from the south, north and northwest, east, and a mixture of origins. It should be noted that the back trajectories and dispersion results did not always show consistent patterns for all flights; hence, the dispersion results and the adjacent variation of sampling were carefully considered for grouping. The flights that correspond to the four groups are: 1) Flight I-1 (FI-1), FII-1, FII-2, FIII-1 and FIII-2 for group 1 (G1), 2)FI-2, FI-3, FI-6, FI-8, FI-9 and FII-4 for group 2 (G2), 3)FI-4, FI-5, FI-7 and FII-3 for group 3(G3), and 4) FI-10, FIII-3 and FIII-4 for group 4(G4). For each group, the box and whisker plots of SO₂, NO_x, O₃, CO, CN and PM_{0.5} are shown in Fig. 2 at 2100m, 900m and 600m, respectively. The box part represents the central 50% of the data, the lower edge is the 25th percentile, and the upper edge is the 75th percentile. The whiskers in the plot represent the error bars. The following discussion will be in the same order as the group numbering.”

Page 10, Line 5, it has been revised as “Flights of G1: air mass origin from the southern transport of pollution”

Page 10, Line 8, *These data need to be put in graphical form. It is very difficult to look at this table and see the patterns that you describe. These can be in the form of bar and whisker plots where you show for each group the average, standard deviation and maximum for each flight and each altitude.*

Thanks, and we have redone the box plots following your suggestions, as the new Figure 2 shows.

Page 10, Line 9-11, *This remains to be proven with the dispersion plots. You should be able to do a simple F-test or student T test between the average values from all the flights in one group with all the flights in the other groups to see if the differences are*

statistically significant.

Thanks, and we have done the T-test between different groups at the three altitudes to test it, as the new Table 3 shows. Many new sentences were added, as “The back trajectories and dispersion results of FI-1 and FIII-1 were shown as examples in Fig. 3, indicating the influences of southern transportation. We have done the student t-tests between G1 and other groups for gases and particles at different altitudes, as shown in Table 3. The results certified the significant differences between G1 and other groups for all gases and particles at the three altitudes. These flights, and the associated back trajectory and dispersion analysis, indicated the probable influence of emissions from the many large cities to the south into the research area.”

Page 10, Line 12, *Describe what makes the features remarkable. This is where the figures are needed and not a table.*

Thanks, and we have revised it as “The gases pollutants showed some remarkable features significantly higher concentrations between G1 and other groups, particularly in the air with high concentration of SO₂ and O₃, as shown in Table 2”.

Page 10, Line 13-16, *This is speculation. Can you prove it?*

Following your suggestions, we have revised it. You know, as only the gases and number concentration of particles were made during the campaigns. It is very difficult to get exact proofs for that. In addition, not too much aircraft measurements especially not so many field campaigns during Beijing around areas in China have been made before. However, we still tried to revise them, as “This may be due to air stagnation under conditions of low wind speeds and highly active photochemistry; the urban emissions of both primary compounds and precursors for secondary ions lead to an additional pollutant on top of the already elevated regional level (Van Pinxteren et al., 2009; Streets et al., 2007). This may be shown from the higher concentration of particles, especially for PM_{0.5} in G1.”

Page 10, Line 24-28 to Page 12, Line 5, *These need to be in the figures I suggested above and don't need to be described in detail for each day. Discuss the average over all the days and their standard deviations and maximum values as a group for each altitude, not each flight.*

Thanks, all the results have been done in Fig. 2, and we have redone the results here, as “For G1, the gases and particle pollutants come mostly from sources nearby to the south, as the back trajectory and the forecast dispersion shows in Fig. 3. In addition, the levels of gas pollutants in G1 is significantly higher than those in other groups, especially for SO₂, NO_x, and O₃. The higher the altitudes, the more variation between flights of G1 and other groups, as shown in Table 2 and Fig. 3. It is obvious that the higher concentration of SO₂ especially in 2100 m may be a good tracer for the southern sources from regional transport. As an example, the SO₂ measured from FI-1 showed tens to hundreds times higher than the average of other flights (4.85 compared to 0.03~0.69 ppbV) at 2100 m, 2~18 times higher at 900 m (11.1 compared to 0.62~3.37 ppbV), and 2~30 times higher at 600 m (11.7 compared to 0.78~5.19 ppbV). The NO_x showed similar variation with SO₂.

For O₃, this group showed concentrations similar to the other groups at 2100 m, which may be the regional level of this height, as shown in Fig. 2 and Table 2. However, it showed more variation between different flights at lower altitudes especially 600 m, which may suggest the differences of the ground transport or dispersion, as Fig. 3 showed. For NO_x pollution, all the flights showed significantly higher contribution of NO₂, which contributes more than 90% of NO_x.”

Page 12, Line 6-7, it has been revised as “Flights of G2: air mass origin from the north and northwest”

Re-do with suggested figures and discussion of only statistics over all flights. Move and prove some speculation comments.

Following your suggestions, we have re-written this part, and added the statistic results in Table 3 and Figure 2, and revised this part as “Most of the G2 flights showed generally low concentrations of gases pollutants at all three different heights, compared to the other groups, as shown in Table 2 and Fig. 2. The back trajectories and dispersion results of FI-2 and FII-4 are shown as examples in Fig. 4, indicating the influences of northern and northwestern transportation. We have done the student t-tests between G2 and other groups for gases and particles at different altitudes, as shown in Table 3. The results certified the significant lower variation between G2 and

other groups for all gases and particles at the three altitudes. These flights indicated the possible influences of transport from the northern or northwestern direction of air that generally has lower pollutants due to fewer sources of gas and particles, as shown in Fig. 4 (Guo et al., 2004; Van Pinxteren et al., 2009).

The NO_x and SO₂ showed significantly lower concentrations in G2, compared with other groups especially G1. The average values for were 30~65% and for 25~75%, respectively, of those measured during the G1 flights. Faster flowing air with fewer pollutants from the north-northwest can clear out the more local pollutants (Guo et al., 2004; Van Pinxteren et al., 2009).

The number concentration of CN and PM_{0.5}, however, showed different characteristics than the gases. The concentration measured by the G2 flights were higher than those in G1 at 900m and 600m. The average values measured by G2 flights were ~3.2-3.8 times greater than the G1 flights for CN and ~1.3 times higher for PM_{0.5}. The inverse characteristics of gases and particles may again verify that the characteristics of transport from the northern or northwestern direction, i.e. lower gases pollutants on all heights, lower CN on high altitudes but higher on lower altitudes. The contribution of dust may be the reason, as many researches on dust events in Beijing and surrounding areas have shown (Zhang et al., 2010; Sun et al., 2010).”

Page 13 Line 13, *Re-do with suggested figures and discussion of only statistics over all flights.*

Following your suggestions, they have been revised as “The G3 flights were in air that had come from the east, especially at 600 and 900 m. This is air, the mixing of the farther sea sources and urban pollutants, as Fig. 5 shown.

For gases pollutants especially NO_x and SO₂, the average concentrations fall between the G1 and G2 flight groups, as shown in Table 2 and Fig. 2. The particle concentrations, however, especially PM_{0.5} showed higher levels at 600m than most flights in other groups, as the average number concentration of PM_{0.5} reached 3.3×10^4 N/cm³, compared with 1.4×10^4 for G1 and 1.9×10^4 N/cm³ for G2, as shown in Fig.2 and Table 2. This may verify the influences of eastern sea sources, lack of

gases pollutants but more sea salt particles, as the most dominant sea salt aerosols (ammonium sulfate and acidic sulfate) dominated a median diameter of 0.14 μm , and sea-salt particles concentrated with modes at 0.2~0.6 μm (McInnes et al., 1997; O'Down et al., 1997). Additionally, the mixing of local sources during the transport contribute more to the gases pollutants.”

Page 13, Line 23, *Re-do with suggested figures and discussion of only statistics over all flights.*

Following your suggestions, this part has been revised as “The G4 flights showed inverse transport directions between back trajectories and forward transport, as Fig. 6 shown. Take the FIII-3 and FIII-4 as examples, it showed eastern or southeastern and western or northwestern directions for the back trajectories, while showed inverse directions for the forecast transport, i.e. western or northwestern for FIII-3 and eastern or northeastern for FIII-4, respectively.

In addition, these flights showed the influences of the mixture of different transport directions for the pollutants along the flight paths. This mixture causes the pollutants to have different characteristics than the other groups, i.e. the transport at lower altitudes was more from the polluted southern direction but at higher altitudes more from the cleaner northern direction, as Fig. 6 shown. Similar to other observations (Van Pinxteren et al., 2009), the lengths of the back trajectories are much shorter for air arriving from the south at lower altitudes that is slower moving, favoring the accumulation of pollution in a stagnant mixed layer before arriving at the sampling sites. The back trajectories from the north or northwest directions are much longer at higher altitudes and these faster moving air masses from cleaner regions is evident in the gas concentrations. The concentration of gases at 600 and 900 m from the G4 flights are similar to the G1 flights when the air was from the same southern sources. The particle concentrations are puzzling at 2100 m since the CN and $\text{PM}_{0.5}$ are higher than the other three groups of flights at this altitude.”

Page 13, Line 23, Following your suggestions, this part has been revised as “The G4 flights showed inverse transport directions between back trajectories and forward transport, as Fig. 6 shown. Take the FIII-3 and FIII-4 as examples, it showed eastern

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Page 14, Line 26, As different from other reactive gases, CO showed special characteristics, and was discussed. The new version of this part has been revised as “The trace gas CO is different from the reactive gases, NO_x, SO₂, and O₃, as it is longer lived and less reactive. It can be transported over longer distance and is a potential tracer of long range transport. However, local sources of CO can complicate the interpretation of CO as a tracer of air mass origin. The variation at the three flight levels showed different characteristics even within the same groups. Fig. 7 gives an example from several flights of the variation of CO concentration at three altitudes during different flights.

For L1, three types of CO-related of flights were identified: 1) CO concentration lower at 2100 m but higher at 600 m and 900 m, 2) higher concentration at 2100 m but lower at 600 m and 900 m and 3) similar on all altitudes.

The back trajectory analysis for the Type 1 CO concentration showed longer

transport at 2100 m (air mass from the northwest direction on Aug. 27 and northern on Sep. 3) and shorter, i.e. or regional sources, at lower altitudes (air mass from the south and more local sources on Aug. 27 and Sep. 3).

The back trajectory analysis for the Type 2 flights showed that air was from the northwest at the three flight altitudes but at the air had come from a longer distance at 2100 m, as illustrated in Fig.4 (a) showed. The northwestern direction at lower altitudes had a “cleaner” effect on the local and regional CO pollution, which has been reported on the influences of the dust storm (Guo et al., 2004). The high concentration of 2100 m may show the transport effect from northwestern direction on higher altitude. This showed that CO may be a good tracer for northwestern direction on 2100 m when all the heights showed the same transport direction, with the consideration of the class 1 flights.

The third class of flights included the flight in the morning and afternoon on Oct. 11 and Oct. 13. It showed similar level at the three heights of the same flight, and the CO level was 0.37~0.52, 0.14~0.18, and 0.53~1.15 ppmV, respectively. The back trajectories showed the combination of class 1 and class 2 above. However, it is different for flights on Oct. 11 and 13, i.e. the same transport direction and similar transport range at the three heights on Oct. 11, but the longer transport western direction on 2100 m and southwestern direction on 900 m and 600 m. The “cleaner” effect from the northwestern transport direction made the CO level on the afternoon flight significantly lower than the morning flight, as Fig. 7 and Table 2 showed.”

Page 16, Line 10, Following your suggestions, this part has been re-written, and the new version are as “The concentrations of particles were always higher when the air masses were from the south, consistent with previous observations (Van Pinxteren et al., 2009; Guinot et al., 2007; Wang et al., 2005; Wehner et al., 2008). Van Pinxteren et al. (2009) investigated two, primary factors that influence particle properties. Firstly, the surrounding areas of Beijing show different characteristics, for example, in the southern direction the region is highly populated and industrialized, whereas in the north or northwest directions, and partially in the east, are mountains or deserts with less anthropogenic emissions. Thus air masses from the southern areas

are influenced by high pollutant emissions and those from northern areas have been impacted less by such emissions. Secondary, wind speeds are often lower during southern advection (Wehner et al., 2008), as was the case during the current study. Similar to other observations (Van Pinxteren et al., 2009), the lengths of the back trajectories are much shorter for air masses from the souths (See Figure 3&6), and the slower movement this air favors the accumulation of gases and particles before arriving at the sampling sites.

The size distribution of particles do not necessarily follow the same trends as were seen with the gases with respect to air mass origin or altitude. Fig. 8 shows the variation of size distributions with the three altitudes. The size distribution at 2100 m has the peaks concentrated between of 20~30 nm. At the lower altitudes, the peaks fall between 80~120 nm. At 2100 m the width of the size distribution, centered around 20 nm, remains fairly constant whereas at 900 and 600 m the width fluctuates with several periods showing larger increases at sizes between 25 and 80 nm, suggesting a mixture between the air in the mixed layer with free tropospheric air.

Local emissions and regional transport may be the most important factors affecting the size distribution of particles. Air masses originating from directions at different heights will bring aerosol particles that have originated from differing sources but then also age differently. e.g. northwestern direction at 2100 m and southern direction at 900 m and 600 m on Aug. 27 flight, as shown in Fig. 3. Correspondingly, the size distribution of 5.6-560 nm particles showed peaks of 20~30 nm at 2100 m and 80~120 nm at 900 m and 600 m. While, it showed no peaks at 2100 m and showed peaks of ~0.7 μm at 900 m and 600 m at size range of 0.5~20 μm , as shown in Fig. 9. This may indicate the influencing effects of the different transport directions, i.e. the cleaner effects of northwestern and polluted effects of southern transportation, which may be consistent with other ground-based researches (Van Pinxteren et al., 2009; Guinot et al., 2007; Wang et al., 2005; Wehner et al., 2008).

Local sources mostly impact the particle characteristics below 1000 m while aging and photochemistry impact those in the free troposphere at 2100 m. As shown in Fig. 8, the size distribution of particles 5.6-560 nm varied a lot over the time

periods when the aircraft was at 900 m and 600 m, and the peaks of size distribution showed tendency to smaller sizes at 4:05~4:08, 4:12~4:16, and 4:43~4:49 am (GMT) at 900 m. Similar results are shown for size distribution at 600 m. In order to show research the differences of the changes during 4:05~4:08, 4:12~4:16, and 4:43~4:49 am in this flight, the average size distribution of the changes were specially listed in Fig. 10 in 900 m, as well as the averages of the sizes at different altitudes. Obviously, the size distributions peaked at 81 nm at 900 m for the average of special flight areas, and 93 nm on other average flight areas at both 600 m and 900 m. As the aircraft measurements were conducted along linear tracks between two sites, several highways and freeways below contribute the differences of the peaks of size, after carefully checking with the flight time and flight areas. Fig. 10 shows the average size distribution of particles at different heights on FI-1 flight, and the distribution above the highway was specially noted.

However, we didn't observe similar 81 nm peaks during other flights over the same flight areas. The meteorological conditions may have contributed to this. There was a moderate rain from a thunderstorm on Aug. 27 (www.wunderground.com). The wet deposition helps scavenge the aged particles (Nilsson, et al., 2001; Elperin et al., 2011). Fresh emission from vehicles was observed, which was consistent with results from Wang et al. (2011). They conducted the on-road emissions of individual diesel vehicles in and around Beijing by a mobile platform equipped with fast response instruments such as EEPs, and got bimodal modes peaking around 10 nm and 80 nm, similar to 81 nm in special flight areas in this study. This confirms the potential impacts from vehicles emission of highways or freeways, in addition to the meteorological factors.”

Page 18, Line 18, For “Summary and Conclusion” part, only the major results and conclusion were summarized in the new version, as “Intensive aircraft measurements of gaseous pollutants and particles in the regions around Beijing were conducted during Aug. 27 to Oct. 13, 2008. The selected flight levels were 600, 900, and 2100 m along three different flight routes. Major findings include:

(1) Based on the back trajectories, forecast transport and pollution variation, the

flights were classified into four groups based on the origin of air masses along the flight tracks: 1) air from the south, 2) air from the north or northwest, 3) air from the east and 4) the mixing of air from the north and south but at different altitudes.

- (2) Results from group 1: The high concentration of SO₂ especially at 2100 m may be a good tracer for the sources from southern transport of pollution from the south, particularly from coal-fired power plants.
- (3) Results from group 2: Most of the flights showed the lowest concentration of gas pollutants, at all three different heights, of the four groups of flights. These flights indicated the possible influences of transport from the cleaner northern or northwestern direction, where large regions are sparsely populated and arid.
- (4) Results from group 3: for the average gas pollutants fell between groups 1 and 2; however, the CN concentrations are higher than most flights at the two heights.
- (5) Results from group 4: In this group of flights with air mass origins depending on the flight altitude, the lengths of the back trajectories are much shorter from the south at lower altitudes, indicating slower movement of air masses that favors the accumulation of pollution before arriving at the sampling sites. The longer back trajectories from the north or northwest at higher altitudes bring cleaner air from arid, less populated regions. These measurements showed the mixture of air mass origins in the concentration of both gases and particles.

These results illustrate the complexity of how anthropogenic emissions evolve in a densely populated region where the dynamics that bring air from one region, mixing it with air from local sources create an interaction between gases and particles that will require a much more in-depth analysis that is currently in progress.”.

In addition, the original conclusion 6 and 7 of“(6) Different from the reactive gases of NO_x, SO₂, and O₃, CO was long lived and non-active. It can be transported for a long range, and could be used as a possible tracer of long range transport. However, local sources of CO are complicated, and may be mixed with transport sources. The variation of different heights showed different characteristics even at the same groups. Results showed that CO may be a good tracer for northwestern direction on 2100 m when all the heights showed the same transport direction, with the

consideration of the class 1 flights.

(7) The variation of size distribution of 5.6~560 nm particles showed three influencing factors, i.e. transport direction, ground emission sources and meteorological factors. Take the flight Aug. 27 as an example, the different transport direction contribute the different size distribution at 2100 m and lower altitudes. For northwestern transport of 2100 m, the peaks concentrated on 20~30 nm. While, for the southern transport of lower altitudes, the peaks concentrated on larger sizes of 80~120 nm. Additionally, the peaks shifted to smaller sizes above the special highways or freeways influenced areas. The ground sources influence much to the particles below 1000 m and no obvious influences on 2100 m. While, the wet deposition on Aug. 27 helps scavenge the aged particles and the emission of freshly emission from vehicles, which could explain the phenomenon was not observed on other flights.” were deleted.

Page 20, Line 22, For the “Acknowledgements” part, we appreciate great help from the reviewers especially Dr. Darrel Baumgardner from Universidad Nacional Autonoma de Mexico. Thus we added some sentences as “The authors wish to thank Dr. Darrel Baumgardner from Universidad Nacional Autonoma de Mexico, and the other anonymous reviewer for their useful comments and language editing which have greatly improved the manuscript.”