Supplemental Material for "Measurement Report: Aircraft Observations of Ozone, Nitrogen Oxides, and Volatile Organic Compounds over Hebei Province, China"

Sarah E. Benish[1], Hao He[1], Xinrong Ren[1,2], Sandra J. Roberts[3], Ross J. Salawitch[1,3], Zhanqing Li[1,4], Fei Wang[4,5], Yuying Wang[6], Fang Zhang[4], Min Shao[7], Sihua Lu[7], Russell R. Dickerson[1]

[1] Department of Atmospheric and Oceanic Science, University of Maryland, College Park, MD 20742, USA.
[2]Air Resources Laboratory, National Oceanic and Atmospheric Administration, College Park, MD 20740, USA.
[3] Department of Chemistry and Biochemistry, University of Maryland, College Park, MD 20742, USA.
[4] State Key Laboratory of Earth Surface Processes and Resource Ecology, College of Global Change and Earth System Science, Beijing Normal University, Beijing, 100875, China.
[5] Key Laboratory for Cloud Physics, Chinese Academy of Meteorological Sciences, Beijing, 100081, China.
[6] Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, School of Atmospheric Physics, Nanjing University of Information Science and Technology, Nanjing, 21004, China.
[7] College of Environmental Science and Engineering, Peking University, Beijing, 100871, China.

Text S1

We exclude two WAS canisters from this analysis due to evidence of contamination. The first sample was collected on May 21 at 399 m pressure altitude. This sample was heavily polluted with i-butane (25.8 ppbv), i-pentane (57.7 ppbv), as well as longer chain alkanes like 2,3-dimethylbutane (4.2 ppbv), 2-methylpentane (5.9 ppbv), cyclopentane (2.7 ppbv), 2-methylheptane (16.1 ppbv), and 3-methylpentane (3.5 ppbv) in addition to aromatics like toluene (41.3 ppbv) and benzene (20.8 ppbv). Many of these compounds are typical of fuel evaporation or from petrochemical industries, indicating this canister may have directly sampled directly in the plume of one of these sources. Since this study is primarily focused with evaluating aloft VOCs away from their direct emission sources, the data from this canister were removed from this analysis.

The second contaminated sample was collected on May 28 at 3:36 UTC. This sample was filled to ambient pressure at 3000 m in relatively clean air, based on *in situ* observations at the time the canister was collected (CO=111 ppbv, CH_4=1890 ppbv, CO_2=406 ppmv, O_3=84 ppbv). The concentrations of VOCs for this sample are outliers relative the associated abundances of the trace gases. This anomaly is indicative of valve leakage during transit or ambient air entering the WAS canister after the flight. The observed CO to acetylene ratio (ppbv/ppbv), often used as a tracer for the age of an air mass, was much smaller in this sample (70 ppbv/ppbv) compared to other samples collected at a similar altitude (~400 ppbv/ppbv).

Table S1. Summary statistics of the 1-second measured concentrations for O_3, NO_2, and CO, flight path descriptions, and weather conditions for each flight during ARIAs. Negative values of NO_2 indicate when the instrument was measuring around the detection limit.

Date (DOY)	Takeoff (LST)	Landing (LST)	Mean O_3 (Range), ppbv	Mean NO_2 (Range), ppbv	Mean NOy (Range), ppbv	Mean CO (Range), ppbv	Flight Description	Weather Conditions
May 8 (129)	10:30	14:32	76.6 (62.7-83.9)	No data	15.6 (9.0-29.4)	No data	Spirals over Julu (400-3500 m) at 10:58 LST, Quzhou (350-3500 m) at 12:00 LST, Xingtai (400-3000 m) at 12:23 LST, and Shijiazhuang (100-3500 m) at 14:05.	A high over the region a weak low to the N moving to the E. Stron winds in daytime (surfa wind speed up to 10 m Cold front passed 2 d prior.
May 15 (136)	12:17	15:04	64.6 (54.4-85.8)	No data	No data	No data	Spirals over Julu (400-3500 m) at 12:43 LST and Quzhou at 13:41.	A high over the region an occluded front to th over the Yellow Sea Surface winds mostly f the NE up to 10 m/s
May 16 (137)	15:03	15:54	85.3 (70.5-96.0)	No data	22.8 (6.1-29.6)	No data	Flight to the southeast to the W of Julu. Flight altitude about 400 m.	A high over the region a cold front to the E of Korean Peninsula. Mor surface winds from the (< 8 m/s) with a shift i morning from the SE (m/s).
May 17 (138)	8:21	11:13	80.1 (45.0-99.1)	8.8 (–0.1-38.4)	30.2 (0.2-89.7)	590.5 (114.3-6053.6)	Low altitude transect to Julu, with spirals at Julu (650-2800 m) at 9:47 LST and Quzhou (400-3000 m) at 10:19 LST.	Surface high pressu conditions throughout region and low pressure the N and W. A 700 h ridge is situated over N Korea. Surface W wind the morning (< 5 m/s) a shift in late-morning f the SE (< 10 m/s).
May 19 (140)	15:42	17:09	97.1 (75.4-130.2)	1.4 (–0.1-6.8)	11.3 (3.6-29.1)	131.4 (90.8-540.0)	Spirals over the airport.	Weak surface high pres conditions over the reg and low pressure syste over S Mongolia and I Mongolia. Upper level hPa trough from the previous day moved E Sea of Okhotsk. Surfa winds from the W in t morning (< 5 m/s) wit shift in late-morning fro the SE (< 7 m/s).
May 21 (142)	11:57	13:41	99.5 (67.1-145.6)	1.9 (–0.1-16.4)	No data	238.5 (80.5-564.5)	Flew to southeast at low altitude (1000 m) to a point (114.9 °E, 37.6 °N). Spirals over Quzhou (300-3000 m) at 12:40 LST and Xingtai (300-2400 m) at 13:34 LST.	Weak surface high pres conditions over the reg with a Siberian anticyc to the N. Surface win from the W in the morn with a shift in late-mor from the E (< 6 m/s)
May 28 (149)	10:16 16:29	13:26 18:24	86.3 (63.5-100.3); 88.9 (72.9-112.3)	3.2 (–0.1-27.0); 2.6 (0.01-10.4)	No data	332.2 (97.1-1264.9); 215.2 (88.1-963.3)	Morning flight flew spirals over Xingtai (350-3000 m) at 11:02 LST and Julu (450-2500 m) at 12:29 LST. During the afternoon flight, spirals over Xingtai (350-3000 m) at 16:57 LST.	High pressure over th region and a stationa front ~1000 km to th near Shanghai. Surfac winds mostly from the (< 5 m/s).
June 2 (154)	13:47	14:53	94.9 (79.6-106.3)	1.5 (–0.1-5.4)	24.3 (14.9-70.7)	256.7 (95.1-487.5)	Spirals over the airport.	High over the region v low pressure over cen China and a stationary f to the S near Shangh

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								Light surface winds (< 5 m/s) mostly from the W.
June 6 (158)	10:08	12:01	99.9 (67.5-134.7)	0.7 (−0.1-4.9)	No data	296.2 (105.1-573.2)	Low altitude (< 2000 m) spirals to the SE of Shijiazhuang. Spirals over Julu at 10:44 LST.	Several weak low pres systems over the region stationary front is over East China Sea. Varia winds less than 5 m/s
June 11 (163)	11:02	13:45	76.7 (57.2-90.8)	2.3 (− 0.1-6.7)	16.9 (11.1-23.8)	187.4 (88.2-412.9)	Low altitude transect (2000 m) to NE Julu. Spirals over Xingtai (600-3000 m) at 11:54 LST and Shijiazhuang (600-3000 m) at 13:12 LST.	Low pressure over reg with a Siberian anticyc over Mongolia. A stationary front is locat the S near Taiwan. Vari surface winds, with th strongest winds (12 m from the N in the morn

Table S2. Summary statistics of alkanes, alkenes/alkynes, and aromatics quantified for all WAS canisters (pptv), as well as the method detection limit (MDL, in pptv), rate constants with OH (kOH), maximum incremental reactivity (MIR) value, and ratio to CO (pptv/ppbv) for compounds with R>0.50. Values less than 1 pptv are not shown.

	Mean (STD)	Min	5th	25th	50th	75th	95th	Max	MDL*	kOH‡	MIR†	
					Alkanes							
Ethane	2648 (710)	1804	1902	2033	2525	2998	4066	4154	50	$6.90 \times 10^{-12} \times e^{-1000/T}$	0.28	
Propane	1391 (231)	978	1044	1196	1356	1509	1769	1887	21	$7.60 \times 10^{-12} \times e^{-585/T}$	0.49	
n-Butane	363 (278)	83	92	207	259	480	1131	1210	30	$9.80 \times 10^{-12} \times e^{-425/T}$	1.15	
2,2-Dimethylbutane	13 (14)	2	3	5	9	17	42	64	7	$3.22 \times 10^{-11} \times e^{-781/T}$	1.17	
2,3-Dimethylbutane	44 (93)	2	2	5	11	27	293	400	5	$1.24 \times 10^{-17} \times T^2 \times e^{-585/T}$	0.97	
i-Butane	624 (997)	56	70	109	246	673	3546	3963	29	$1.16 \times 10^{-17} \times \times T^2 \times e^{225/T}$	1.23	
n-Pentane	119 (113)	19	26	54	71	155	400	479	5	$2.44 \times 10^{-17} \times \times T^2 \times e^{183/T}$	1.31	
i-Pentane	674 (1255)	32	48	118	168	413	3785	5444	12	3.70×10^{-12}	1.45	-
Cyclopentane	34 (64)	2	2	5	12	25	168	296	26	$2.67 \times 10^{-11} \times e^{-590/T}$	2.39	-
Methylcyclopentane	26 (29)	2	3	7	15	28	91	115	8	7.66×10^{-12}	2.19	-
2-Methylpentane	111 (144)	8	11	39	61	103	363	667	5	5.30×10^{-12}	1.5	-
3-Methylpentane	53 (88)	3	3	9	26	55	224	395	7	5.40×10^{-12}	1.8	-
2,3-Dimethylpentane	27 (33)	4	4	9	19	26	86	152	4	$1.95 \times 10^{-11} \times e^{-330/T}$	1.34	-
2,4-Dimethylpentane	31 (53)	3	3	6	11	25	137	228	5	$2.49 \times 10^{-11} \times e^{-443/T}$	1.55	-
2,2,4-Trimethylpentane	433 (1117)	9	11	28	57	236	2120	5422	3	$2.09 \times 10^{-12} \times (\frac{T}{298})^{2.00} \times e^{140/T}$	1.26	-
2,3,4-Trimethylpentane	232 (652)	9	9	15	33	96	987	3253	8	$9.85 \times 10^{-12} \times e^{-124/T}$	1.03	-
n-Hexane	123 (180)	6	8	24	46	130	541	699	16	$1.53 \times 10^{-17} \times T^2 \times e^{414/T}$	1.24	-
Cyclohexane	15 (13)	1	2	5	9	27	44	44	16	$2.88 \times 10^{-17} \times T^2 \times e^{-309/T}$	1.25	-
Methylcyclohexane	17 (23)	3	5	6	10	14	54	114	8	1.18×10^{-11}	1.70	-
2-Methylhexane	39 (72)	6	8	13	18	27	147	362	8	6.86×10^{-12}	1.19	-
3-Methylhexane	44 (88)	7	7	12	16	33	178	438	6	7.15×10^{-12}	1.61	-

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n-Heptane	41 (52)	9	12	16	22	39	142	255	7	$1.59 \times 10^{-17} \times T^2 \times e^{478/T}$	1.07	-
2-Methylheptane	399 (1106)	11	13	32	63	172	1718	5515	8	$2.51 \times 10^{-17} \times T^2 \times e^{447/T}$	1.07	-
3-Methylheptane	15 (13)	7	7	8	11	14	56	59	9	$2.51 \times 10^{-17} \times T^2 \times e^{447/T}$	1.24	-
Octane	26 (19)	10	10	15	23	29	59	102	12	$2.76 \times 10^{-17} \times T^2 \times e^{378/T}$	0.90	-
n-Nonane	22 (12)	13	14	15	17	25	39	72	21	$2.51 \times 10^{-17} \times T^2 \times e^{447/T}$	0.78	-
n-Decane	58 (57)	14	14	24	38	76	155	288	10		0.68	
Alkenes/Alkynes												
Acetylene	803 (465)	234	284	454	578	1175	1506	1934	48	$1.69 \times 10^{-12} \times e^{-233/T}$	0.95	1.4
Ethylene	884 (923)	185	191	281	405	1093	2941	3536	30	$2.14 \times 10^{-12} \times e^{411/T}$	9.00	2.9
Propylene	168 (44)	102	104	143	164	199	223	308	25		11.66	-
1-Butene	23 (10)	10	11	17	19	25	43	46	30	$6.60 \times 10^{-12} \times e^{465/T}$	9.73	-
cis-2-Butene	3 (6)	-	-	1	1	2	7	31	23	$1.10 \times 10^{-11} \times e^{487/T}$	14.24	-
trans-2-Butene	3 (3)	-	-	1	2	4	9	16	31	$1.01 \times 10^{-11} \times e^{550/T}$	15.16	-
Isoprene	35 (39)	2	5	8	20	37	117	138	15	$2.70 \times 10^{-11} \times e^{390/T}$	10.61	-
1-Pentene	8 (3)	4	4	6	7	9	14	18	9	$5.86 \times 10^{-12} \times e^{500/T}$	7.21	-
cis-2-Pentene	2 (3)	-	-	1	1	2	4	16	8	6.54×10^{-11}	10.38	-
trans-2-Pentene	2 (3)	-	-	1	1	2	12	14	8	6.69×10^{-11}	10.56	-
1-Hexene	6 (5)	3	3	4	5	6	9	27	11	3.70×10^{-11}	5.49	-
Aromatics												
Benzene	510 (521)	63	96	188	330	570	1819	2183	7	$2.30 \times 10^{-12} \times e^{-190/T}$	0.72	1.8
Toluene	757 (1188)	31	43	159	300	627	4064	4402	5	$1.80 \times 10^{-12} \times e^{340/T}$	4.00	-
Styrene	14 (12)	4	4	6	8	18	43	45	13	5.80×10^{-11}	1.73	-
m/p-Xylene	108 (155)	16	22	43	62	117	345	789	2	1.87×10^{-11}	7.80	-
o-Xylene	43 (51)	8	11	17	28	44	119	263	3	1.36×10^{-11}	7.64	-
Ethylbenzene	73 (85)	12	15	27	45	83	198	423	3	7.00×10^{-12}	3.04	-
Isopropylbenzene	15 (8)	7	7	10	13	18	28	48	20	6.61×10^{-12}	2.52	0.02
n-Propylbenzene	15 (20)	4	4	7	10	15	37	104	16	5.80×10^{-12}	2.03	0.06
2-Ethyltoluene	14 (17)	5	5	7	9	13	35	89	10	1.86×10^{-11}	5.59	0.05
3-Ethyltoluene	19 (18)	4	5	9	13	18	59	88	20	1.18×10^{-11}	7.39	0.05
4-Ethyltoluene	19 (26)	4	4	7	11	18	60	132	20	1.19×10^{-11}	4.44	0.07
1,3-Diethylbenzene	20 (28)	4	4	6	8	12	64	248	10	1.86×10^{-11}	7.10	0.15
1,4-Diethylbenzene	27 (40)	8	8	11	18	22	53	218	10	1.18×10^{-11}	4.43	0.12
1,2,3-Trimethylbenzene	18 (24)	7	7	9	13	16	43	130	2	3.27×10^{-11}	11.97	0.07
1,2,4-Trimethylbenzene	30 (30)	8	9	15	23	31	65	160	3	3.25×10^{-11}	8.87	0.08
1,3,5-Trimethylbenzene	10 (11)	3	3	4	6	8	28	54	4	5.67×10^{-11}	11.76	-

* TO-15 method, where the standard deviation of seven replicates near the detection limit are multiplied by 3.14 (Student's t value with 99% confidence).

‡ Reaction rate coefficient with OH.

† Maximum Incremental Reactivity (MIR, units=g O_3/g VOC), from Carter, (2010).

Figure S1. Left: Picture of the gas (alt-facing) and aerosol inlet (forward facing) on top of the Y-12 aircraft. Right: Picture of the Cloud Water Inertial Probe (CWIP) on the Y-12 aircraft installed under the port wing.

235 **Figure S2. Scatter plot of 1-minute average O_x (O_3+NO_2) as a function of NO_z (NO_y-NO_x) less than 30 ppbv below 1500 m. The color shows the local hour of collection. The line is the linear regression with the slope (k) and Pearson R correlation coefficient.**

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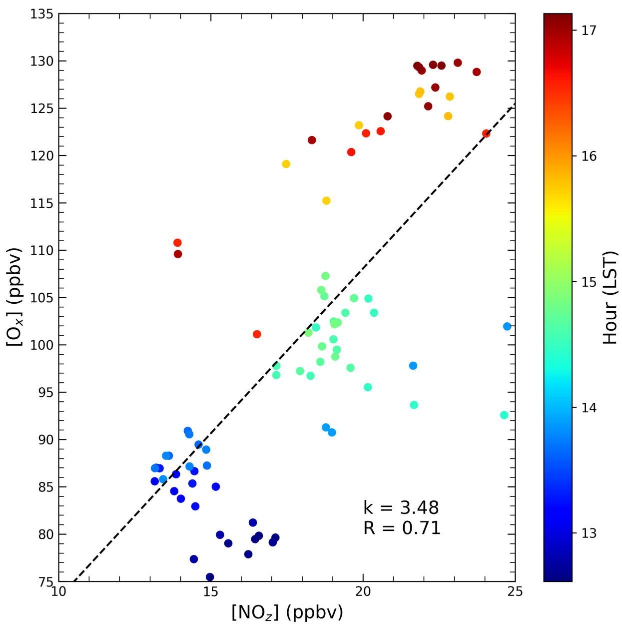

Figure S3. Vertical profiles (N=19) of 1-second O_3 concentrations (ppbv) from the Y-12 (circles) compared to concurrent average concentrations measured at the A^2BC site in Xingtai (diamonds). The average surface O_3 concentration was computed by averaging the 5-minute data interval starting 30 minutes before the spiral until 30 minutes after the spiral was completed.

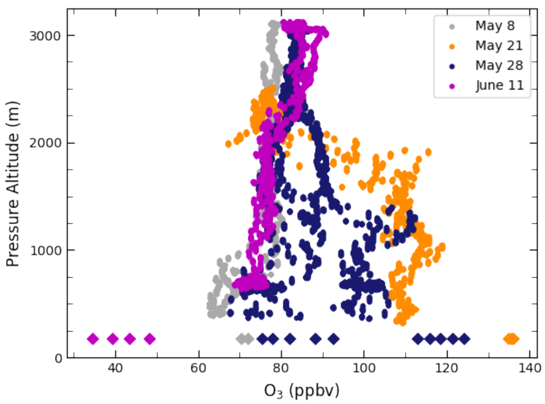

Figure S4. Scatter plot of 1-second CO (ppbv) and CO_2 (ppmv) (left) and SO_2 (ppbv) and CO_2 (right) sampled during a plume over Julu on June 6.

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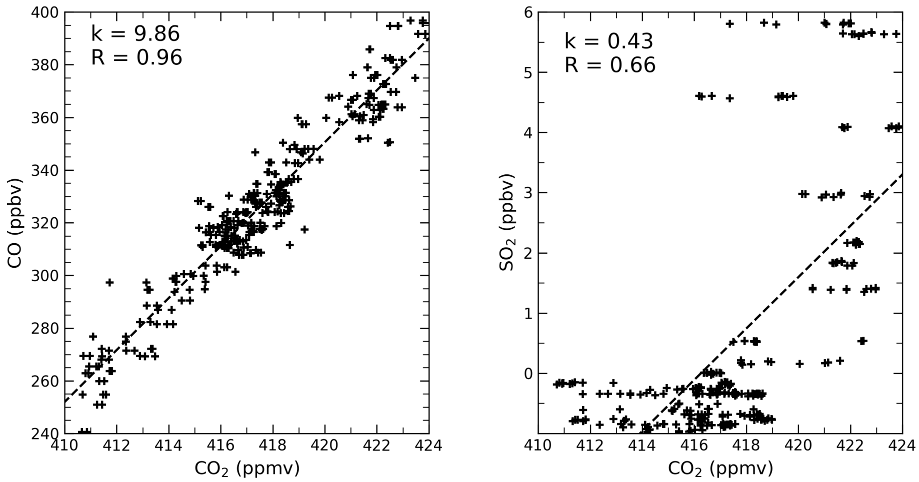

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