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Evaluation of Anthropogenic Emissions and Ozone Pollution in the North China Plain: Insights from the Air Chemistry Research in Asia (ARIAs) Campaign

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Atmospheric Chemistry and Physics Discussions

Abstract

To study the air pollution in the North China Plain (NCP), the Air Chemistry Research in Asia (ARIAs) campaign conducted airborne measurements of air pollutants including O_3, CO, NO and NO_2 in spring 2016. High concentrations of pollutants, >100 ppbv of O_3, >500 ppbv of CO, and >10 ppbv of NO_2, were observed throughout the boundary layer during the campaign. CMAQ simulations with the 2010 EDGAR emissions can capture the basic spatial and temporal variations of ozone and its major precursors such as CO, NO_x and VOCs, but significantly underestimate their concentrations. Observed emission enhancements of CO and NO_x with respect to CO_2 suggest the existence of combustion with high emissions such as biomass burning in the NCP. The comparison with emission factors from the 2010 EDGAR emission inventory indicates that the contribution of combustion with high emissions has been overestimated. Differences between CMAQ simulations with 2010 emissions and satellite observations in 2016 can reflect the change in anthropogenic emissions. NO_x emissions decreased in megacities such as Beijing and Shanghai confirming the effectiveness of recent control measures in China, while in other cities and rural areas NO_x emissions slightly increased, e.g., CMAQ predicts only ~80% of NO_x observed in the aircraft campaign area. CMAQ also underestimates HCHO (a proxy of VOCs, by ~20%) and CO (by ~60%) over the NCP, suggesting adjustments of the 2010 EDGAR emissions are needed to improve the model performance. $HCHO/NO_2$ column ratios derived from OMI measurements and CMAQ simulations show that VOC-sensitive chemistry dominates the ozone photochemical production in eastern China, suggesting the importance of tightening regulations on VOCs emissions. We adjusted EDGAR emissions based on satellite observations, conducted sensitivity experiments of CMAQ, and achieved better model performance in simulating ozone, but underestimation still exists. Because of the VOC-sensitive environment in ozone chemistry over the NCP, future study and regulations should focus on VOCs emissions with the continuous controls on NO_x emissions in China.

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Atmospheric Chemistry and Physics Discussions

1. Introduction

With rapid economic growth in the past three decades, the consumption of energy in China increased dramatically (Zhang and Cheng, 2009; Guan et al., 2018; Shan et al., 2018). Fossil fuels dominate total energy consumption, with coal still accounting for more than 50% of the carbon dioxide (CO_2) emissions in China (Shan et al., 2018). This drastic increase in fossil fuel energy consumption is accompanied with deterioration of air quality (Chan and Yao, 2008; Fang et al., 2009), posing a threat to public health (Tie et al., 2009; Kan et al., 2012; Chen et al., 2013; Lelieveld et al., 2015). Particulate matter (PM) pollution, especially $PM_{2.5}$ in the North China Plain (NCP), drew public concern and governmental actions (He et al., 2001; Ye et al., 2003; Wang et al., 2005; Sun et al., 2006; Yang et al., 2011; Zhang et al., 2012; Zhang et al., 2013). PM pollution also has complex interactions with the planetary boundary layer (PBL) and its evolution, which can further degrade the air quality (Guo et al., 2016; Li et al., 2017b). Recent studies showed that tropospheric ozone (O_3) pollution increased in China which exacerbated its complex air pollution problem (Xue et al., 2014; Verstraeten et al., 2015; Wang et al., 2017b; Ni et al., 2018).

Elevated ozone concentrations have adverse impacts on both human health (WHO, 2003; Anderson, 2009; Jerrett et al., 2009) and the ecosystem (Adams et al., 1989; Chameides et al., 1999; Ashmore, 2005). Tropospheric ozone absorbs thermal radiation and acts as the third most important anthropogenic contribution to radiative forcing of climate (Ramanathan and Dickinson, 1979; Lacis et al., 1990; IPCC, 2014). In the lower troposphere, the photolysis of ozone is an important source of atmospheric hydroxyl (OH) radicals that control the lifetimes of atmospheric species such as CO and volatile organic compounds (VOCs) (Logan et al., 1981; Thompson, 1992; Finlayson-Pitts and Pitts, 1999). Tropospheric ozone has a relatively long lifetime of several days to weeks (Stevenson et al., 2006; Young et al., 2013), leading to significant long-range transport of ozone and its precursors (Jacob et al., 1999; Derwent et al., 2004; Lin et al., 2008). Thus, investigation of ozone pollution in China is essential to support the national and international policy decision for air quality and the climate.

Tropospheric ozone is produced through complex photochemical reactions of precursors including nitrogen oxides (NO_x = NO + NO_2) and VOCs in the presence of sunlight (Haagensmit, 1952; Crutzen, 1974; Fishman et al., 1979; Seinfeld and Pandis, 2006). In China, sectors of power generation, industry, and transportation dominates the NO_x emissions (Streets et

al., 2003; Ohara et al., 2007; Zhao et al., 2013a). Before 2010, NO_x emissions in China increased substantially (Lin et al., 2010a; Zhao et al., 2013c). Analysis of satellite data revealed that recently NO_x emissions have started decreasing in highly developed regions such as the Pearl River Delta (PRD), but still increased in other regions (Gu et al., 2013; Duncan et al., 2016; Liu et al., 2016). Anthropogenic VOCs emissions had a similar increasing trend in the past decades (Bo et al., 2008; Wei et al., 2011; Kurokawa et al., 2013; Zhao et al., 2017) and are projected to increase in the future (Zhang et al., 2018). Therefore the recent increase of tropospheric ozone in China could likely be explained by the enhanced anthropogenic emissions of ozone precursors.

Due to the complex O_3-NO_x-VOCs chemistry, we need to investigate the photochemical regime for local ozone production, i.e., NO_x-sensitive or VOC-sensitive (Dodge, 1987; Kleinman, 1994). Duncan et al. (2010) used the ratio of tropospheric columns of formaldehyde (HCHO) and nitrogen dioxide (NO_2) observed by the National Aeronautics and Space Administration (NASA) Aura Ozone Monitoring Instrument (OMI) to characterize ozone sensitivity. Studies show that a NO_x-sensitive regime dominates in the United States, except in megacities such as Los Angles and New York City where the local ozone production is in VOC-sensitive or in transition regimes (Duncan et al., 2010; Jin et al., 2017; Ring et al., 2018). However, VOC-sensitive and transition regimes for ozone photochemical production exist ubiquitously in China due to large amount of NO_x emissions, especially over the NCP (Chou et al., 2009; Xing et al., 2011; Jin and Holloway, 2015; Jin et al., 2017). As such, although the current regulations in China focus only on reduction of NO_x emissions (Wang and Hao, 2012; Wang et al., 2014a), air quality might also benefit from VOCs controls.

Aircraft measurements are essential to study the precursor emissions, photochemical production, and transport of ozone pollution at regional scale. However, airborne campaigns are sparse in China (Dickerson et al., 2007; Zhang et al., 2014; Ding et al., 2015; Huang et al., 2015; Wang et al., 2017a). To better understand the characteristics of ozone pollution, the Air Chemistry Research in Asia (ARIAs) aircraft campaign was conducted in Hebei Province of the NCP during May-June 2016, which was affiliated with the surface Aerosol Atmosphere Boundary-Layer Cloud (A^2BC) experiment (Wang et al., 2018a; Wang et al., 2018b). Concentrations of major air pollutants in the lower atmosphere were measured during 11 research flights in the NCP, which were conducted in association with the NASA Korea U.S. – Air Quality (KORUS-AQ) campaign in downwind South Korea. Measurements collected in the

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Atmospheric Chemistry and Physics Discussions

ARIAs research flights and the A^2BC surface observations provide a comprehensive dataset to thoroughly study the tropospheric ozone pollution and emissions of its precursors in China.

In this study, we evaluate anthropogenic emissions and the ozone pollution in the NCP using a combination of aircraft measurements, surface monitoring data, satellite observations, and modeling results. The Environmental Protection Agency (EPA) Community Multiscale Air Quality (CMAQ) model was used to simulate the atmospheric chemistry for the ARIAs campaign. We evaluate the emission data by comparing with the aircraft measurements and satellite products, and then adjust emissions to improve the CMAQ performance. Lastly, we investigate the sensitivity of ozone production derived from CMAQ simulations and OMI observations and discuss the future ozone pollution in China.

2. Data and Method

2.1 Aircraft campaign in the NCP

With more than 250 million tons of iron and steel produced in 2016 (data from http://data.stats.gov.cn, accessed in September 2018), Hebei Province in the NCP is the most industrialized area in China. Due to its high emissions and proximity to megacities Beijing and Tianjin, the Beijing-Tianjin-Hebei area has experienced severe air pollution in the past decade (Zhao et al., 2013b; Wang et al., 2014b). In May and June 2016, the ARIAs aircraft campaign was conducted over Hebei Province to investigate the emissions, chemical evolution, and transport of air pollutants. The airborne campaign was coordinated with the A^2BC field campaign in Xingtai (XT, 37.18 °N, 114.36 °E, 182 m above sea level, ASL) and the NASA KORUS-AQ campaign to expand the study to East Asia. A Harbin Y12 research airplane (similar to the de Havilland Twin Otter) was employed to measure concentrations of air pollutants including O_3, carbon monoxide (CO), CO_2, NO_2, and aerosol optical properties. The research airplane was located in Luancheng airport, hereafter referred to as LC (LC, 37.91 °N, 114.59 °E, 58 m ASL), south of Shijiazhuang, the capital city of Hebei province with a population of 10 million. Eleven research flights were conducted during the ARIAs campaign (Fig. 1a). Vertical profiles of air pollutants from near surface (~100 m above ground level, AGL) to the free troposphere (> 3000 m) were conducted over LC, XT (the supersite of the A^2BC campaign), Julu (JL, 37.22 °N, 115.02 °E, 30 m ASL), and Quzhou (QZ, 36.76 °N, 114.96 °E, 40 m ASL).

The airborne measurements of ozone were conducted using a commercially available

analyzer (Model 49C, Thermo Environmental Instruments, TEI, Franklin, Massachusetts) (Taubman et al., 2006). NO_2 was measured using a modified commercially available cavity ring-down spectroscopy (CRDS) detector (Castellanos et al., 2009; Brent et al., 2013). Concentrations of CO and CO_2 were monitored with a 4-channel Picarro CRDS instrument (Model G2401-m, Picarro Inc., Santa Clara, CA), calibrated with CO/CO_2 standards certified at the National Institute of Standards and Technology (Ren et al., 2018). All the instruments were routinely serviced, calibrated and used for airborne measurements in the United States and China (Taubman et al., 2006; Dickerson et al., 2007; Hains et al., 2008; He et al., 2012; He et al., 2014; Ren et al., 2018; Salmon et al., 2018). Measurements of ambient air pollutants were made at 1 Hz frequency and synchronized with time, geolocation and altitude from the Global Position System (GPS).

In the ARIAs research flights, 28 whole air samples (WAS) were collected in vertical spirals at different altitudes from ~400 m to ~3500 m. The WAS were analyzed using gas chromatography (GC) with Flame Ionization Detection (FID) and Mass Spectroscopy (MS) by the College of Environmental Sciences and Engineering at Peking University. 74 species of alkanes, alkenes/alkynes, aromatics, and halocarbons were identified and quantified for a study on ozone photochemical chemistry (see details in Benish et al., 2019). Detection limits for the compounds ranged from 2 to 50 pptv. Surface observation of trace gases including O_3, CO, NO, and NO_x were measured at the A^2BC Xingtai supersite using analyzers manufactured by Ecotech (Wang et al., 2018b); detailed description of the analyzers is discussed in Zhu et al. (2016). Surface HCHO concentrations were monitored using a formaldehyde analyzer (AERO LASER, Germany, Model 4021) based on fluorometric Hantzsch reactions (Gilpin et al., 1997; Rappenglück et al., 2010). All surface observations were collected as 1-min averaged data and processed to hourly mean values.

2.2 Satellite products

To evaluate the emissions and atmospheric chemistry in the NCP and greater East Asia, we used satellite observations of CO, NO_2, and HCHO for May and June 2016. The Measurements of Pollution In the Troposphere (MOPITT) instrument onboard the NASA Terra satellite retrieved CO column contents with ~10:30 am local overpass time (Deeter et al., 2003). We used the latest version 7 MOPITT Level 3 daily gridded average products (1° × 1° spatial

resolution, available at https://eosweb.larc.nasa.gov/project/mopitt/mop03j_v007) for the ARIAs campaign period (MOPITT Science Team, 2013). MOPITT thermal-infrared and near-infrared (TIR + NIR) products shows improved sensitivity to near surface CO in China (Worden et al., 2010). We used MOPITT near surface CO (~ 900 hPa) products and related averaging kernels (AKs) to evaluate the CMAQ results (Deeter et al., 2012).

OMI, onboard the NASA Aura satellite, is a UV/Vis solar backscatter spectrometer in a polar sun-synchronous orbit with a ~1:35 pm local overpass time. With high spatial resolution (13 km × 24 km for the center at nadir) and nearly daily coverage, OMI has provided monitoring of trace gases and aerosol properties since 2005 (Levelt et al., 2006). The Version 3 OMI Level 2 NO_2 products (https://disc.gsfc.nasa.gov/datasets/OMNO2_V003/summary) (Krotkov et al., 2018) were used to evaluate the emissions and atmospheric chemistry in East Asia. Under clear sky, tropospheric NO_2 columns from OMI has precision of ~0.5×10^{16} molecules cm^{-2} and an accuracy of ±30% (Krotkov et al., 2017). OMI HCHO Version 3 Smithsonian Astronomical Observatory (SAO) (https://disc.gsfc.nasa.gov/datasets/OMHCHO_V003/summary) Level 2 products were used in this study (Chance, 2007; González Abad et al., 2015). The precision of column HCHO is ~1.0×10^{16} molecules cm^{-2} and SAO products have an accuracy of ±25-30% without cloud (Millet et al., 2006; Boeke et al., 2011). Data in OMI pixels affected by the row anomaly and contaminated by clouds were filtered out using quality flags for both NO_2 and HCHO columns.

2.3 Model set-up

We used CMAQ version 5.2 (EPA, 2017) to simulate atmospheric chemistry for the ARIAs campaign. The Weather Research and Forecasting (WRF) model Version 3.8.1 (Skamarock et al., 2008) was driven by the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim products (ds627.0, https://rda.ucar.edu/datasets/ds627.0) (Dee et al., 2011) to simulate meteorological fields. Two domains with spatial resolution of 36 km and 12 km (Fig. 1b) were used to cover East Asia, with 35 layers from the surface to 50 hPa and ~20 layers in the lowest 2 km. Major physical options in WRF include the Rapid Radiative Transfer Model (RRTM) radiation scheme (Clough et al., 2005), the Pleim-Xiu surface layer and land surface model (Pleim and Xiu, 1995; Xiu and Pleim, 2001), the Asymmetric Convective Model (ACM2) boundary layer scheme (Pleim, 2007), the Kain-Fritsch cumulus scheme (Kain, 2004),

and the WRF Single-Moment 6 (WSM-6) microphysics (Hong and Lim, 2006). The National Centers for Environmental Prediction (NCEP) ADP Global Surface and Upper Air Observational Weather Data (ds461.0 and ds351.0, https://rda.ucar.edu) were used to perform observational and analysis nudging on all domains following the method developed for NASA aircraft campaigns (He et al., 2014; Mazzuca et al., 2016). WRF outputs were processed by the EPA Meteorology-Chemistry Interface Processor Version 4.3 (MCIP v4.3, released in November 2015) for emission processing and CMAQ simulations.

Anthropogenic emissions were from the Emissions Database for Global Atmospheric Research Version 4.2 (EDGAR v4.2, 0.1° × 0.1° resolution) of year 2010, which are widely used for chemical transport modeling (European Commission, 2011). We used the EPA Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system Version 4.5 (UNC, 2017) to project EDGAR emissions to the modeling domain. Emissions of air pollutants were speciated into Carbon Bond 05 chemical mechanism (Yarwood et al., 2005) and updated AERO6 aerosol module (Appel et al., 2013). The EDGAR v4.2 inventory has emissions for energy, industry, residential, and transport sectors. Without stack height information for power plants in the energy sector, we followed the approach developed in He et al. (2012) to locate these anthropogenic emissions at ~200 m above the surface as an approximation for averaged stack height and plume rise. We used the United States Geological Survey (USGS) 24 category land use dataset combined with the Biogenic Emission Inventory System (BEIS) emission factors table to generate the input files for the CMAQ inline biogenic emissions modeling. Biogenic emissions were estimated using the BEIS module inline in CMAQ (EPA, 2017).

CMAQ v5.2 uses the updated Carbon Bond 6 (CB6r3) chemical mechanism (Yarwood et al., 2010) including improved chemistry mechanism for organic nitrates and peroxyacyl nitrates (PAN) chemistry and will lead to better performance for simulating Secondary Organic Aerosols (SOA) and tropospheric ozone in the United States (Appel et al., 2016). CMAQ was run with a coarse domain and a nested domain (Fig. 1b). Chemical initial and boundary conditions for the coarse domain were obtained from the default concentration profiles built in CMAQ (EPA, 2017). Results from the CMAQ coarse domain were used to generate boundary conditions for the nested domain. The WRF-CMAQ system was run from mid-April to June with the first 2 weeks as spin-up. Hourly concentrations of air pollutants were saved for further analysis and model evaluation.

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Atmospheric Chemistry and Physics Discussions

3. Results and discussion

3.1 Air Pollution in the NCP and CMAQ performance

Figure 2 summarizes all aircraft measurements of O_3, NO_2, CO, and CO_2 over the NCP from eleven research flights. Generally, we observed high concentrations of trace gases, such as >100 part per billion by volume (ppbv) of O_3, >20 ppbv of NO_2, >500 ppbv of CO, and >450 part per million by volume (ppmv) of CO_2, in the aircraft campaign area (defined as 36.5-38.5°N, 114.0-115.5°E hereafter). We conducted vertical spirals over XT (the A^2BC supersite), LC (the airport in south of Shijiazhuang), and two rural areas (JL and QZ) during the ARIAs research flights. Figure 3a summarizes vertical distributions and the mean profiles of air pollutants over XT, with mean O_3 concentrations of 80 ppbv in the lower atmosphere. We observed isolated plumes with >10 ppbv of NO_2, >1000 ppbv of CO, and >440 ppmv of CO_2 over XT, usually with a secondary maximum between 800 and 1200 m. These plumes aloft can play an important role in long-range transport of air pollutants to downwind regions. Profiles over LC (Fig. 3b) show higher O_3 concentrations (>100 ppbv) and relatively moderate NO_2 (~3 ppbv) and CO (~250 ppbv). The rural areas, JL and QZ, have relatively clean environment with <80 ppbv of O_3, <2 ppbv of NO_2, and <300 ppbv of CO (Fig. 3c and 3d). Even the concentrations of air pollutants over the rural region in the NCP are comparable or higher than values in urban areas in North America and Europe. In summary, we found the south-north and east-west gradients of air pollution, i.e., higher concentrations of air pollutants in the west XT-LC corridor near the mountain as compared with east side of JL and QZ. Thus, the ARIAs research flights have good coverage of regions with both high and moderate concentrations of air pollutants and can fairly represent the regional nature of air pollution over the NCP.

Comparison of the surface trace gas observations at the Xingtai supersite and the CMAQ simulations driven by the EDGAR inventory (named baseline CMAQ case hereafter) reveals that CMAQ generally underestimates concentrations of major air pollutants (Fig. S1 in the supplementary material). The baseline CMAQ run successfully captures the diurnal and daily variations of surface ozone in Xingtai, although consistently underpredicts its concentrations. For CO and NO_x, two important ozone precursors, CMAQ substantially underestimates their concentrations in Xingtai by more than 50% and especially fails to capture the extremely high values such as 6~7 ppmv of CO and ~100 ppbv of NO_x. This underestimation could be caused by local sources poorly represented in the 12-km model simulations. For ambient HCHO, an

273 important byproduct of VOC oxidization in ozone photochemical production, the baseline
274 CMAQ run captures the variations, but substantially underestimates its concentrations. These
275 results suggest that the underestimation of ozone precursors in CMAQ could lead to the poor
276 model performance of simulating tropospheric ozone and other pollutants.
277 Similar analyses were conducted to investigate air pollutant concentrations in the lower
278 troposphere over the NCP observed by the aircraft. A case of the research flight on June 11, 2016
279 (Fig. S2 in the supplementary material) shows that CMAQ well captures the vertical gradient of
280 air pollutants, while substantially underestimates concentrations of all trace gases except NO_y.
281 Following the approach described in Goldberg et al. (2016), we calculated the 10-min average
282 O_3, CO, NO, and NO_2 concentrations from aircraft measurements and compared them with the
283 baseline CMAQ simulations (Fig. 4) and found similar underestimation (50% to 75% for all air
284 pollutants) as compared with surface measurements (Fig. S1 in the supplementary material).
285 CMAQ overestimates NO_y but substantially underestimates NO and NO_2, which suggests that a
286 significant amount of reactive nitrogen compounds could exist in the format of organic nitrates
287 or nitrate aerosols in the model. Figure 5 compares total VOCs concentrations from WAS
288 samples and CMAQ simulations, indicating that VOCs levels are significantly underestimated by
289 80%. The model evaluation with surface and aircraft measurements suggest that in the NCP
290 ozone pollution has been significantly underestimated in the baseline CMAQ run, which could
291 be due to the uncertainty introduced by using the 2010 EDGAR emissions to simulate the 2016
292 ARIAs campaign period. Thus, we need to evaluate the emissions inventory data to improve the
293 CMAQ performance and investigate the sensitivity of ozone production.
294
295 **3.2 Evaluation of emissions inventory in the NCP**
296 The EDGAR v4.2 emission inventory in East Asia was created based on the 2010 MIX
297 emission inventory (Li et al., 2017a), so substantial changes were anticipated when used for the
298 ARIAs campaign in 2016. Anthropogenic emission inventories are usually based on the "bottom-
299 up" approach, which relies on the statistics of fossil fuel usage and emission factors (EFs) for
300 each sector defined as the ratio of the amount of air pollutants released by a unit of CO_2
301 emissions, e.g. CO/CO_2 and NO_x/CO_2. To evaluate the emission inventory data in the NCP, we
302 used a 60-s moving window and conducted linear regression of observed air pollutant (CO, NO_x,
303 etc.) concentrations vs. CO_2 concentrations, i.e. $\Delta CO/\Delta CO_2$ and $\Delta NO_x/\Delta CO_2$, defined as

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Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

emission enhancements (EEs). Through only selecting EEs that are in the PBL (below 1.5 km AGL in this study) and statistically significant ($R^2 > 0.6$), the values of EEs can act as a proxy of EFs in the air mass observed (Halliday et al., 2018).

EEs observed during the research flights have a broad range of values. $\Delta CO/\Delta CO_2$ ranges from below 1%, a typical value of modern automobile emissions, to higher than 10%, a value indicating fossil fuel combustion with high emissions such as biomass burning (Fig. 6a and 6b). The mean of observed EE for CO (3.7%) is close to that calculated from the EDGAR inventory (4.0%) in the aircraft campaign area. Observed $\Delta NO_x/\Delta CO_2$ ratios also have isolated high values (>0.1%) with a mean value of 0.05%, which is substantially higher than the EF (~0.03%) derived from the EDGAR inventory. Since estimation of anthropogenic CO_2 flux in an urban/suburban area is challenging (Cambaliza et al., 2014; Heimburger et al., 2017), the underestimation of CO and NO_x in the NCP could be caused by either underestimated EFs or uncertainty in anthropogenic CO_2 emission data used in the 'bottom-up' approach.

To further investigate the characteristics of air pollutant emissions in the NCP, we conducted a similar analysis of $\Delta NO_x/\Delta CO$, which are usually co-emitted in combustion processes. Since around half of the CO and NO_x are from mobile sources in the EDGAR emission inventory, this ratio can approximately represent the emission characteristic of mobile sources in the NCP. The mean observed $\Delta NO_x/\Delta CO$ ratio is ~1.3%, significantly lower than 5.6% based on the EDGAR emission inventory (Fig. 6c). These results suggest that the EDGAR emission inventory substantially overestimates the ratios of NO_x/CO, while the automobile emissions over the NCP in 2016 have been greatly improved due to recent regulations focusing on NO_x, i.e., EDGAR overestimates the contribution from combustion with high emissions. It is worth noting that we only evaluated the emission ratios (EEs or EFs) in the EDGAR inventory, while the underestimation of CO and NO_x emissions could be caused by inaccurate CO_2 emissions which have not been examined in this study.

3.3 Evaluation of CO, NO_x, and VOCs emissions using satellite data

Satellite observations are widely used to evaluate the anthropogenic emissions in East Asia sometimes supplemented by model simulations, e.g., CO emissions using the MOPITT CO products (Jiang et al., 2015; Zheng et al., 2018), anthropogenic NO_x emissions using OMI NO_2 products (Wang et al., 2012; de Foy et al., 2015; Qu et al., 2017), and VOCs emissions using

OMI HCHO products (Stavrakou et al., 2016). In this study, we used measurements from multiple satellite instruments to evaluate the CMAQ performance of NO_2, HCHO, and CO. Since NO_2 and HCHO can be treated as proxy of NO_x and VOCs emissions, we can further improve the 2010 EDGAR emissions over the NCP base on satellite data.

We followed the approach developed in Canty et al. (2015) to compare the tropospheric column contents of NO_2 from OMI products and CMAQ simulations. Level 2 OMI NO_2 swatch information including row anomaly and quality flags were used to sample NO_2 vertical profiles from CMAQ outputs, and then CMAQ NO_2 column was calculated using the OMI averaging kernel (AK). Lastly, we averaged OMI and CMAQ NO_2 column contents to create daily 0.25° × 0.25° Level 3 products (see details in Canty et al., 2015). A similar approach was used to integrate HCHO column contents from CMAQ simulations based on OMI HCHO retrievals (see details in Ring et al., 2018) and construct daily 0.25° × 0.25° Level 3 HCHO products. For tropospheric CO, we selected the CO concentrations at ~ 900 hPa in CMAQ and averaged them to 1.0° × 1.0° daily products using MOPITT CO averaging kernel (MOPITT Science Team, 2013). All gridded daily data of satellite and CMAQ were averaged in May and June 2016 for comparison.

Figure 7a shows strong signals over the NCP of the OMI NO_2 observations. CMAQ underestimates NO_2 columns over the aircraft campaign area, and only predicts 81% of NO_2 column as compared with OMI observations. However, in urban regions such as Beijing, the Yangtze River Delta (YRD), and the PRD, CMAQ substantially overestimates column NO_2 by up to 30%. Because the baseline CMAQ simulations used the 2010 anthropogenic emission data, these differences should reflect the changes in NO_x emissions due to recent air pollution regulations. The comparison of NO_2 column suggests that NO_x pollution of megacities in China has been substantially improved after 2010 while NO_x pollution in smaller cities and rural area has worsened, consistent with results from independent studies using OMI (Duncan et al., 2016; Krotkov et al., 2016). OMI HCHO retrievals also show high values over the NCP in spring when plants' photosynthetic activity is relatively low, reflecting that the domination of anthropogenic VOCs emissions in north China (Zhao et al., 2017). CMAQ has good agreement with OMI HCHO within the aircraft campaign area (<20% underestimation), but substantially underestimates HCHO columns in south China where biogenic VOCs dominate (Fig. 7b). The MOPPIT products show high near-surface CO concentrations over the eastern China (Fig. 7c),

while the baseline CMAQ run substantially underestimates CO concentrations over north China and only predicts 42% of the CO over the aircraft campaign area.

Using NO_2 and HCHO as proxies of NO_x and anthropogenic VOCs emissions, the comparison of satellite observations and the baseline CMAQ simulations suggests that both NO_x and VOCs emissions in the aircraft campaign area need to be adjusted for a better simulation of tropospheric ozone. Also, the underestimation of CO, as an important precursor, can lead to underprediction of tropospheric ozone. We calculated the model/satellite ratios of NO_x, HCHO, and CO in East Asia (Fig. 8) and used these ratios to adjust their anthropogenic emissions in CMAQ. The results will be discussed in Section 3.4.

3.3 Tropospheric ozone production sensitivity from OMI and CMAQ

Photochemical production of tropospheric ozone is highly non-linear and dependent on concentrations of NO_x and VOCs (Kleinman, 1994; Sillman, 1999; Kleinman, 2000). A maximum rate of ozone production can be achieved with an optimal VOCs/NO_x ratio. With other VOCs/NO_x ratios, ozone production can be either in the VOC-sensitive regime (the rate of ozone production is controlled by VOCs concentrations) or in the NO_x-sensitive regime (the rate of production is controlled by NO_x concentrations). Different pollution control strategies can be implemented to reduce the tropospheric ozone levels in these two regimes. For instance, in a VOC-sensitive environment, reducing NO_x emissions will lead to limited effects until the ozone production has been changed to a NO_x-sensitive environment with the continuous removal of NO_x from the atmosphere. Duncan et al. (2010) developed an approach using OMI HCHO/NO_2 column ratio to estimate the ozone production sensitivity as: 1) HCHO/NO_2 <1: VOC-sensitive regime; 2) HCHO/NO_2 1~2: transition regime; 3) HCNO/NO_2 > 2: NO_x-sensitive regime. Studies show that urban areas in the U.S. such as Los Angeles, New York City and Houston are in VOC-sensitive or transition regimes, which lead to difficulty in local regulation of air quality (Duncan et al., 2010; Mazzuca et al., 2016; Ring et al., 2018). Recent studies suggest new threshold values of HCHO/NO_2 ratios between VOC-sensitive, transition, and NO_x-sensitive regimes in the U.S. (Jin et al., 2017; Schroeder et al., 2017).

Using the Duncan et al. (2010) approach, studies using OMI products suggest large areas of eastern China are either in VOC-sensitive regime (mostly megacities such as Beijing) or in transition regime (Jin and Holloway, 2015; Jin et al., 2017; Xing et al., 2018). We follow the

approach described in Ring et al. (2018) to calculate the column $HCHO/NO_2$ ratios from OMI observations and CMAQ simulations for East Asia. OMI column $HCHO/NO_2$ ratios suggest that the ozone photochemical production is VOC-sensitive or in transition region over the NCP and other major urban areas such as YRD and PRD (Fig. 9a) if the Duncan et al. (2010) approach is applicable for these areas. CMAQ successfully captured the spatial distribution of the regional nature of ozone production sensitivity in eastern China but predicted that the rate of ozone production is controlled more by VOCs with the CMAQ $HCHO/NO_2$ ratio lower than 1.0 in Beijing, YRD, and PRD (Fig. 9b). The VOC-sensitive environment suggests the rate of ozone photochemical production in the NCP is controlled not only by NO_x emissions, but also by VOCs emissions which currently lack regulations in China. With continuous reduction of anthropogenic NO_x emissions in China, VOCs controls might be efficient in these VOC-sensitive regions.

3.4 Improvements of tropospheric ozone simulation using satellite products

Results of the previous two sections show that the baseline CMAQ run substantially underestimates the concentrations of ozone and its major precursors in the NCP. To identify the individual and combined effects of the emission discrepancy of impacting major ozone precursors in the NCP, we designed a series of sensitivity experiments with emissions adjusted to satellite observations. Unlike the top-down approach using global chemical transport models such GEOS-Chem (Lin et al., 2010b; Qu et al., 2017), here we simply applied the ratios of air pollutant column contents from satellite observations and CMAQ simulations on each 0.25 degree grids (Fig. 8) as: CO_{CMAQ}/CO_{MOPITT}, NO_{2CMAQ}/NO_{2OMI}, and $HCHO_{CMAQ}/HCHO_{OMI}$ ratios for anthropogenic CO, NO_x, and VOCs emissions, respectively. To estimate the contribution from biogenic VOCs emissions, we conducted one more run with the in-line BEIS module turned off. Table 1 shows the emission adjustments for the five sensitivity experiments. CMAQ was run for the nested 12 km domain (D02) with the same meteorology, initial conditions, and boundary conditions derived from the coarse domain simulations.

Figure 10 presents the evaluation of surface observations with respect to two sensitivity experiments (CMAQ_baseline and CMAQ_all, comparison with all CMAQ runs are presented in Fig. S3 in the supplementary material). CMAQ still might not capture the extreme high values of surface O_3 and CO (Fig. 10a and 10b). For instance, the maximum CO concentration from

CMAQ simulations are ~1700 ppbv while surface observations have CO peaks higher than 6000 ppbv (Fig. 10b). The adjustments of the emission inventory have improved the model simulations of NO_2/NO (Fig. 10c and 10d) and HCHO (Fig. 10e). During the ARIAs flights, we observed various sources of emissions in the aircraft campaign area such as small factories and biomass burning, which are not included in the EDGAR emission inventory. Thus, the reason for the model underestimation could be that the spatial resolution (12 km) of the nested CMAQ domain cannot represent the detailed emissions and resolve the local air pollution hotspots. However it is worth noting that even our CMAQ system is still not capable to reproduce the surface air quality at Xingtai, the adjustments of EDGAR emissions based on satellite observations reduce the underestimation.

The ARIAs flights covered a large area (~10^4 km^2) in Hebei Province, which represent the regional nature of air pollution over the NCP. A case comparison of CMAQ_All case and Y12 measurements on June 11, 2016 (Fig. 11) shows better results in both concentrations and vertical gradient of air pollutants (compared with Fig. S2 in the supplementary material), indicating the effectiveness of improving the emission inventories based on satellite observations. Table 2 summarizes the model performance of CMAQ as compared with aircraft measurements. The adjustments of the EDGAR emissions with satellite observations moderately improved simulations of ozone pollution, with the root mean square error (RSME) decreasing from 25.1 ppbv (the baseline case) to 21.2 ppbv (CMAQ_All case) and the mean ratio of CMAQ simulations to aircraft observations increasing from 0.75 to 0.82. The model performance of CO has also been improved, with the RMSE decreasing from 247.0 ppbv to 203.6 ppbv and the mean ratio increasing from 0.40 to 0.66. For nitrogen compounds including NO_2, NO, and NO_y, the adjustments of EDGAR emissions have small impacts on improving the CMAQ performance. The reason could be that the ozone photochemistry is mainly VOC-sensitive over the NCP, so the adjustments of NO_x emissions have limited impacts close to sources.

4. Conclusions and Discussion

The ARIAs campaign conducted aircraft measurements over the NCP and observed high concentrations of air pollutants including O_3, CO, and NO_x. CMAQ simulations driven by the 2010 EDGAR emissions substantially underestimate the levels of ozone and its precursors in the campaign region. Analysis of emission enhancements of CO and NO_x with respect to concurrent

459 CO_2 measurements suggests that the usage of the 2010 EDGAR emissions for the 2016 ARIAs
460 campaign could introduce substantial uncertainty due to the recent changes of anthropogenic
461 emissions in China. Comparison of atmospheric columns of NO_2 from CMAQ simulations and
462 satellite observations suggests that NO_x emissions decreased in megacities such as Beijing and
463 Shanghai but increased in rural areas from 2010 to 2016. Similar analysis of HCHO and CO
464 shows that the EDGAR VOCs and CO emissions could also be underestimated in the NCP.
465 $HCHO/NO_2$ column ratio from OMI observations indicates tropospheric ozone production is
466 mainly in the VOC-sensitive regime in the NCP, which is also confirmed by CMAQ simulations.
467 To test a hypothesis that the poor model performance is due to emission biases, we adjusted the
468 EDGAR emissions over East Asia based on satellite observations. Better performance of
469 simulating ozone and its precursors is achieved, while underestimation still exists.

470 Both satellite observations and CMAQ simulations indicate that the VOC-sensitive
471 chemistry dominates the ozone photochemical production in eastern China, so the rate of local
472 ozone production is mainly controlled by the VOCs emissions. In the past few years, despite
473 implementation of control measures mainly on SO_2 and NO_x, ozone concentrations have
474 increased in China. Our study indicated that high NO_x concentrations were pervasive in the PBL
475 over rural areas of the NCP, where anthropogenic VOCs were also abundant. Reducing NO_x
476 emissions is essential to control ozone on the regional scale, but our model simulations indicated
477 that reducing VOCs emissions can lower the rate of photochemical smog production.

478 Currently, studies and regulations on anthropogenic VOCs emissions in China are
479 lacking, so with expectation of further decreasing NO_x emissions in China, more severe ozone
480 pollution could be anticipated. It is worth noting that while VOCs controls can have beneficial
481 impact on the local rate of ozone production in the VOC-sensitive regime, the ozone levels will
482 not decrease until NO_x emissions are substantially lower, i.e., regulations on VOCs are needed as
483 well as the continuous controls on NO_x emissions in China. These results can also partially
484 explain why ozone pollution emerged in the past few years while $PM_{2.5}$ pollution has been
485 substantially improved with strict regulations on anthropogenic emissions. New datasets such as
486 the updated 'bottom-up' emissions inventory for East Asia and high resolution satellite
487 observations such as TROPOMI and GEMS products are needed to improve the modeling of
488 ozone pollution in China, which can provide scientific evidence for future national and
489 international regulations on air quality.

Author contribution

X.R., R.D., H.H. and Z.L. designed the aircraft campaign; H.H., X.R., F.W., X.D., and F.L. performed the research flights; Y.W., X.R., and T.Z. conducted the surface observations; H.H., T.P., and Y.H. developed the modeling system; H.H., X.R., and S.B. analyzed the data; H.H., X.R., S.B. and R.D wrote the paper

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Disclaimer

The scientific results and conclusions, as well as any views or opinions expressed herein, are those of the author(s) and do not necessarily reflect the views of NOAA or the Department of Commerce.

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Atmospheric Chemistry and Physics Discussions

926 **Tables and Figures**

927

928 **Table 1.** List of CMAQ simulations with adjusted emissions based on satellite observations.
929 Anthropogenic CO, NO_x, and VOCs emissions were adjusted using MOPITT CO, OMI NO_2, and
930 OMI HCHO observations.

931

Run NO.	Experiment Name	Bio. VOCs	Anthro. CO	Anthro. NO_x	Anthro. VOCs
1	CMAQ_baseline	BEIS	EDGAR	EDGAR	EDGAR
2	CMAQ_noBEIS	N/A	EDGAR	EDGAR	EDGAR
3	CMAQ_CO	BEIS	Adjusted	EDGAR	EDGAR
4	CMAQ_NO_x	BEIS	EDGAR	Adjusted	EDGAR
5	CMAQ_VOCs	BEIS	EDGAR	EDGAR	Adjusted
6	CMAQ_All	BEIS	Adjusted	Adjusted	Adjusted

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Atmospheric Chemistry and Physics Discussions

933 **Table 2.** Statistics of CMAQ performance of six sensitivity experiments compared with ARIAs
934 aircraft measurements over the NCP.
935

No.	Name	Mean Diff	Slope	Stdev	Corr. R	NMB	NME	RMSE	Mean Ratio
		ppbv	Unitless	ppbv	Unitless	%	%	ppbv	Unitless
O_3									
1	CMAQ_Baseline	-21.35	0.56	13.25	0.37	-25.14	25.86	25.10	0.75
2	CMAQ_noBEIS	-23.74	0.49	12.85	0.41	-27.94	28.34	26.97	0.72
3	CMAQ_NOx	-19.83	0.59	13.63	0.34	-23.34	24.18	24.03	0.77
4	CMAQ_VOCs	-19.26	0.66	13.66	0.36	-22.67	23.81	23.58	0.77
5	CMAQ_CO	-20.35	0.61	13.52	0.36	-23.96	24.83	24.40	0.76
6	CMAQ_All	-15.18	0.81	14.83	0.33	-17.87	20.33	21.18	0.82
CO									
1	CMAQ_Baseline	-183.56	0.21	165.92	0.23	-60.26	60.26	246.98	0.40
2	CMAQ_noBEIS	-186.34	0.21	165.52	0.25	-61.17	61.17	248.79	0.39
3	CMAQ_NOx	-184.25	0.21	165.76	0.24	-60.48	60.50	247.39	0.40
4	CMAQ_VOCs	-181.89	0.22	166.32	0.22	-59.71	59.78	246.01	0.40
5	CMAQ_CO	-148.55	0.36	167.90	0.22	-48.76	50.32	223.67	0.51
6	CMAQ_All	-104.45	0.52	175.48	0.21	-34.29	45.03	203.60	0.66
NO_2									
1	CMAQ_Baseline	-1.72	0.31	3.09	0.58	-59.91	64.59	3.52	0.40
2	CMAQ_noBEIS	-1.73	0.31	3.09	0.58	-60.47	64.90	3.52	0.40
3	CMAQ_NOx	-1.45	0.38	2.99	0.60	-50.61	61.26	3.31	0.49
4	CMAQ_VOCs	-1.76	0.31	3.10	0.58	-61.60	65.66	3.55	0.38
5	CMAQ_CO	-1.70	0.31	3.09	0.58	-59.28	64.20	3.51	0.41
6	CMAQ_All	-1.47	0.38	3.01	0.59	-51.23	61.62	3.33	0.49
NO									
1	CMAQ_Baseline	-0.25	0.99	0.47	0.68	-32.23	45.4	0.53	0.68
2	CMAQ_noBEIS	-0.24	1.02	0.48	0.68	-31.09	45.66	0.54	0.69
3	CMAQ_NOx	-0.08	1.31	0.59	0.67	-9.75	50.01	0.59	0.90
4	CMAQ_VOCs	-0.30	0.89	0.45	0.68	-38.63	46.58	0.54	0.61
5	CMAQ_CO	-0.26	0.96	0.47	0.68	-33.08	45.26	0.53	0.67
6	CMAQ_All	-0.16	1.13	0.52	0.67	-20.31	45.46	0.54	0.80
NO_y									
1	CMAQ_Baseline	-15.26	0.30	10.15	0.39	-77.58	77.58	18.27	0.22
2	CMAQ_noBEIS	-15.50	0.29	10.15	0.40	-78.81	78.81	18.47	0.21
3	CMAQ_NOx	-14.24	0.37	10.20	0.37	-72.39	72.39	17.46	0.28
4	CMAQ_VOCs	-15.23	0.30	10.16	0.39	-77.42	77.42	18.25	0.23
5	CMAQ_CO	-15.26	0.30	10.15	0.39	-77.56	77.56	18.27	0.22
6	CMAQ_All	-14.21	0.37	10.20	0.37	-72.26	72.26	17.44	0.28

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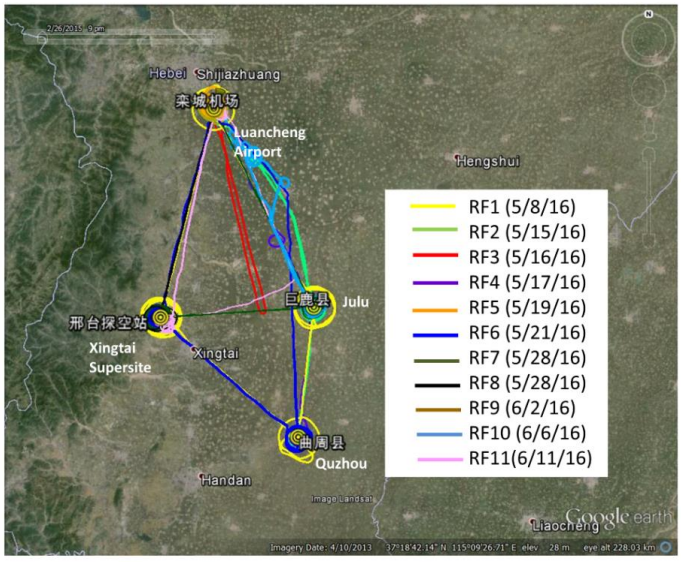
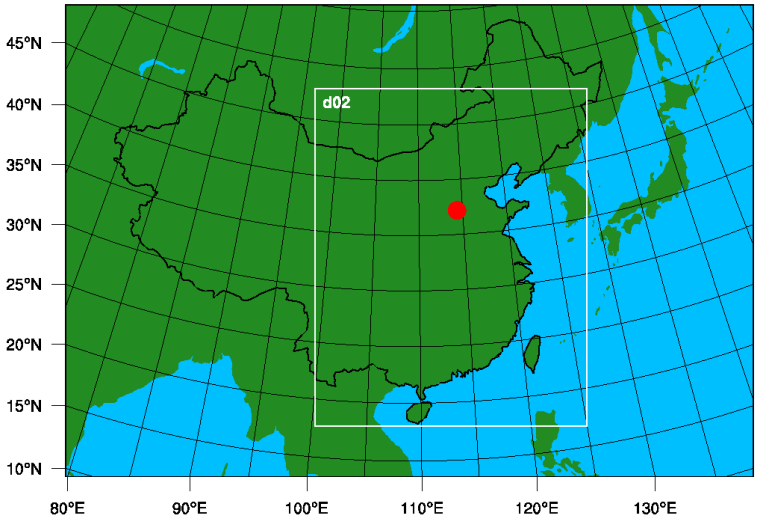
Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-248
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

937 **Figure 1.** ARIAs flights over the NCP and the WRF-CMAQ domains. Eleven Research Flights
938 (RF) were conducted in May to Mid-June 2016. CMAQ has two domains, the coarse domain
939 (d01, 36 km resolution) covering East Asia and the nested domain (d02, 12 km resolution)
940 focusing on eastern China. a) Summary of flight routes; b) WRF-CMAQ modeling domain (the
941 red dot represents the location of the Xingtai supersite).
942 a)



943
944 b)



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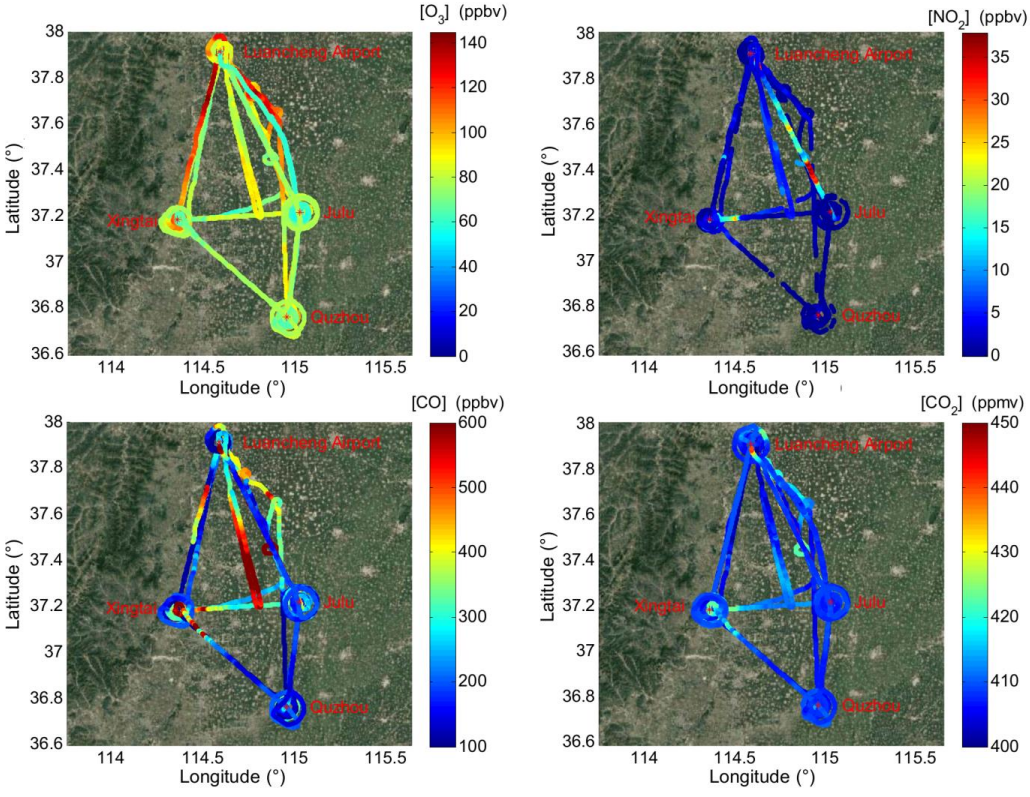
Discussion started: 29 March 2019

946 **Figure 2.** Summary of air pollutant concentrations in the NCP observed by Y12 aircraft. a) O_3, b)
947 NO_2, c) CO, and d) CO_2.
948



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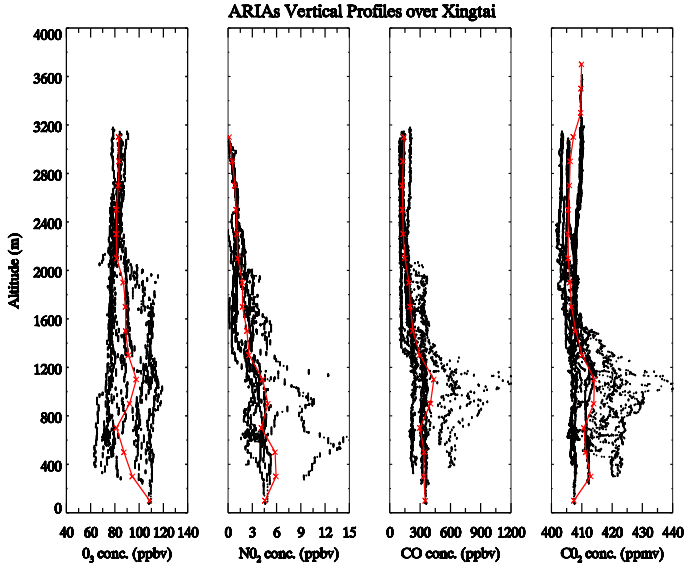
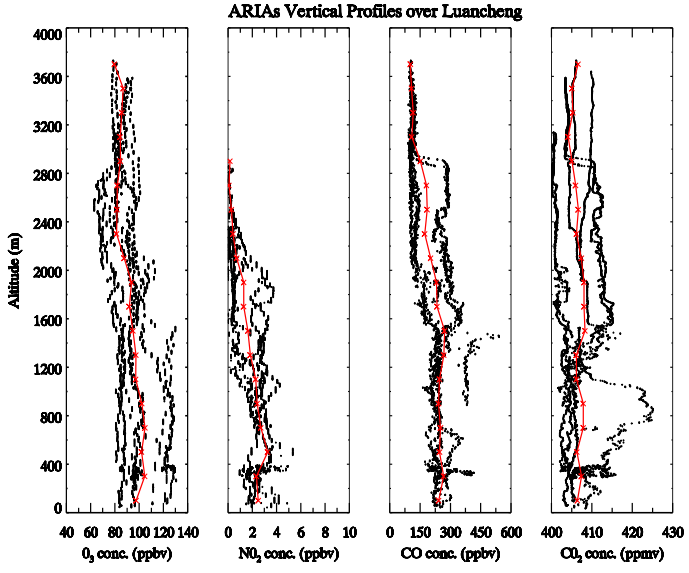
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

951 **Figure 3.** Vertical profiles of air pollutants over four locations in the NCP. a) Xingtai (XT), b)
952 Luancheng (LC), c) Julu (JL), and d) Quzhou (QZ). Red lines show the mean profiles.
953 a)

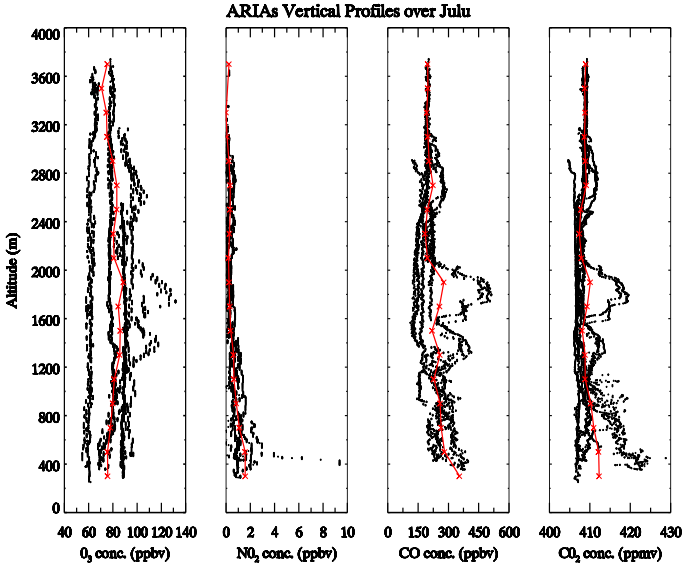


954
955 b)

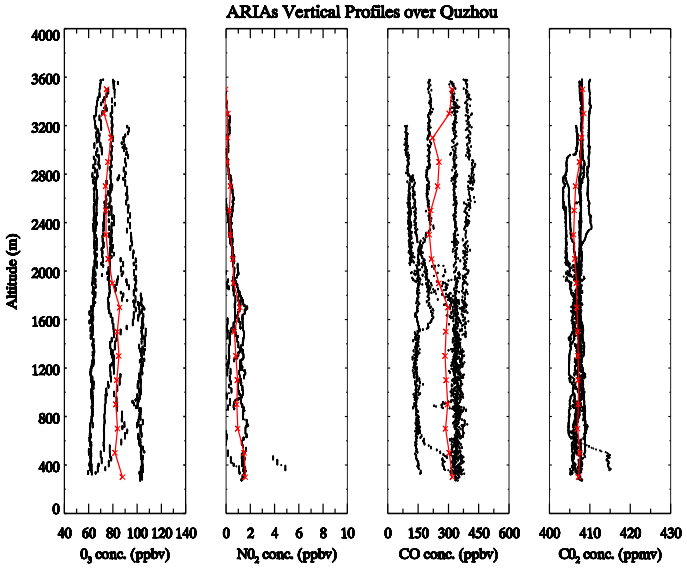


956

957 c)



958
959 d)



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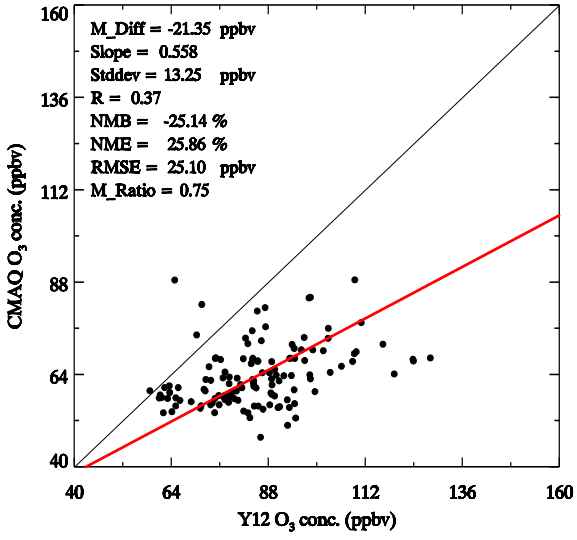
Discussion started: 29 March 2019

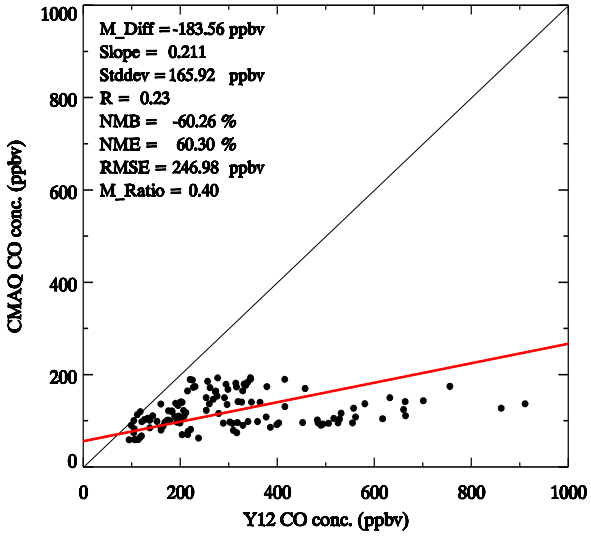
Atmospheric Chemistry and Physics Discussions

961 **Figure 4.** Comparison of 10-min averaged aircraft data and CMAQ simulations from 11 ARIAs
962 research flights. a) O_3, b) CO, c) NO, and d) NO_2. Black line shows the 1:1 ratio; red line stands
963 for the linear regression fitting line. M_Diff: mean difference; R: correlation; NMB: normalized
964 mean bias; NME: normalized mean error; RMSE: root-mean square error; M_Ratio: mean ratio.
965 a)



966

967 b)



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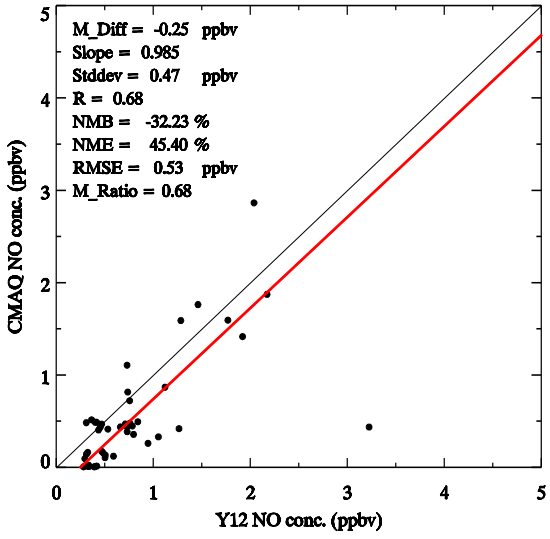
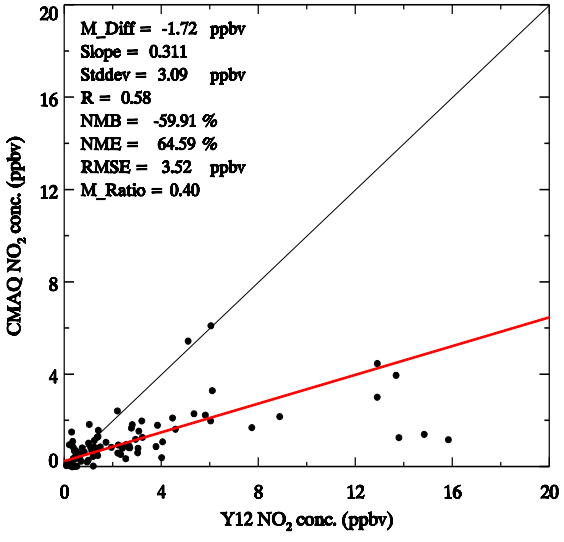
969 c)



970

971 d)



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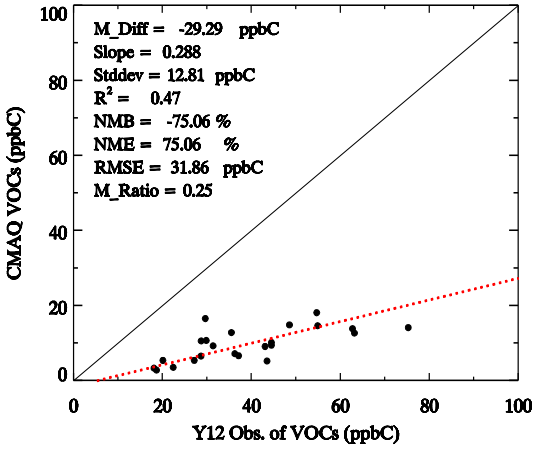
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

973 **Figure 5.** Comparison of total VOCs concentrations from WAS samples and CMAQ simulations.
974 Values are in unit of parts per billion Carbon (ppbC). Black line shows the 1:1 ratio; red line
975 stands for the linear regression fitting line. M_Diff: mean difference; R: correlation; NMB:
976 normalized mean bias; NME: normalized mean error; RMSE: root-mean square error; M_Ratio:
977 mean ratio.



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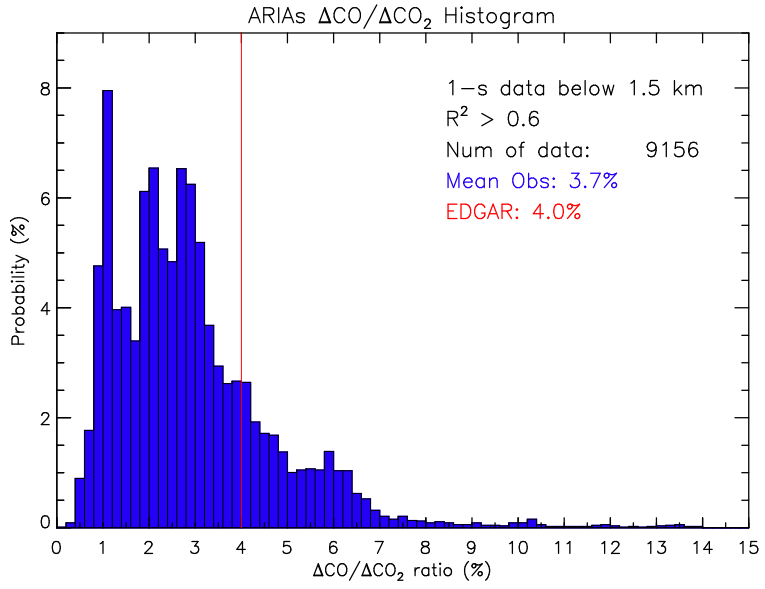
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 29 March 2019

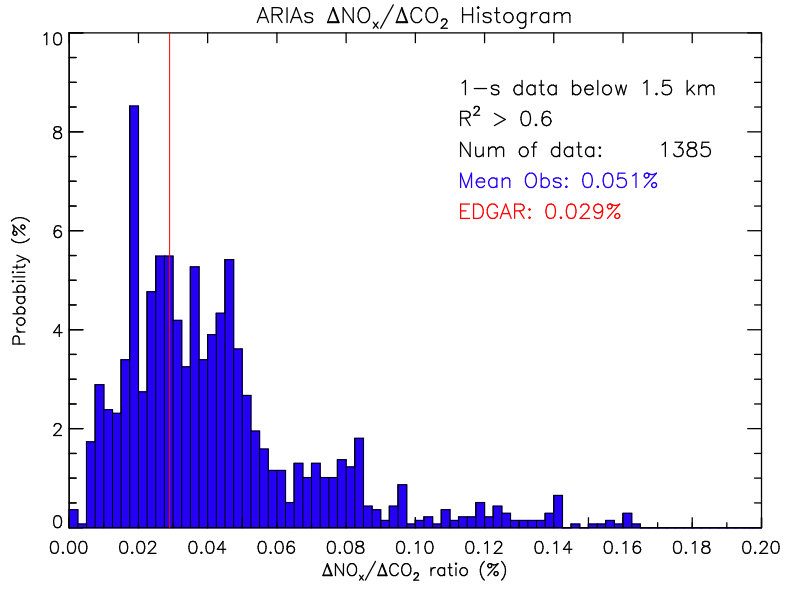
Atmospheric Chemistry and Physics Discussions

Figure 6. Comparison of emission enhancements (EEs) from the ARIAs campaign and emission factors (EFs) from the EDGAR emission inventory. a) $\Delta CO/\Delta CO_2$, b) $\Delta NO_x/\Delta CO_2$, c) $\Delta NO_x/\Delta CO$. Blue histogram shows the distribution of EEs observed by the Y12 aircraft; red line shows the ratio calculated using the EDGAR anthropogenic emissions.

a)



b)



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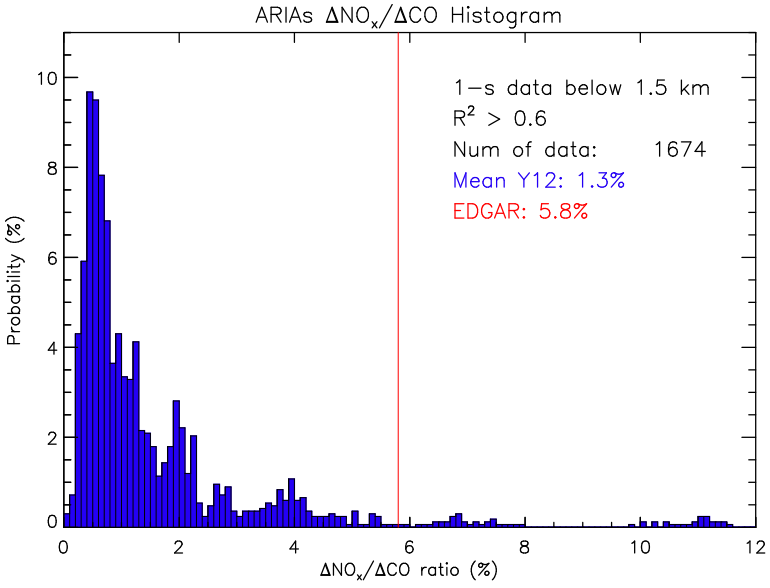
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

988 c)



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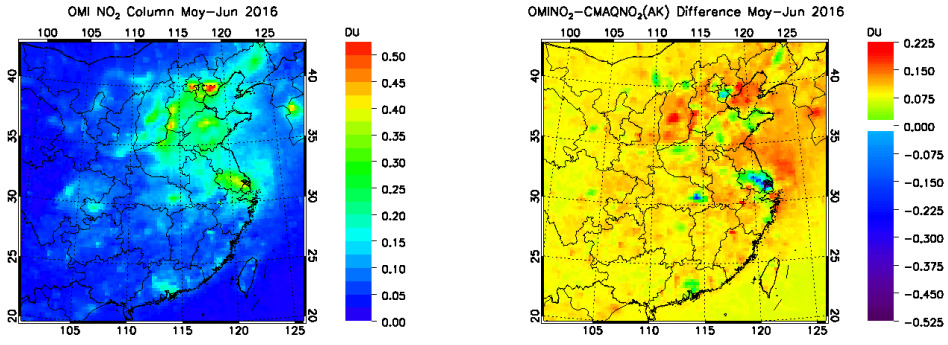
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 29 March 2019

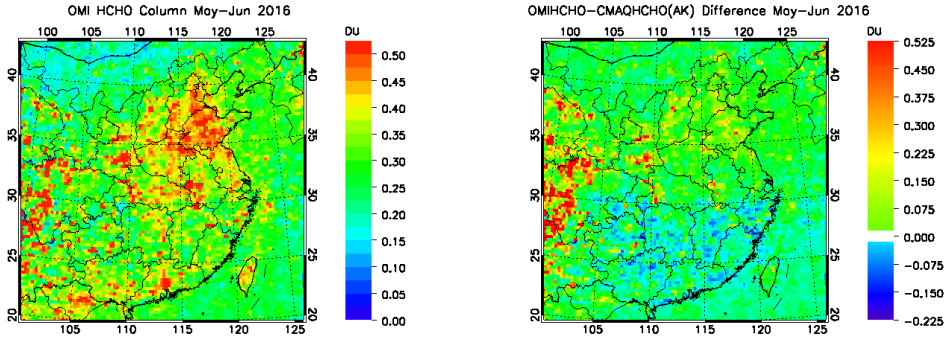
Atmospheric Chemistry and Physics Discussions

Figure 7. Comparison of air pollutants from satellite observations and CMAQ simulations. a) OMI NO_2 column (left) and the difference between OMI and CMAQ (right), Unit: Dobson Unit (1 DU = 2.69×10^{20} molecules/cm^2); b) OMI HCHO column (left) and the difference between OMI and CMAQ (right), Unit: DU; c) MOPITT near surface CO (left) and the difference between MOPITT and CMAQ (right), Unit (ppbv).

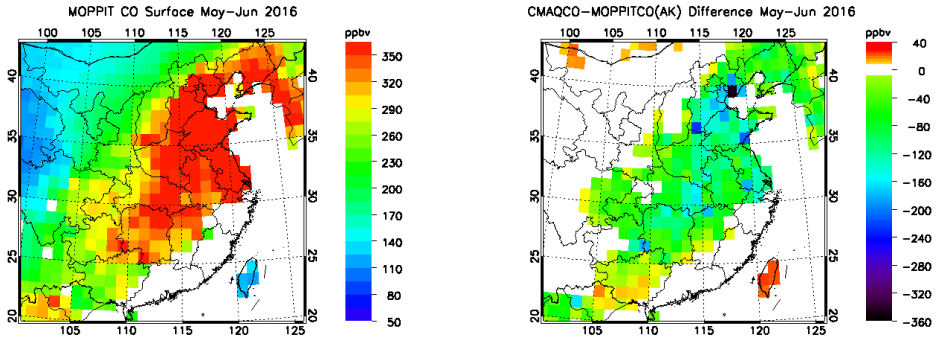
a)



b)



c)



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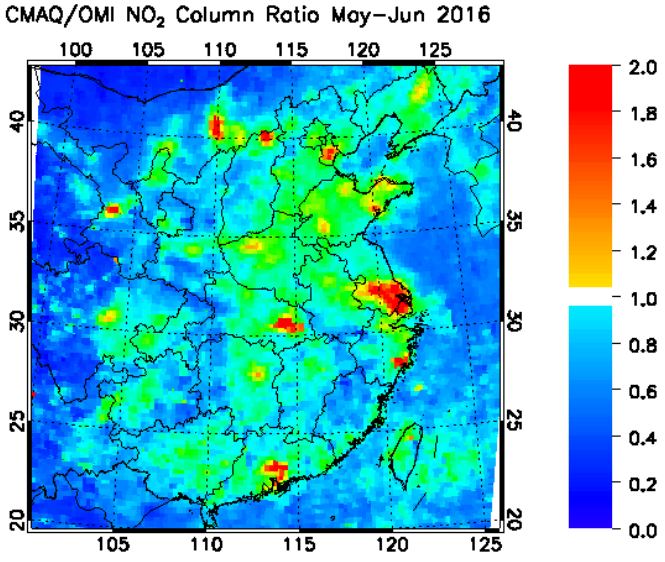
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

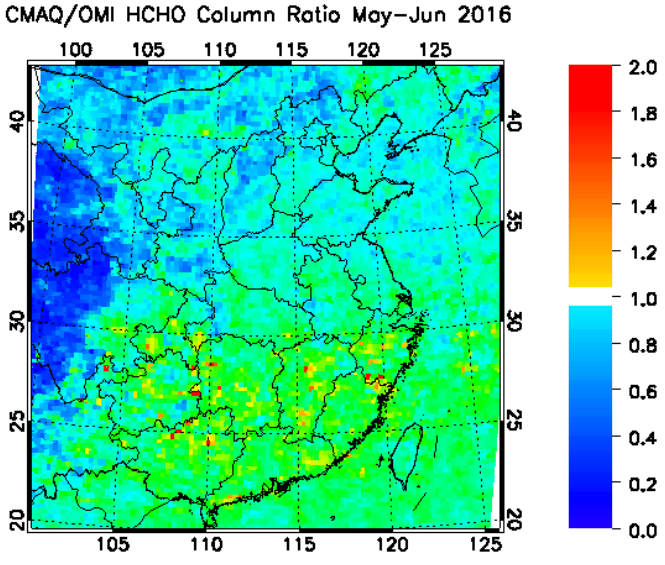
1002 **Figure 8**. Ratios of column contents of the baseline CMAQ simulations and satellite
1003 observations. a) CMAQ/OMI NO_2; b) CMAQ/OMI HCHO; c) CMAQ/MOPITT CO.

1004 a)



1005

1006 b)



1007

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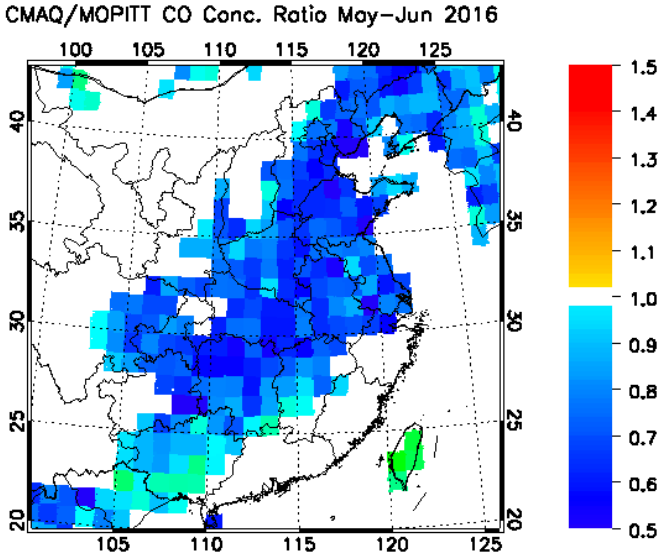
Discussion started: 29 March 2019

1008 c)



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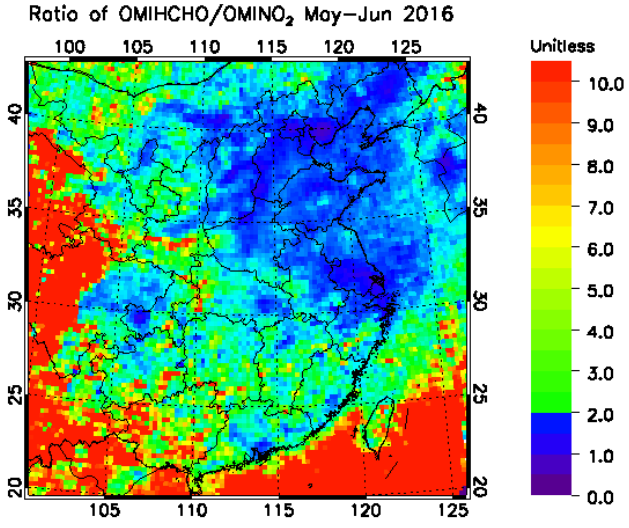
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

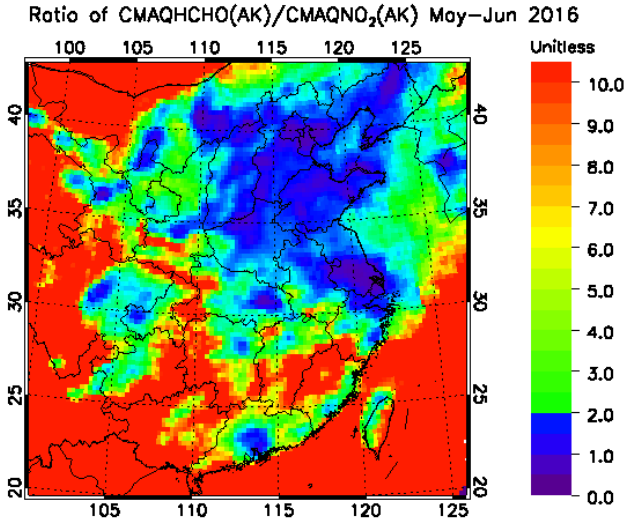
1010 **Figure 9.** Column HCHO/NO_2 ratios over East Asia in spring 2016. a) Ratio derived from
1011 collocated OMI HCHO and NO_2 observation; b) Ratio calculated from CMAQ simulations with
1012 OMI quality information and averaging kernel (AK).

1013 a)



1014

1015 b)



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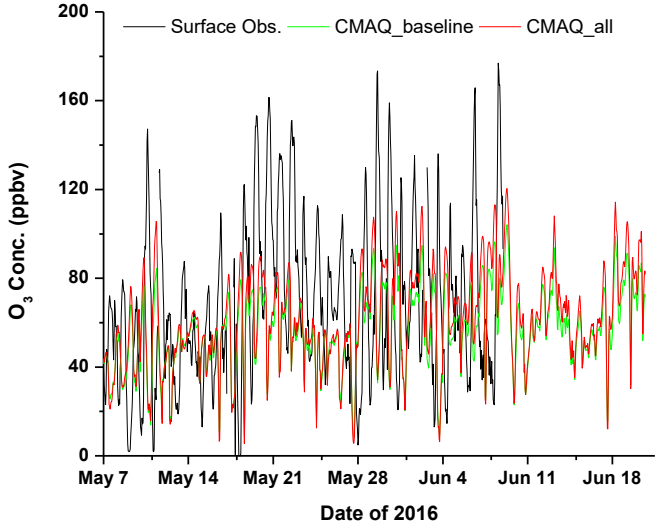
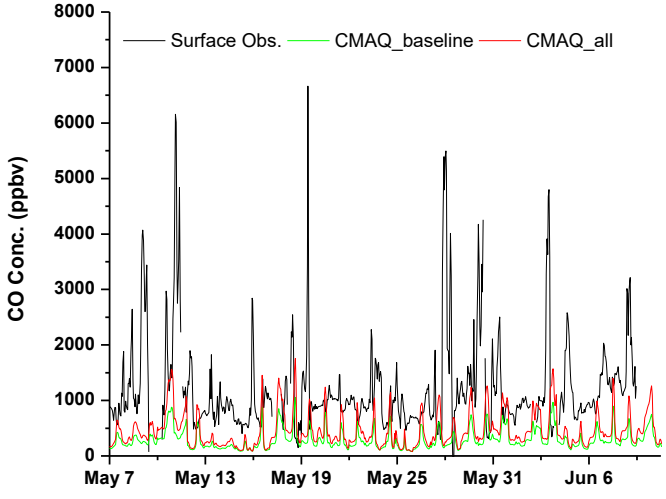
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

1017 **Figure 10.** Comparison of surface hourly observations of air pollutants and CMAQ simulations
1018 at the Xingtai supersite from May to mid-June 2016. a) O_3, b) CO, c) NO_2^*, d) NO_x, and e)
1019 HCHO. *Surface NO_2 is inferred as NO_x-NO from surface observations.
1020 a)



1021
1022 b)



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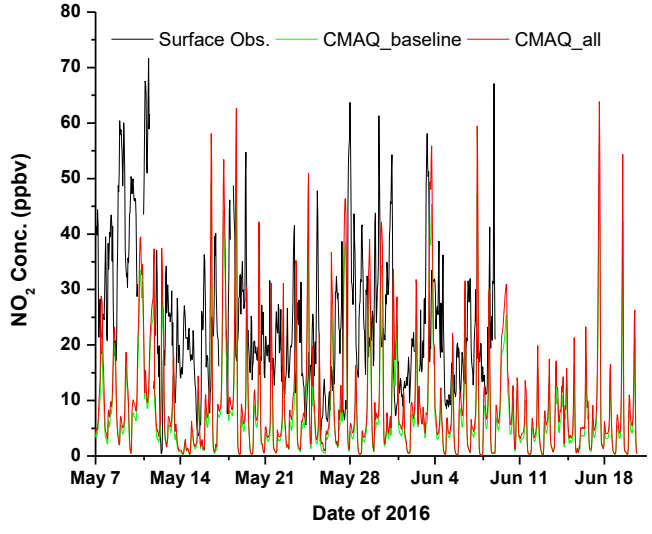
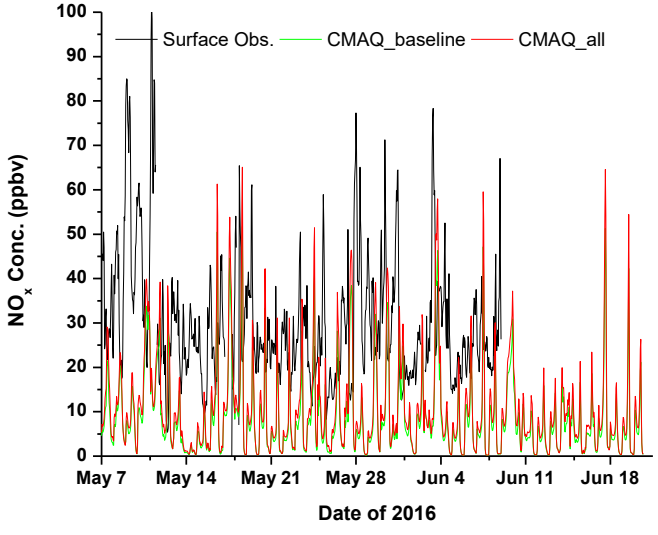
Discussion started: 29 March 2019

1024 c)



1025
1026 d)



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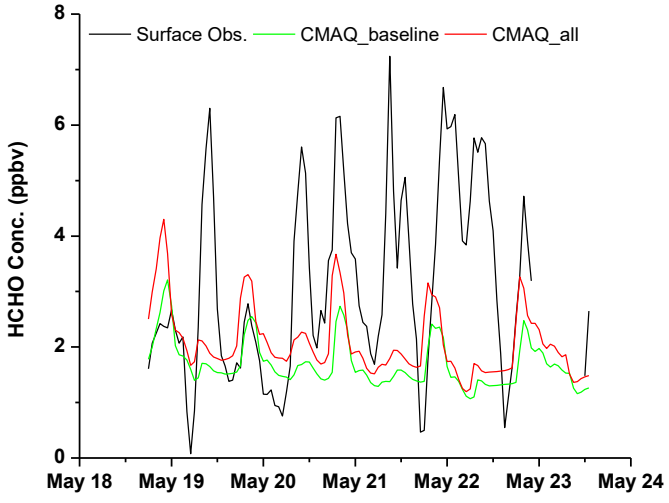
1029 e)



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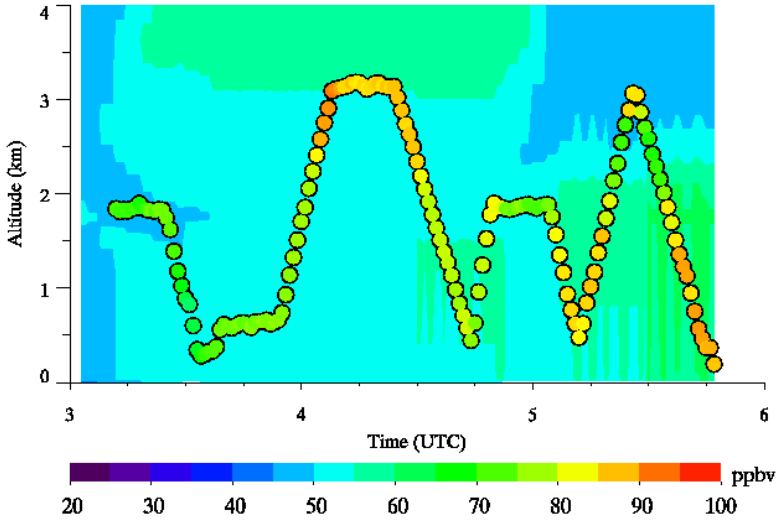
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

1031 **Figure 11.** A case study comparing aircraft observations and the CMAQ_All case results on June
1032 11, 2016. Background: CMAQ simulations. Overlay: 1 min Y12 measurements. a) O_3, b) CO, c)
1033 NO_2, d) NO, and e) NO_y.
1034 a)



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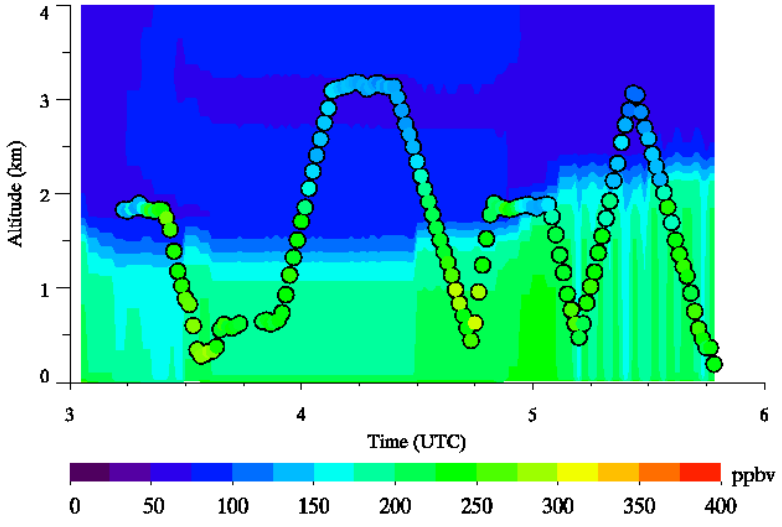
1036 b)



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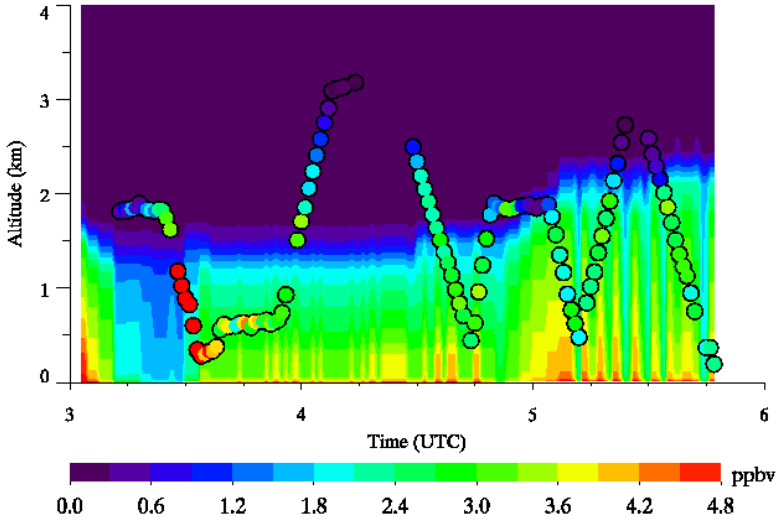
1038

1039 c)



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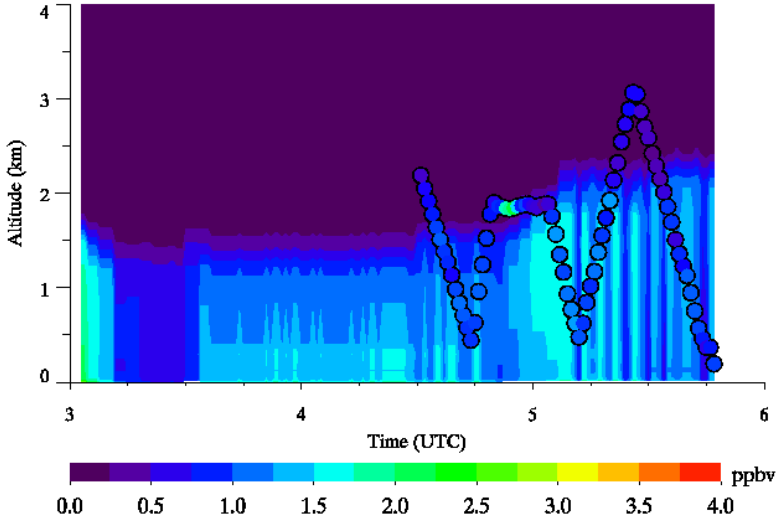
1041
1042 d)



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Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-248
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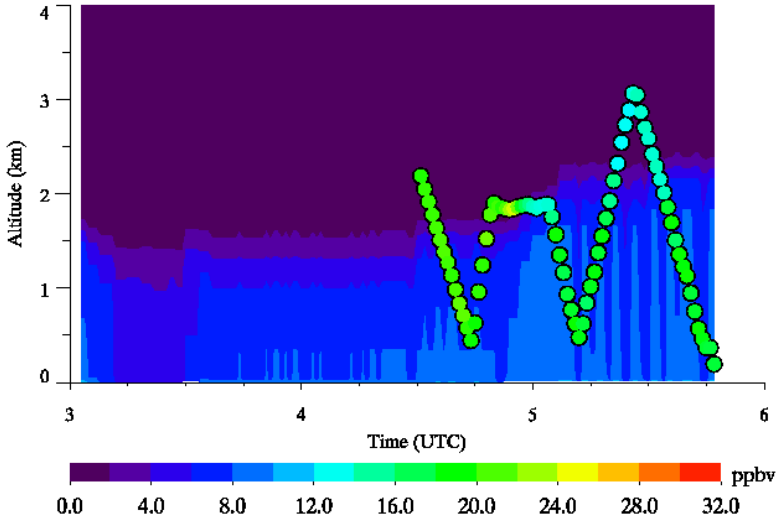
Discussion started: 29 March 2019

Atmospheric Chemistry and Physics Discussions

1045 e)



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