Wet removal of black carbon in Asian outflow: Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign

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[1] The Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign was conducted over East Asia in March–April 2009. During the A-FORCE campaign, 120 vertical profiles of black carbon (BC) and carbon monoxide (CO) were obtained in the planetary boundary layer (PBL) and the free troposphere. This study examines the wet removal of BC in Asian outflow using the A-FORCE data. The concentrations of BC and CO were greatly enhanced in air parcels sampled at 3–6 km in altitude over the Yellow Sea on 30 March 2009, associated with upward transport due to a cyclone with modest amounts of precipitation over northern China. In contrast, high CO concentrations without substantial enhancements of BC concentrations were observed in air parcels sampled at 5–6 km over the East China Sea on 23 April 2009, caused by uplifting due to cumulus convection with large amounts of precipitation over central China. The transport efficiency of BC (TE$_{BC}$, namely the fraction of BC particles not removed during transport) in air parcels sampled above 2 km during the entire A-FORCE period decreased primarily with the increase in the precipitation amount that air parcels experienced during vertical transport, although their correlation was modest ($r^2 = 0.43$). TE$_{BC}$ also depended on the altitude to which air parcels were transported from the PBL and the latitude where they were uplifted locally over source regions. The median values of TE$_{BC}$ for air parcels originating from northern China (north of 33°N) and sampled at 2–4 km and 4–9 km levels were 86% and 49%, respectively, during the A-FORCE period. These median values were systematically greater than the corresponding median values (69% and 32%, respectively) for air parcels originating from southern China (south of 33°N). Use of the A-FORCE data set will contribute to the reduction of large uncertainties in wet removal process of BC in global- and regional-scale models.


1. Introduction

[2] Black carbon (BC) particles have been recognized as one of the most important aerosols for climate forcing because they efficiently absorb solar radiation and lead to heating of the atmosphere [Hansen et al., 1997; Ackerman et al., 2000; Ramanathan et al., 2001; Jacobson, 2001, 2002, Menon et al., 2002; Koren et al., 2004; Wang, 2004; Ramanathan and Carmichael, 2008]. BC particles are emitted into the atmosphere by incomplete combustion of fossil fuels (diesel and coal), biomass, and biofuels. BC particles emitted at the ground surface are transported from the planetary boundary layer (PBL) to the free troposphere (FT) by uplifting processes, including warm conveyor belts (WCBs) and cumulus convection, followed by efficient horizontal transport on a regional-to-hemispheric scale by the westerlies. BC particles coated with sufficient water-soluble compounds are cloud condensation nuclei (CCN) active [e.g., Kuwata et al., 2009] and therefore can be efficiently removed from the atmosphere by precipitation during transport, while a small fraction of the remaining interstitial BC particles can be removed through collection of cloud or rain droplets [Seinfeld and Pandis, 2006].

[3] An understanding of the wet removal of BC is critically important because it directly controls vertical profiles of BC and amounts of BC transported from source regions to receptor regions (i.e., long-range transport). Previous modeling studies showed that the direct radiative forcing by aerosols depends strongly on the vertical profile of BC...
[Hansen et al., 2005], and some fraction of BC particles uplifted from the PBL to the FT over source regions is transported over long distances to receptor regions [Park et al., 2005; Koch and Hansen, 2005; Stohl, 2006; Koch et al., 2007; Hadley et al., 2007; Liu et al., 2011], such as the Arctic troposphere, exerting a substantial impact on regional-to-global-scale radiative forcing [Hansen and Nazarenko, 2004]. A multimodel comparison of general circulation models (GCMs) by Shindell et al. [2008] found large inter-model differences in BC concentrations calculated in the Arctic. Koch et al. [2009] also found large differences in vertical profiles of BC concentration between GCM simulations and observations. One of the main causes of these differences is considered to be large uncertainties in the wet removal process of BC adopted in aerosol models [Textor et al., 2006; Vignati et al., 2010]. Despite the importance of the wet removal process of BC, a quantitative understanding of this process is still limited.

[4] In recent years, removal of BC and its transport from source regions to receptor regions have been studied by several aircraft campaigns conducted over receptor regions, such as the NASA Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission in spring and summer 2008 [Jacob et al., 2010] and the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observations (HIPPO1) campaign in January 2009 [Schwarz et al., 2010]. In these aircraft campaigns, high-accuracy measurements of BC particles were made with single-particle soot photometer (SP2) instruments [Schwarz et al., 2008; Kondo et al., 2011a]. During ARCTAS, the median values of the transport efficiency of BC (TE_{BC}, namely the fraction of BC particles not removed during transport) in air parcels transported from Asia to the Arctic troposphere were estimated to be 13% and 0.8% in spring and summer, respectively [Matsui et al., 2011]. The quantity TE_{BC} was first defined by Koike et al. [2003], and the definition is slightly modified in this study as described in section 3.3. These Asian air parcels had undergone strong uplifting associated with WCBs being influenced by precipitation during transport. The differences in TE_{BC} of these air parcels between spring and summer were found to arise from seasonal differences in precipitation. During HIPPO1, vertical profiles of BC concentration over the Pacific Ocean were found to depend on latitude [Schwarz et al., 2010] and thus the wet removal of BC in global models might need to be evaluated separately in different latitudinal regions. However, these studies on the wet removal of BC were focused on receptor regions with rather coarse spatial resolution (i.e., on continental scales). In order to improve our understanding of the wet removal of BC, aircraft measurements closer to source regions, such as East Asia, with a higher spatial resolution are essential, because most BC particles are considered to be removed by precipitation near source regions within several days of transport.

[5] East Asia is the largest source of anthropogenic BC, according to current emission inventories [Streets et al., 2003; Bond et al., 2004; Zhang et al., 2009]. Over East Asia, several aircraft campaigns have been conducted, such as the Asian Aerosol Characterization Experiment (ACE Asia) in spring 2001 [Huebert et al., 2003] and the NASA Transport and Chemical Evolution over the Pacific (TRACE-P) in spring 2001 [Jacob et al., 2003]. During these campaigns, Uno et al. [2003] discussed transport processes of BC within the PBL and Park et al. [2005] estimated the atmospheric lifetime of BC during TRACE-P. However, studies on the wet removal of BC over East Asia were quite limited, because there were only very small amounts of data available on BC obtained by aircraft in the FT. In fact, there have been no aircraft observations with high-accuracy BC measurements covering the entire altitude range of the FT over East Asia since the TRACE-P mission in 2001, although some aircraft BC measurements were conducted in the lower troposphere, such as the Cheju ABC Plume-Monsoon Experiment (CAMPEX) in summer 2008 (0–4 km in altitude) [Ramana et al., 2010].

[6] The Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign was conducted over East Asia in March–April 2009 to investigate transport and removal processes of aerosols, their physical and chemical properties, and cloud microphysical properties in Asian outflow. During the campaign, 120 vertical profiles of BC particles were obtained using an SP2 instrument at 0–9 km in altitude. The major objective of this study is to understand the spatial distributions of BC over East Asia and the wet removal of BC in Asian outflow using the A-FORCE data. In particular, we focus on the importance of precipitation, which influences the removal rate of BC from the atmosphere. We also describe meteorological conditions during the A-FORCE period, which influenced the wet removal of aerosols.

2. Aircraft Measurements

[7] During the A-FORCE aircraft campaign, there were a total of 21 flights conducted over the Yellow Sea, the East China Sea, and the western Pacific Ocean between 18 March and 25 April 2009 using a King Air aircraft, operated by Diamond Air Service (DAS) Inc. (Figure 1 and Table 1). These flights were conducted over the area where concentrations of BC and carbon monoxide (CO) were high due to their transport from China (Figure 1). In situ measurements of gaseous and aerosol species and cloud microphysical properties were made onboard the King Air aircraft (Table 2). A brief description of the measurements used in this study is given below.

[8] The CO mixing ratio was measured with a vacuum ultraviolet (VUV) resonance fluorescence instrument (AL5002, Aero-Laser GmbH) with a time resolution of 1 s [Gerbig et al., 1999]. By calibrating this instrument during observational flights regularly, its accuracy was estimated to be 2%, and its precision was about 0.5% for 10-s average data.

[9] Aerosols in ambient air were introduced to the instruments mounted within the cabin using a forward-facing iso-kinetic inlet [McNaughton et al., 2007] attached to the fuselage (roof). The volume flow rate was controlled to maintain iso-kinetic flow through the inlet tip and minimize inertial enhancement of particle concentrations.

[10] Size distributions of BC particles, namely BC-containing particles excluding their coating materials (i.e., BC cores), were measured using the SP2 instrument based on the laser-induced incandescence technique with a time resolution of 1 s [Schwarz et al., 2006; Moteki and Kondo, 2007]. The SP2 instrument detected BC cores in the size range of 75–850 nm volume equivalent diameter, assuming
a density of 2.0 g cm\(^{-3}\) [Moteki and Kondo, 2010]. Total BC mass concentration was derived from the sum of the measured BC cores over the observed size range. The fitting of a lognormal function to the mass size distributions of BC cores showed that more than 90% of the BC mass in the lognormal mode was detected. The SP2 instrument also measured the coating thicknesses of BC-containing particles with diameters of 200–850 nm [Kondo et al., 2011b]. In addition to BC-containing particles, the size distributions of light-scattering (i.e., BC-free) particles were measured in the size range of 170–850 nm volume equivalent diameter. The refractive index of light scattering particles was assumed to be 1.52. Absolute uncertainties of BC particles and light-scattering particles were estimated to be within 10% (by volume) and 20% (by volume), respectively.

Figure 1. (a) Flight tracks for the King Air aircraft over East Asia during the A-FORCE aircraft campaign (18 March to 25 April 2009). Estimates of anthropogenic emissions of BC in the year 2006 at 0.5\(^{\circ}\) x 0.5\(^{\circ}\) resolution [Zhang et al., 2009] are shown in gray scale. Only the grid boxes in which BC emissions are greater than 0.01 Gg yr\(^{-1}\) are plotted with shading. (b) Enlarged map of the area enclosed within the blue solid line in Figure 1a. The red, green, and black lines denote the flight tracks during flight 8, flight 19, and the other flights, respectively.
et al., 2001] with a time resolution of 1 s (M. Koike et al., Measurements of regional-scale aerosol impacts on cloud microphysics over the East China Sea: Possible influences of warm sea surface temperature over the Kuroshio Ocean Current, submitted to Journal of Geophysical Research, 2012). The CAPS instrument was mounted under the wing of the King Air aircraft. The precision (1-s data) and absolute accuracy of the bulk LWC measurements used in this study were estimated to be 0.03 m g m⁻³ and 18%, respectively.

In this paper, we report aerosol concentrations at standard temperature and pressure (STP, 273.15 K and 1013.25 hPa). Only the data obtained outside of clouds were used for the purpose of the estimation of wet removal effects of BC. Cloud-free conditions were identified using the 1-min averaged LWC and ice water content (IWC) from the CAPS measurements. We excluded the BC data with a sum of LWC and IWC greater than 0.002 g m⁻³. Data influenced from local pollution at the airports was also excluded.

3. Methodology

3.1. Back Trajectories

Five-day kinematic back trajectories of air parcels measured onboard the aircraft were calculated every one minute based on the method described by Tomikawa and Sato [2005]. For calculating the trajectories, 6-hourly meteorological data from the National Centers for Environmental Prediction (NCEP) Final (FNL) operational global analysis were used, which are available on a regular grid with a resolution of 1° in both latitude and longitude at the 21 standard
Correlation between BC mass concentration and air parcels. The unit of the slope is ng m$^{-3}$/C0 (parts per billion by volume (ppbv)) [2011]. The value, D03204/D, is the value of the air parcels). Figure 2 shows the resulting BC with air parcels (1-min average data that was within the range of the Koike Park et al. – "air parcels are 4.84 ng m$^{-3}$). In the trajectory model, an TE craft below 2 km in altitude during A-FORCE (red circles). The red solid line is the linear regression line for the "dry PBL" air parcels. The unit of the slope is ng m$^{-3}$/ppbv$^{-1}$. For comparison, BC mass concentrations and CO mixing ratios for all the air parcels (1-min average data that match the back trajectories) sampled during A-FORCE are denoted by black circles. See the text for details.

We used the 3-D Weather Research and Forecasting (WRF) model [Skamarock et al., 2005] to estimate precipitation and PBL height. The model consisted of 120 $\times$ 72 grid boxes in the horizontal, each of which occupying an area of 81 $\times$ 81 km$^2$, covering the entire East Asian region. In the vertical direction, the model had 21 layers reaching up to the 100-hPa level, at finer intervals within the PBL (11 layers below a sigma level of 0.8). The WRF model simulation was performed for the period from 1 February 2008 through 31 May 2009 [Kondo et al., 2011c], with four-dimensional data assimilation ("FDFA nudging") where the circulation and thermal fields in the model were nudged toward the 6-hourly NCEP FNL data. A validation of the WRF precipitation in comparison with observations is presented in section 4.1.

3.2. Meteorological Model Simulation

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3.3. Definition of the Transport Efficiency of BC

[15] The $T_{BC}$ (transport efficiency of BC) value for sampled air parcels transported from source regions can be estimated from changes in observed BC-to-CO ratios, because emission sources of BC and CO are generally similar [Streets et al., 2003], and CO can be used as an inert combustion tracer within a timescale of a few weeks, although BC can be removed by wet deposition. The $T_{BC}$ is defined for individual air parcels as follows:

$$T_{BC} = \frac{\Delta[BC]}{\Delta[CO]}$$

where $\Delta[BC]$ and $\Delta[CO]$ are the differences between observed and background values of BC mass concentration and CO mixing ratio in individual air parcels, respectively, and $R_{BC-CO}$ is the value of the $\Delta[BC]/\Delta[CO]$ slope in air parcels that had not been influenced by wet processes during transport from the source regions (i.e., representing the emission ratio of BC to CO over the source regions). This approach is basically similar to that described by Koike et al. [2003], Park et al. [2005], and Matsui et al. [2011]. The definition of the "background value" of an air mass in this study is the concentration, which the air mass would have had if it had not been influenced by emissions over the East Asian region. The specific values are given near the end of this section. If a sampled air parcel is not influenced by wet removal during transport from the source region, the $\Delta[BC]/\Delta[CO]$ value of the air parcels does not change as long as the background concentrations of BC and CO are constant.

[16] Application of equation (1) requires the $R_{BC-CO}$ value, which has been estimated from the slope of the correlation between BC mass concentrations and CO mixing ratios observed for "dry PBL" air parcels (1-min average data that match the back trajectories). The "dry PBL" air parcels are defined by the following criteria: (1) air parcels sampled below 2 km in altitude outside of clouds, (2) their trajectories found in the lower troposphere (between the surface and the 700-hPa level) over the Asian continent (the longitudinal sector between 90°E and 122°E), and (3) the average RH of the highest 10% of RH values along the 5-day trajectory smaller than an RH value of 85%. The criterion (3) may be rather strict, but it was adopted to ensure exclusion of wet removal effects during transport, because the occurrence of precipitation always requires high RH conditions. At the same time, the particular threshold RH value (85%) is modest enough to ensure the sufficient number of data points for the "dry PBL" air parcels. Figure 2 shows the resulting BC and CO correlation for the "dry PBL" air parcels (based on 1-min average data, indicated by red circles in Figure 2) sampled during the A-FORCE campaign. The BC mass concentrations and CO mixing ratios for all air parcels sampled during A-FORCE are also shown for comparison (1-min average data, indicated by black circles). As indicated by the red line in Figure 2, the resulting $R_{BC-CO}$ value is 4.84 ng m$^{-3}$ (parts per billion by volume (ppbv))$^{-1}$ with 20% variability (i.e., the standard deviation of the $\Delta[BC]/\Delta[CO]$ values over all the “dry PBL” air parcels). The high BC-CO correlation ($r^2 = 0.95$) supports the validity of the identification of dry air parcels within the PBL. The $R_{BC-CO}$ value of 4.84 ng m$^{-3}$ was within the range of the values of the $\Delta[BC]/\Delta[CO]$ slopes (3.5–5.8 ng m$^{-3}$) observed in the Beijing area [Han et al., 2009] that contributed dominantly to the BC emissions in northeastern China [Guinot et al., 2007]. We note here that although Koike...
et al. [2003] used emission ratios of BC to CO instead of the $R_{BC-CO}$ value in equation (1), our approach has the advantage of excluding uncertainties in emission inventories. Uncertainties included in the $R_{BC-CO}$ and $TE_{BC}$ values are discussed in section 7.4.1.

[17] The background concentration of BC was assumed to be zero because the atmospheric lifetime of BC is estimated to be several days [e.g., Cooke et al., 2002; Park et al., 2005]. The background value of CO was chosen to be 118 ppbv, which is the value of the x-intercept of the regression line at the bottom of Figure 2. The median values of observed BC mass concentration (49 ng m$^{-3}$) and CO mixing ratio (120 ppbv) for the “free tropospheric” air parcels that remain above the 700-hPa level within the last 5 days prior to measurement (see section 7.2) are nearly the same as the background concentrations chosen in this study, confirming that our assumptions of the background concentrations are reasonable. We only use air parcels in which $\Delta[CO]$ values are greater than 30 ppbv for estimates of $TE_{BC}$ following Koike et al. [2003], in order to select air parcels that are clearly influenced by emissions over East Asia.

3.4. Estimates of Origins of Sampled Air Parcels

[18] To identify the origins of the air parcels sampled from the aircraft in the FT (above 2 km in altitude) during the entire A-FORCE period, we examined the geographical locations where the 5-day back trajectories of these air parcels first crossed the top of the PBL. The PBL height was derived from the WRF model simulation. This procedure is basically similar to that described by Oshima et al. [2004]. These trajectory-crossing locations are hereafter referred to as “uplifted locations” or “origins” of the air parcels. We also refer to air parcels whose trajectories crossed those locations as “uplifted air parcels” or for simplicity “sampled air parcels” hereafter. The origins of sampled air parcels are discussed in sections 7.1 and 7.2.

3.5. Estimates of Precipitation Influencing Sampled Air Parcels

[19] In general, it is difficult to evaluate the influence of precipitation on wet removal of aerosols from air parcels during vertical transport based on daily precipitation data, because the timescale of the wet removal of aerosols is considered to be smaller than a day. Since no high-resolution observational data of precipitation at intervals of a few hours are available over the entire region of East Asia, we use hourly output from the 3-D WRF model simulation.

[20] For our evaluation of the influence of precipitation on the air parcels sampled in the FT (above 2 km in altitude) during transport, we utilized the results of the 5-day back trajectories of the “uplifted air parcels” in combination with the precipitation water content (i.e., sum of rain, snow and graupel contents) available in the WRF 3-D output and precipitation available in the WRF surface output. Specifically, we checked the precipitation water content in each WRF 3-D grid box along each of the trajectories at hourly intervals. The surface precipitation amount was then integrated in the Lagrangian sense along each trajectory when and where a trajectory had passed through the precipitation water content grid box anywhere from the “uplifted location” to the sampling point (the flight track). The amount of integrated precipitation is hereafter referred to as “accumulated precipitation along trajectory (APT),” which gives a measure of the influence of precipitation on the wet removal of aerosols from air parcels during transport. This approach is similar to that described by Matsui et al. [2011]. The use of the WRF 3-D precipitation water content is necessary for identifying an air parcel influenced by precipitation during transport, while the use of only surface precipitation data cannot identify the altitude relationship between air parcels along trajectories and the occurrence of precipitation (e.g., we cannot exclude the case that air parcels are above precipitating clouds).

4. Meteorological Conditions During the A-FORCE Period

4.1. Spatial Distributions of Mean Meteorological Fields

[21] Mean meteorological fields during the A-FORCE period (17 March through 30 April 2009) based on NCEP FNL data are shown in Figures 3a–3d. The low-level circulation (850-hPa level) over East Asia during the period was characterized by two types of airflow in the course of the seasonal transition from winter to summer. The midlatitude region (30°–50°N), including northern China, the Yellow Sea, the East China Sea, and areas around Japan, was still under the influence of the modest monsoonal northwesterly flow associated with the weakening Siberian high (Figure 3a). In contrast, the subtropical region (15°–30°N), including southern China and the South China Sea, was under the influence of the persistent southerlies associated with a strengthening subtropical anticyclone over the central North Pacific (Figure 3a). As shown in Figure 3b, the low-level southerly flow advected warm, moist air into southern/central China and the western Pacific Ocean. Part of that flow was confluent to the south of Japan with the prevailing northwesterlies from the continent (Figure 3a). The rest of the southerlies were converging into a frontal zone (as denoted by a heavy solid line in Figures 3a and 3b) that extended zonally along the Yangtze River (around 30°N). The frontal zone was marked by a sharp meridional gradient in lower-tropospheric equivalent potential temperature ($\theta_e$) (Figure 3b) and a distinct mid-tropospheric updraft (500-hPa level) (Figure 3c). The mean upper-tropospheric circulation (250-hPa level) over East Asia was characterized by a northwesterly subtropical jet stream and a westerly subtropical jet stream, both of which were confluent over Japan to form a distinct jet core (Figure 3d).

[22] Figure 4a shows the 24-h precipitation observed at World Meteorological Organization (WMO) surface stations as the average during the A-FORCE period (17 March through 30 April 2009). The data were archived by the National Climatic Data Center (NCDC). Figure 4b shows mean precipitation during the same period, based on Global Precipitation Climatology Project (GPCP) data [Huffman et al., 2001]. The daily precipitation is available for the GPCP data on a regular grid with a resolution of 1° in both latitude and longitude, based on satellite and rain gauge measurements. Figures 4a and 4b show similar spatial distributions of the mean precipitation, indicating the consistency between these data sets. As shown in Figure 4b, the mean precipitation was pronounced over the Indochina Peninsula, southern/central China, the East China Sea,
As shown in Figure 3b, an abundant moisture supply into these regions by the low-level southerlies contributed to the locally enhanced precipitation, while precipitation was much less over northern China (Figures 4a and 4b). In fact, the amounts of precipitation estimated from the GPCP data over the midlatitude region (35°–50°N, 80°–140°E) and subtropical region (20°–35°N, 80°–140°E) of East Asia averaged over the A-FORCE period were 0.82 and 2.6 mm day$^{-1}$, respectively.

In the following, the precipitation predicted by the WRF model simulation during the A-FORCE period is compared with the corresponding GPCP data, because our estimation of the APT value depends on the reproducibility of the WRF precipitation. Figure 4c shows the mean WRF precipitation during the A-FORCE period (17 March through 30 April 2009). The WRF precipitation (1.3 mm day$^{-1}$) averaged within the midlatitude region (35°–50°N, 80°–140°E) over the entire A-FORCE period overestimated the corresponding average of GPCP precipitation (0.82 mm day$^{-1}$) by 57%. For the subtropical region (20°–35°N, 80°–140°E), the average WRF precipitation (3.0 mm day$^{-1}$) overestimated the GPCP precipitation (2.6 mm day$^{-1}$) by 13%. The correlations between WRF and GPCP precipitation (i.e., spatially and temporally averaged for each 5° × 5° grid box and 1 day, respectively) within the midlatitude and the subtropical regions showed r$^2$ values of 0.43 and 0.39, respectively. Given some uncertainties in representing precipitation in the GPCP data, we conclude that the WRF simulation overall reproduces the spatial distribution of the observed precipitation reasonably during the A-FORCE period. In fact, Verma et al. [2011] also pointed out

Figure 3. Mean meteorological fields over East Asia during the A-FORCE period (17 March to 30 April 2009) based on NCEP FNL data. (a) Mean sea level pressure (hPa, contours) and mean horizontal winds at the 850-hPa level (m s$^{-1}$, vectors with scaling near the lower-right corner). The heavy solid line denotes the approximate location of the frontal zone over central/southern China. Regions without data correspond to those of high-altitude mountains. (b) Mean equivalent potential temperature $\theta_e$ (K, filled contours) and total meridional moisture transport ($qv$ values in vector form with scaling near the lower-right corner) at the 850-hPa level (m s$^{-1}$ g K g$^{-1}$). $qv$ vectors with magnitudes greater than 10 m s$^{-1}$ g K g$^{-1}$ are plotted. (c) Mean vertical wind velocity ($w$-velocity) at the 500-hPa level (Pa s$^{-1}$, filled contours). (d) Mean westerly wind speed (m s$^{-1}$, filled contours) and mean horizontal winds at the 250-hPa level (m s$^{-1}$, vectors with scaling near the lower-right corner).

the western Pacific (south and east of Japan). As shown in Figure 3b, an abundant moisture supply into these regions by the low-level southerlies contributed to the locally enhanced precipitation, while precipitation was much less over northern China (Figures 4a and 4b). In fact, the amounts of precipitation estimated from the GPCP data over the midlatitude region (35°–50°N, 80°–140°E) and subtropical region (20°–35°N, 80°–140°E) of East Asia averaged over the A-FORCE period were 0.82 and 2.6 mm day$^{-1}$, respectively.

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4.2. Temporal Variations of Cyclones and Convection

As a measure of the activity of migratory cyclones and anticyclones, the instantaneous deviation of the meridional wind velocity ($v'$) from its 5-day running mean at the 850-hPa level was evaluated at each grid point. Time-longitude variations of the 500-hPa omega-velocity superimposed on those of the 850-hPa $v'$ over East Asia, both averaged over 35°–45°N and 25°–35°N, are shown in Hovmöller diagrams in Figures 5a and 5b, respectively, between 100°E and 150°E. These diagrams indicate the close association between mid-tropospheric updraft (black dashed lines) and migratory cyclone (blue and green shading). The mid-tropospheric updraft was enhanced occasionally at mean intervals of 5.4 days (7–8 times) and 5.3 days (8–9 times) for 35°–45°N and 25°–35°N, respectively, on the passage of synoptic-scale cyclonic disturbances. These results indicate that upward transport of CO and BC over both northern and southern China was associated with frontal cyclones (WCBs).

Figure 6a shows a time-latitude cross section of 500-hPa omega velocity averaged between 105°E and 120°E. Solid lines in Figure 6a indicate latitudes at which the longitudinally average $\theta_e$ difference between the 500- and 925-hPa levels ($\Delta \theta_e$) became zero. The lines separate dry, cool midlatitude air with stable stratification ($\Delta \theta_e > 0$) from warmer and/or moister subtropical air whose stratification is less stable ($\Delta \theta_e < 0$) and thus favorable for moist convection. Consistent with Figure 5b, the mid-tropospheric updraft over southern/central China (between 20°N and 35°N) was enhanced occasionally for a few days during the A-FORCE period, namely around 21, 27 March and 4, 11, 15, 18, 22 April. Each of these sporadic updraft events occurred immediately after the poleward intrusion (up to 25°–35°N) of convectively unstable subtropical air masses with negative $\Delta \theta_e$ values. As pointed out by Oshima et al. [2004], the passage of cyclonic disturbance can act as a trigger for moist convection over central China in spring. Figure 6b shows a time-latitude cross section of the average of the lowest 5% of the equivalent blackbody temperatures ($T_{BB}$) between 105°E and 120°E, obtained by Multifunctional Transport Satellite (MTSAT) infrared (IR) images, as an indicator of cloud top altitudes of the deepest convective clouds in this longitudinal sector. Between 20°N and 40°N, the timing and latitudinal position of occurrence of the upper-level clouds with low $T_{BB}$ (e.g., lower than 230 K) were in general agreement with those of the sporadic updraft events shown in Figure 6a, indicating that the strong updrafts associated with the upper-level clouds were due to deep cumulus convection. These results indicate that moist convection also played an important role in upward transport of CO and BC over southern/central China during the A-FORCE period, in

Figure 4. Mean precipitation (mm day$^{-1}$) obtained from (a) the NCDC at WMO surface stations, (b) GPCP data, and (c) a WRF model simulation during the A-FORCE period (17 March to 30 April 2009). The locations of the stations that reported data on fewer than 10 days during the period are denoted with gray circles (Figure 4a). The evaluations of WRF precipitation were conducted within the two rectangular domains (20°–35°N, 80°–140°E and 35°–50°N, 80°–140°E) denoted by white lines (Figures 4b and 4c). Note that the GPCP data were available on a regular grid with a horizontal resolution of 1.0° in both latitude and longitude, and the WRF simulation was conducted with a horizontal grid resolution of 81 × 81 km$^2$. 

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Figure 5. Hovmöller diagrams based on NCEP FNL data showing time-longitude variations of the 500-hPa omega-velocity (Pa s\(^{-1}\), black lines) superimposed on those of the 850-hPa \(v'\) (m s\(^{-1}\), filled contours) over East Asia, both averaged latitudinally (a) between 35\(^{\circ}\)N and 45\(^{\circ}\)N and (b) between 25\(^{\circ}\)N and 35\(^{\circ}\)N during the A-FORCE period, where \(v'\) is the instantaneous deviation of the meridional wind velocity from its 5-day running mean at each grid point. Black solid and dashed lines denote the \(\omega\)-velocity of 0.15 Pa s\(^{-1}\) (downward motion) and −0.15 Pa s\(^{-1}\) (upward motion), respectively.
Figure 6. (a) Time-latitude cross section of the 500-hPa $\omega$-velocity averaged longitudinally between 105°E and 120°E (Pa s$^{-1}$, filled contours) during the A-FORCE period based on NCEP FNL data. The black thick lines denote locations where the $\theta_e$ difference between the 500-hPa and 925-hPa levels was zero ($\Delta \theta_e = 0$). Convectively unstable air masses (negative $\Delta \theta_e$) penetrated up to 25°–35°N, in association with the intrusions of the warm, moist low-level southerlies. (b) Time-latitude cross section of the average of the lowest 5% of $T_{BB}$ (equivalent blackbody temperature (K) derived from IR images obtained by the MTSAT) values between 105°E and 120°E during the A-FORCE period, as an indicator of the cloud top altitudes of the deepest convective clouds (color-coded).
Figure 7. Vertical profiles of 10-s mean (a, c, and e) CO mixing ratio and (b, d, and f) BC mass concentration measured at 34°–38°N (Figures 7a and 7b), 30°–34°N (Figures 7c and 7d), and 26°–30°N (Figures 7e and 7f) latitude ranges during the A-FORCE aircraft campaign (18 March to 25 April 2009) (black lines). The red circles denote the median values of 1-min mean CO (Figures 7a, 7c, and 7e) and BC (Figures 7b, 7d, and 7f) for each 1-km altitude range, and the red horizontal lines denote the central 67% ranges.
addition to large-scale updrafts associated with frontal cyclones (WCBS).

5. Spatial Distribution of BC During A-FORCE

[26] Figure 7 shows vertical profiles of CO mixing ratio (left) and BC mass concentration (right) observed in three latitude ranges (34°–38°N, 30°–34°N, and 26°–30°N) during A-FORCE. Median values and central 67% ranges of CO and BC concentrations observed at each 1-km altitude step for the three latitude ranges are also shown in Figure 7 and summarized in Table 3. Figure 8 shows horizontal distributions of the median values of CO and BC concentrations observed in each 1° × 1° grid box for three altitude ranges (0–2 km, 2–4 km, and 4–6 km).

[27] In general, large enhancements of CO and BC were frequently observed at the 0–2 km level during flights over the Yellow Sea and the East China Sea (Figures 7a–7f and Figures 8a and 8b). In particular, pronounced large enhancements of CO (i.e., 300–900 ppbv) and BC (500–3500 ng m⁻³) were observed over the Yellow Sea (black lines in Figures 7a and 7b). The back trajectories of these air parcels indicate that these enhancements were due to horizontal transport of CO and BC within the PBL originating from anthropogenic emissions over the northern and central parts of China (Figure 1).

[28] In the lower FT (2–4 km), high concentrations of CO (greater than 200 ppbv) and BC (greater than 400 ng m⁻³) were observed during several flights both over the Yellow Sea and the East China Sea (black lines in Figures 7a–7d). These high concentrations were also marked by relatively high median values of CO (greater than 180 ppbv) and BC (greater than 250 ng m⁻³) along the flight tracks (Figures 8c and 8d). In the middle FT (4–6 km), in contrast, CO enhancements (greater than 200 ppbv) with relatively low BC concentrations (smaller than 400 ng m⁻³) were frequently observed over the East China Sea (black lines in Figures 7c and 7d), although simultaneous enhancements of CO (greater than 200 ppbv) and BC (greater than 400 ng m⁻³) were occasionally observed over the Yellow Sea (black lines in Figures 7a and 7b). The difference in the BC concentrations in the FT over the Yellow Sea and the East China Sea was likely due to the difference in the degree of wet removal of BC during upward transport, as discussed in sections 6 and 7.

6. Two Case Studies of Upward Transport of BC

[29] As presented in section 4, uplifting mechanisms (i.e., frontal cyclones and convective updrafts) and precipitation amounts during the A-FORCE period exhibited certain regional characteristics between northern and southern/central China. In this section, those regional characteristics are examined in detail, in highlighting two events of upward transport of BC observed in the FT over the Yellow Sea and the East China Sea.

6.1. Cyclone-Induced Transport Over Northern China

[30] Figure 9a shows vertical profiles of BC mass concentration and CO mixing ratio observed over the Yellow Sea around 37°N, 126°E during flight 8 on 30 March 2009 (Figure 1). Large enhancements of both BC (greater than 800 ng m⁻³) and CO (greater than 400 ppbv) were observed in air parcels sampled between 3 and 6 km in altitude. Figure 10 shows the 5-day back trajectories of these air parcels. The trajectories indicate that these air parcels were situated in the lower troposphere (800-hPa level or below) over northern China about 12 h prior to the measurement and had likely been influenced by anthropogenic emissions over northern China (Figure 1). They then underwent rapid uplifting from the 800-hPa up to the observed mid-tropospheric levels within the following 12 h over the Shandong Peninsula around 37°N, 120°E in China.

[31] Meteorological fields at the time of the rapid uplift of these air parcels (1800 UTC on 29 March 2009) are shown in Figures 11a–11c. In the lower troposphere (850-hPa level), relatively warm and moist southerly converged into a frontal zone extending zonally between 110°E and 122°E around 37°N (as denoted by a heavy solid line in Figures 11a–11c), where the meridional gradient of θ_e was pronounced (Figure 11a). In good agreement with the enhanced low-level convergence (Figure 11a), mid-tropospheric upward motion was also enhanced along the front (Figure 11b). The updraft was strongest in the vicinity of a low-level cyclone around 37°N, 120°E, marked by a local maximum of relative vorticity.
The rapid uplifting of the air parcels to the FT as revealed in the trajectory analysis was therefore likely due to the cyclone activity (i.e., frontal lifting). An increase in potential temperature (>0.5 K/hour) along the back trajectories (not shown) suggests the occurrence of diabatic heating associated with precipitation during upward transport of the air parcels. Figure 11d shows the distribution of 24-h precipitation observed at WMO surface stations as an average for 29 and 30 March 2009. Modest amounts of precipitation were observed over the
Shandong Peninsula around 37°N, 120°E (1.9–9.4 mm, 5.0 mm averaged for the 36°–38°N and 118°–122°E domain), where the air parcels were uplifted in association with the cyclone. Changes in the WRF precipitation water content along the trajectories suggest the occurrence of precipitation during their upward transport (Figure 10b). These results suggest that some of the BC particles were removed from the air parcels by precipitation during the upward transport from the PBL to the FT.

The TE\textsubscript{BC} and APT values were estimated for the air parcels, sampled between the 3- and 6-km levels shown in Figure 9a. The mean values of TE\textsubscript{BC} and APT were 0.53 and 3.8 mm, respectively. This result is consistent with the occurrence of moderate precipitation during the upward transport during flight 8 (Figure 11d).

6.2. Convective Transport Over Central China

Figure 9b shows vertical profiles of BC and CO observed over the East China Sea around 33°N, 128°E during flight 19 on 23 April 2009 (Figure 1). In contrast to the flight 8 case, only enhancements of CO (greater than 200 ppbv) and no substantial enhancements of BC (smaller than 200 ng m\textsuperscript{-3}) were observed in air parcels sampled between 5 and 6 km in altitude. The 5-day back trajectories of these air parcels (Figure 12) indicate that these air parcels were situated in the lower troposphere over the central China region (along the Yangtze River (around 30°N)) about 24 h prior to the measurement and had likely been influenced by anthropogenic emissions over the region. They then underwent rapid uplifting to the mid-troposphere within the following 9 h over inland central China around 30°N, 110°E. Finally, these air parcels were transported horizontally toward the observation area over the East China Sea by the westerly subtropical jet.

Meteorological fields at the time of the rapid uplift of the air parcels (1200 UTC on 22 April 2009) are shown in Figures 13a and 13b. Comparing with the flight 8 case, much warmer and moister southerlies in the lower troposphere (850-hPa level) converged into a frontal zone extending between 30°N, 110°E and 25°N, 120°E (as denoted by a heavy solid line in Figures 13a and 13b), marked by a tight meridional \theta\textsubscript{e} gradient (Figure 13a). In association with the pronounced lower-tropospheric moisture transport into central China, mid-tropospheric upward motion was also enhanced in the vicinity of the frontal zone (Figure 13b). An IR image by the MTSAT shown in Figure 13c indicates the formation of deep convective clouds with T\textsubscript{BB} values below 235 K (or, equivalently, cloud top altitudes higher than 9.5 km) around the convergence zone. Despite certain limitations in resolving the effects of subgrid-scale convection on motions of the air parcels in our grid-scale trajectory calculation, the consistency among the rapid uplifting of the air parcels by the trajectories, the deep convective clouds identified by the IR image, and the poleward intrusion (up to 30°N) of the convectively unstable air mass shown by the NCEP FNL data (Figure 6a) during the uplifting period provides evidence that the rapid uplifting of the air parcels to the FT during flight 19 was quite likely due to deep convection over inland central China.

Similar to the flight 8 case, an increase in potential temperature (>1.0 K/hour) along the trajectories (not shown) suggests the occurrence of latent heat release associated with precipitation in the uplifted air parcels. The 24-h precipitation observed at the WMO surface stations averaged for 22 and 23 April 2009 are shown in Figure 13d. Large amounts of precipitation were observed over inland central China (1.5–48 mm, 21 mm averaged for the 28°–32°N and 108°–112°E domain), where the air parcels were uplifted. The amounts of precipitation were greater in this case than those in the flight 8 case, in the presence of an abundant moisture supply by the lower-tropospheric southerlies (Figures 11b and 13b). Changes in the WRF precipitation water content along the
trajectories also suggest the occurrence of precipitation during their upward transport (Figure 12b). It is likely that the removal of the large portion of BC particles from the air parcels was due to heavy precipitation during their upward transport.

[37] The mean $TE_{BC}$ and $APT$ values estimated for the air parcels were 0.12 and 11 mm, respectively. This result is consistent with the occurrence of the large amount of precipitation during the upward transport during flight 19 (Figure 13d). The distinct difference in the precipitation amount between the flight 8 and flight 19 cases is the likely cause of the large difference in BC concentrations observed in the FT.

7. Wet Removal of BC During the A-FORCE Period

7.1. Dependence of Wet Removal of BC on Precipitation

[38] We examined the dependence of wet removal of BC on precipitation for air parcels sampled in the FT using the
entire A-FORCE data set (based on 1-min average data that match the back trajectories). Figure 14 shows the relationship between the $TE_{BC}$ and the APT values for the “uplifted air parcels” sampled above 2 km in altitude during the entire A-FORCE period (black circles). The median values and the central 67% ranges of $TE_{BC}$ and APT within each APT range (i.e., APT values were divided into eight even intervals between 0.01 mm and 100 mm based on a constant common ratio) are also shown in Figure 14 (red circles and lines). Although the negative correlation between $TE_{BC}$ and the logarithm of APT for the air parcels was modest (i.e., black circles, $r^2 = 0.43$), the good correspondence between their median values (i.e., red circles, $r^2 = 0.88$) indicates a clear tendency that the $TE_{BC}$ value decreases with the increase in the APT value. This result indicates that $TE_{BC}$ primarily depended on APT, namely the wet removal of BC from air parcels primarily depended on the precipitation amount that the air parcels experienced during vertical transport from the PBL to the FT. The decreasing trend is also applicable for the case studies based on the flights 8 and 19 (open black triangles and upside-down triangles, respectively, in Figure 14), namely the mean $TE_{BC}$ and APT values for the flight 19 case were found smaller and greater than those for the flight 8 case, respectively. It should be noted that APT is a relative value that represents the degree of the effect of precipitation on the air parcels, and the absolute value can change depending on the calculation method (e.g., the integration time along trajectories and precipitation data used for the

Figure 11. Figures 11a–11c show meteorological fields based on NCEP FNL data at the time when air parcels in which large enhancements of both BC mass concentration and CO mixing ratio were observed between 3 and 6 km in altitude during flight 8 were uplifted over the Shandong Peninsula in China (18 UTC on 29 March 2009). (a) $\theta_e$ (K, filled contours) and horizontal wind at the 850-hPa level (m s$^{-1}$, vectors with scaling near the lower-right corner). The heavy solid line denotes the approximate location of a surface front. (b) Meridional moisture transport ($qv$ values in vector form with scaling near the lower right corner) at the 850-hPa level (m s$^{-1}$ g K$^{-1}$) and vertical wind velocity ($\omega$-velocity) at the 500-hPa level (Pa s$^{-1}$, filled contours). $qv$ vectors with magnitudes greater than 50 m s$^{-1}$ g K$^{-1}$ are plotted. (c) Relative vorticity at the 850-hPa level (s$^{-1}$, filled contours). (d) Twenty-four-hour average precipitation observed at WMO surface stations (mm day$^{-1}$) for 29 and 30 March 2009 (colored circles). The locations of the stations that did not report precipitation for the period are denoted with gray circles.
integration). The variability in $TE_{BC}$ shown in Figure 14 is primarily explained in terms of $APT$ ($r^2 = 0.43$), however the remaining variability will be contributed to by other factors, including uncertainties in estimates of $APT$ and/or $TE_{BC}$ (see section 7.4). For example, the uncertainties in estimates of $APT$ arose from errors in the WRF-simulated precipitation and/or in the trajectory calculations. Another factor that can weaken the $APT$-$TE_{BC}$ correlation may be modifications of the $TE_{BC}$ values of air parcels through their mixing with different background air that could occur during their long-distance transport [e.g., Pisso et al., 2009].

[39] In order to understand the spread in the $TE_{BC}$ values among the individual air parcels in Figure 14, we examined relationships among $TE_{BC}$, $APT$, and the origins of the “uplifted air parcels” (see section 3.4). The origins (or the “uplifted locations”) of the air parcels sampled above 2 km in altitude during the entire A-FORCE period are shown in Figure 15. The “uplifted air parcels” originating from northern China (north of 33°N) and southern China (south of 33°N) are hereafter referred to as “NC air parcels” and “SC air parcels,” respectively. In Figures 15a and 15b, the $TE_{BC}$ and $APT$ values, respectively, estimated for the air parcels sampled on board the aircraft are assigned at their “uplifted locations” with different colors. As shown in Figure 15a, $TE_{BC}$ values for the NC air parcels were systematically greater than those for the SC air parcels. In good agreement with the spatial pattern of the $TE_{BC}$ values, the $APT$ values for the NC air parcels were
smaller than those for the SC air parcels (Figure 15b). The latitudinal difference in APT is consistent with that in precipitation over East Asia in spring (Figure 4). These results indicate that the regional-scale distribution of precipitation was important in controlling the spatial distribution of TE\textsubscript{BC} over East Asia.

Figures 16a and 16b show the relationship between the TE\textsubscript{BC} and the APT values for the “uplifted air parcels” sampled at 2–4 km and 4–9 km in altitude, respectively, during the entire A-FORCE period (filled and open black circles). The decreasing trend in the TE\textsubscript{BC} value with the increase in the APT value is also seen for both altitudes in Figures 16a and 16b, although these correlations are not strong (gray lines in Figure 16). Figure 16 also shows that the TE\textsubscript{BC} and APT values depend on the altitude at which the air parcels were sampled by the aircraft. Namely, the TE\textsubscript{BC} and APT values for the air parcels sampled at 4–9 km in altitude (Figure 16b) are generally smaller and greater than those sampled at 2–4 km (Figure 16a), respectively. It is suggested that, as uplifted from the PBL to higher altitude in the FT, an air parcel tends to have a greater chance of being influenced by precipitation, accounting for the altitude-dependence of the TE\textsubscript{BC} and APT values shown in Figure 16.

For statistical analysis, we classified the sampled air parcels into four categories, on the basis of the sampling altitude (2–4 km and 4–9 km) and latitude of origin (southern China (20°–33°N) and northern China (33°–50°N)) of the air parcels (i.e., SC and NC air parcels). The median values and the central 67% ranges of TE\textsubscript{BC} and APT for the four categories are summarized in Table 4 and shown in Figure 16. As shown in Table 4, the median values of TE\textsubscript{BC} estimated for the NC air parcels are 0.86 (2–4 km) and 0.49 (4–9 km) and those estimated for the SC air parcels are 0.69 (2–4 km) and 0.32 (4–9 km). The decreasing trend in the TE\textsubscript{BC} values with the increase in the APT for the individual air parcels is also seen in the median values of the four categories. When the air parcels with the same origins are compared, the median values of TE\textsubscript{BC} and APT for the air parcels sampled at 4–9 km in altitude (Figure 16b) are found systematically smaller and greater than those sampled at...
Figure 14. Relationship between the $TE_{BC}$ (transport efficiencies of BC, defined by equation (1) in section 3.3) and the $APT$ (accumulated precipitation along trajectory, defined in section 3.5) values for the “uplifted air parcels” sampled above 2 km in altitude during the entire A-FORCE period (black circles). Open black triangles denote the flight 8 case in which large enhancements of both BC mass concentration and CO mixing ratio were observed in air parcels sampled between 3 and 6 km in altitude on 30 March 2009. Open black upside-down triangles denote the flight 19 case in which only enhancements of the CO mixing ratio were observed in air parcels sampled between 5 and 6 km in altitude on 23 April 2009. The black solid line is the regression line for the “uplifted air parcels” (black circles). The red circles denote the median values of $TE_{BC}$ and $APT$ within each $APT$ range (i.e., $APT$ values were divided into eight even intervals between 0.01 mm and 100 mm based on a constant common ratio), and the red vertical and horizontal lines denote the central 67% ranges. Note that the air parcels with $\Delta[CO]$ values greater than 30 ppbv are shown. See the text for details.

2–4 km (Figure 16a), respectively (Table 4). When the air parcels sampled within the same altitude levels are compared, the median values of $TE_{BC}$ and $APT$ for the SC air parcels are found systematically smaller and greater than those for the NC air parcels, respectively (Figures 16a and 16b and Table 4). These results indicate that $TE_{BC}$ primarily depended on $APT$, which in turn depended on the altitudes and origins of the sampled air parcels.

One may consider that the wet removal of BC also depended on the CCN activity of BC-containing particles. We discuss here the effect of the CCN activity of BC-containing particles on the wet removal of BC. Freshly emitted BC particles are generally bare [Weingartner et al., 1997; Sakurai et al., 2003], and they are gradually coated by internally mixing with other aerosols due to aging processes in the atmosphere [e.g., Moteki et al., 2007; Oshima et al., 2009a]. Previous studies showed that the timescale of the conversion of BC particles from hydrophobic to hydrophilic (internally mixed) is typically within 24 h [e.g., Riemer et al., 2004; Park et al., 2005]. Considering timescales of uplifting events associated with migratory synoptic-scale cyclones over China (about 5 days on average) and convective activity over southern China (at intervals of 6–7 days on average) during the A-FORCE period (see section 4.2), it is likely that most BC-containing particles were coated by water-soluble species to be CCN active within the PBL prior to uplifting. A previous modeling study also showed that 87% of BC-containing particles (by BC mass concentration) can act as CCN at a supersaturation level of 0.1% within the PBL over the Japanese anthropogenic source region in spring [Oshima et al., 2009b]. In addition to the anthropogenic emissions, some of the BC particles observed during A-FORCE might have been influenced by biomass burning. Kondo et al. [2011a] showed that BC-containing particles originating from biomass burning in North America and Asia were thickly coated by organic aerosols, with shell/BC-core ratios of about 1.4 several hours after emission. The increase in the shell/BC-core ratios continued for a few days, leading to an increase in the volume of coating materials by a factor of 2. This suggests that the BC-containing particles emitted from biomass burning were likely coated by water-soluble species to be CCN active within the timescales of the uplifting events during the A-FORCE period.

7.2. BC Concentrations in Uplifted Air Parcels

Figures 15c and 15d show the origins (or “uplifted locations”) of the air parcels sampled above 2 km in altitude during the entire A-FORCE period, colored with the values of CO mixing ratios and BC mass concentrations observed on board the aircraft, respectively. For statistical analysis, the median values and the central 67% ranges of CO mixing ratio and BC mass concentration are shown for the four categories (see section 7.1) in Table 4. For comparison, the corresponding median values and the central 67% ranges are also shown for the “free tropospheric” and “dry PBL” air parcels (see section 3.3) in Table 4.

As evident in Table 4, the median values of CO mixing ratios are systematically greater in the “uplifted air parcels” for the four categories (180–220 ppbv) as compared with “free tropospheric” air parcels (120 ppbv). We concluded from Figure 15c that these enhanced CO mixing ratios in the air parcels were likely caused by anthropogenic emissions, in recognition of the good spatial correspondence between the origins and the locations of high anthropogenic CO emissions [Zhang et al., 2009].

Likewise, the median values of BC mass concentration were systematically greater in the “uplifted air parcels” for the four categories (110–370 ng m$^{-3}$) as compared with “free tropospheric” air parcels (50 ng m$^{-3}$). Nevertheless, the median BC values differed among the four categories depending on the corresponding $APT$ values (Table 4). As shown in Figure 15d, the BC concentrations were greater for the NC air parcels than those for the SC air parcels. These differences are considered to be dependent on both the spatial distribution of BC emission (Figure 1) and that of precipitation (Figure 4). The largest (smallest) median value of BC mass concentration was obtained for the NC (SC) air parcels sampled at 2–4 km (4–9 km) in altitude (Table 4).
These results explain the difference in the BC concentration observed in the FT over the Yellow Sea and the East China Sea during A-FORCE, described in section 5. These results also suggest that BC particles uplifted to the FT over northern China tended to have a greater chance of efficient long-range transport and therefore may have exerted a larger impact on the radiation budget on a regional scale during the A-FORCE period.

7.3. Air Influenced by Cloud and Precipitation

Formation of clouds and precipitation causes modification and removal of aerosols in air parcels. In order to examine the influence of cloud and precipitation on the air parcels sampled in the FT (2–9 km) during A-FORCE, we classified the “uplifted air parcels” into three categories as follows: (1) air parcels influenced by heavy precipitation (APT was greater than 1 mm), (2) those influenced by light precipitation (APT was smaller than 1 mm), and (3) those influenced by neither cloud nor precipitation (APT was equal to 0 mm). The median values and the central 67% ranges of the altitude of sampling points, $TE_{BC}$, APT, and concentrations of CO and BC for the three categories (i.e., “heavy-rain,” “light-rain,” and “no-cloud/rain” air parcels) are summarized in Table 5. The median values of $TE_{BC}$ and sampling altitude in the “heavy-rain” air parcels are smaller and greater, respectively, than those in the “light-rain” air parcels. The median value of BC concentrations for the “heavy-rain” air parcels is half that of the “light-rain” air parcels, while CO concentrations are comparable between the two types of air parcels. This result indicates that uplifting associated with heavy precipitation carried air parcels to higher altitudes while removing a greater fraction of BC particles, which is consistent with the results shown in Figure 16.

The median values of BC mass concentration and sampling altitude for the “no-cloud/rain” air parcels were greatest and smallest, respectively, among the three types of air parcels, while the median CO concentrations was comparable with the other two types of air parcels. In fact, the median $TE_{BC}$ value was 1.0 for the “no-cloud/rain” air parcels. It should be pointed out that only 7% of the whole “uplifted air parcels” were free from any influence by cloud and precipitation during the A-FORCE period, suggesting...
observed in the “dry PBL” air parcels. Trajectories of the “dry PBL” air parcels indicate that most of the “dry PBL” air parcels were transported over a large area of anthropogenic emissions over northern China (not shown), suggesting that the $R_{BC-CO}$ value used in this study mainly represents the emission ratio of BC to CO over the northern and coastal regions of China.

[48] Verma et al. [2011] estimated $R_{BC-CO}$ values using BC and CO concentrations observed at Cape Hedo (26.9°N, 128.3°E) on Okinawa Island, Japan, from March 2008 to May 2009. The $R_{BC-CO}$ value for air parcels horizontally transported in the PBL from North China to Cape Hedo in March–May 2009 was $6.79 \pm 2.17$ ng m$^{-3}$ ppbv$^{-1}$. The $R_{BC-CO}$ value estimated during A-FORCE (4.84 ng m$^{-3}$ ppbv$^{-1}$) is smaller by 29% than that estimated by Verma et al. [2011], although it is within the range of the variability given by Verma et al. [2011]. The difference in the $R_{BC-CO}$ values may arise from the different methodologies for estimating $R_{BC-CO}$ in the two studies and also from the spatial variability in $R_{BC-CO}$ between Cape Hedo and the A-FORCE region. The overall uncertainty in the $TE_{BC}$ is about 35%, estimated from the square root of the sum of the squares of the 29% difference and the 20% variability (see section 3.3) in the $R_{BC-CO}$ values.

[50] It should be noted that we applied the identical value of $R_{BC-CO}$ (4.84 ng m$^{-3}$ ppbv$^{-1}$) to every air parcel sampled during the A-FORCE campaign, although the $R_{BC-CO}$ value could vary depending on emission source. According to Zhang et al. [2009], the emission ratios of BC to CO over inland southern China are greater than those over the northern and coastal regions of China. This result suggests that the $R_{BC-CO}$ value for the air parcels originating from inland southern China could be greater than that used in this study. The application of a greater $R_{BC-CO}$ value for the SC air parcels gives smaller $TE_{BC}$ values and thus enhances the latitudinal contrast of the $TE_{BC}$ values.

### 7.4.2. Uncertainties in Estimates of the APT Value

[51] In order to evaluate the uncertainties in the estimates of the APT values based on the WRF precipitation (section 3.5), we applied another method to estimate the APT values using the GPCP data, following the method of Matsui et al. [2011]. The daily precipitation is available for the GPCP data on a regular grid with a resolution of 1° in both latitude and longitude (see section 4.1). We calculated the APT values by integrating the amount of hourly precipitation in the Lagrangian sense along each of the trajectories using the time-interpolated GPCP data. As a result, the APT values estimated from the GPCP data were statistically similar to those estimated from the WRF precipitation. Namely, the median value of APT for the air parcels sampled at 4–9 km in altitude was greater than that sampled at 2–4 km, and the median value of APT for the NC air parcels was smaller than that for the SC air parcels. However, the correlation coefficient ($r^2$) between $TE_{BC}$ and the logarithm of APT of the air parcels was 0.07. The poor correlation was likely due to large uncertainties brought into our estimation of APT by the usage of the daily GPCP precipitation data, because the timescale of the wet removal of BC is considered to be shorter than a day and we cannot take into account the altitude relationship between air parcels along trajectories and the occurrence of precipitation in using the GPCP data. Nevertheless, the certain

![Figure 16](image-url)  
*Figure 16. Same as Figure 14 but for the air parcels sampled at (a) 2–4 km and (b) 4–9 km in altitude. The filled and open black circles denote the air parcels whose latitudes of “uplifted locations” or “origins” are 33°–50°N and 20°–33°N (i.e., the NC and SC air parcels), respectively. The solid gray lines are the regression lines. The red and blue circles denote the median values of $TE_{BC}$ and APT for each category, and the vertical and horizontal lines denote the central 67% ranges. See the text for details.*

that most air parcels uplifted from the PBL to the FT had been influenced by cloud and precipitation during transport.

### 7.4. Uncertainties

#### 7.4.1. Uncertainties in Estimates of the $TE_{BC}$ Value

[48] Here we discuss uncertainties in the estimates of the $TE_{BC}$ value. As presented in section 3.3, we estimated the $R_{BC-CO}$ value based on the BC and CO concentrations
similarity between the results obtained using two different methods gives confidence in the validity of our conclusions.

[52] The estimation of APT described in section 3.5 requires the WRF 3-D precipitation water content to identify the air parcels influenced by precipitation during transport, and then the WRF surface precipitation is used for the integration along each of the trajectories. For the along-trajectory integration, the hourly WRF 3-D precipitation water content can be used instead of the WRF surface precipitation. As a result, a similar relationship between $TE_{BC}$ and APT was obtained (not shown), although the correlation between $TE_{BC}$ and the logarithm of APT was poorer ($r^2 = 0.28$) compared with the result based on the WRF surface precipitation ($r^2 = 0.43$). The poorer correlation may be due to more difficulty in vertically distributing the precipitation than in providing the surface amount in meteorological models. However, possible causes were not identified by this study because of the complexity of the wet removal processes.

[53] It should be noted that the spatially and temporally averaged WRF precipitation over the midlatitude region (35°–50°N, 80°–140°E) of East Asia during the A-FORCE period overestimated the GPCP precipitation by 57% (see section 4.1). The overestimation of precipitation in the WRF simulation could lead to the overestimation of the APT values for the NC air parcels. However, this does not alter the conclusions derived from this study in any quantitative sense, because the use of the smaller precipitation amount (i.e., closer to the observations) over the midlatitude region would enhance the latitudinal contrast of the APT values for the sampled air parcels.

8. Summary and Conclusions

[54] The A-FORCE aircraft campaign was conducted over the Yellow Sea, the East China Sea, and the western Pacific Ocean during March–April 2009. During the A-FORCE campaign, 120 vertical profiles of BC particles were obtained using an SP2 instrument at 0–9 km in altitude. Both BC mass concentrations and CO mixing ratios were greatly enhanced in air parcels sampled at 3–6 km in altitude over the Yellow Sea (around 37°N, 126°E) on 30 March 2009. These air parcels were uplifted during a passage of a cyclone that accompanied modest precipitation (5.0 mm day$^{-1}$ on average) over northern China (around 37°N, 120°E), resulting in a 47% removal of BC on average. In contrast, BC concentrations did not show substantial increase despite high CO concentrations in air parcels sampled at 5–6 km in altitude over the East China Sea (around 33°N, 128°E) on 23 April 2009. These air parcels were uplifted quite likely due to cumulus convection that accompanied heavy precipitation (21 mm day$^{-1}$ on average) over inland central China (around 30°N, 110°E), resulting in large removals of BC (88% on average).

[55] Our analysis based on the entire A-FORCE data set showed that the wet removal of BC primarily depended on the precipitation amount that an air parcel had experienced during vertical transport from the PBL to the FT. Specifically, the

### Table 4. Median Values and Central 67% Ranges of Various Species for the Sampled Air Parcels From Different Origins

<table>
<thead>
<tr>
<th>Classification</th>
<th>Number of Data Points</th>
<th>Altitude (km)</th>
<th>$TE_{BC}$</th>
<th>APT (nm)</th>
<th>CO (ppbv)</th>
<th>BC (ng m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NC (2–4 km)</td>
<td>126</td>
<td>3.10 (2.05–3.68)</td>
<td>0.86 (0.54–1.14)</td>
<td>0.069 (0.00042–1.4)</td>
<td>221 (161–265)</td>
<td>371 (185–682)</td>
</tr>
<tr>
<td>SC (2–4 km)</td>
<td>51</td>
<td>2.52 (2.04–3.86)</td>
<td>0.69 (0.28–1.10)</td>
<td>0.13 (0–7.3)</td>
<td>210 (172–288)</td>
<td>265 (79.4–684)</td>
</tr>
<tr>
<td>NC (4–9 km)</td>
<td>56</td>
<td>5.47 (4.57–6.11)</td>
<td>0.49 (0.20–0.58)</td>
<td>3.4 (0.53–8.7)</td>
<td>202 (162–311)</td>
<td>154 (52.8–503)</td>
</tr>
<tr>
<td>SC (4–9 km)</td>
<td>128</td>
<td>5.86 (4.83–6.80)</td>
<td>0.32 (0.11–0.73)</td>
<td>6.6 (0.34–14)</td>
<td>179 (162–250)</td>
<td>105 (45.1–229)</td>
</tr>
<tr>
<td>Dry PBL</td>
<td>231</td>
<td>1.12 (0.623–1.68)</td>
<td>—</td>
<td>—</td>
<td>266 (174–381)</td>
<td>686 (220–1264)</td>
</tr>
</tbody>
</table>

*Median values of altitude of sampling points, $TE_{BC}$ (transport efficiency of BC), APT (accumulated precipitation along trajectory), CO mass mixing ratio, and BC mass concentration for the air parcels sampled by the aircraft during A-FORCE. Values in parentheses are the central 67% ranges. Values are for the uplifted air parcels with $\Delta$[CO] values greater than 30 ppbv (the top four lines).

bNC (2–4 km) indicates “uplifted air parcels” originating from northern China (latitudes of “uplifted location” or “origin” are north of 33°N) and sampled by the aircraft at 2–4 km in altitude. “SC” indicates “uplifted air parcels” originating from southern China (latitudes of “uplifted location” or “origin” are south of 33°N).

Number of the $TE_{BC}$ data points for “uplifted air parcels” (the top four lines) and those of the 1-min BC data for the “free tropospheric” and the “dry PBL” air parcels (the bottom two lines).

### Table 5. Median Values and Central 67% Ranges of Various Species for the Sampled Air Parcels Influenced by Precipitation

<table>
<thead>
<tr>
<th>Classification</th>
<th>Number of Data Points</th>
<th>Altitude (km)</th>
<th>$TE_{BC}$</th>
<th>APT (nm)</th>
<th>CO (ppbv)</th>
<th>BC (ng m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heavy-rain (2–9 km)</td>
<td>190</td>
<td>5.53 (3.84–6.76)</td>
<td>0.35 (0.11–0.58)</td>
<td>6.7 (1.8–14)</td>
<td>196 (164–262)</td>
<td>117 (43.4–247)</td>
</tr>
<tr>
<td>Light-rain (2–9 km)</td>
<td>145</td>
<td>3.13 (2.05–5.52)</td>
<td>0.86 (0.44–1.19)</td>
<td>0.098 (0.0087–0.57)</td>
<td>210 (161–272)</td>
<td>224 (93.8–519)</td>
</tr>
<tr>
<td>No-cloud/rain (2–9 km)</td>
<td>26</td>
<td>2.36 (2.01–3.64)</td>
<td>1.04 (0.81–1.16)</td>
<td>0</td>
<td>222 (169–330)</td>
<td>515 (229–908)</td>
</tr>
</tbody>
</table>

*Median values of altitude of sampling points, $TE_{BC}$ (transport efficiency of BC), APT (accumulated precipitation along trajectory), CO mass mixing ratio, and BC mass concentration for the “uplifted air parcels” sampled by the aircraft during A-FORCE. Values in parentheses are the central 67% ranges. Values are for air parcels with $\Delta$[CO] values greater than 30 ppbv.

bHeavy-rain (2–9 km) indicates “uplifted air parcels” whose APT values are greater than 1 mm and sampled by the aircraft at 2–9 km in altitude. “Light-rain” indicates the “uplifted air parcels” whose APT values are smaller than 1 mm. “No-cloud/rain” indicates the “uplifted air parcels” that had been influenced by neither cloud nor precipitation during transport.

*Number of the $TE_{BC}$ data points.
$TE_{BC}$ (transport efficiency of BC) value of the air parcels sampled above 2 km in altitude decreased with the increase in the APT (accumulated precipitation along trajectory) value, although the negative correlation between $TE_{BC}$ and APT was rather modest ($r^2 = 0.43$). The remaining variability in the correlation may arise from other factors, including the errors in estimates of $TE_{BC}$ and APT and the effects of mixing in the evolution of air parcels during transport. The $TE_{BC}$ values for the sampled air parcels generally decreased with the increase in altitude of the air parcels, because of the increase in APT with altitude.

The $TE_{BC}$ values for the sampled air parcels originating from northern China (north of 33°N, NC air parcels) were systematically greater than those from southern China (south of 33°N, SC air parcels). The median values of $TE_{BC}$ for the NC air parcels sampled at 2–4 km and 4–9 km in altitude were 0.86 and 0.49, respectively, whereas the corresponding values for the SC air parcels (0.69 at 2–4 km and 0.32 at 4–9 km) were considerably smaller. The regional-scale distribution of precipitation played an important role in controlling the spatial distribution of $TE_{BC}$ over East Asia.

The median values of BC mass concentration for the NC air parcels sampled at 2–4 km and 4–9 km in altitude were 371 ng m$^{-3}$ and 154 ng m$^{-3}$, respectively, which were considerably greater than the corresponding values for the SC air parcels (265 and 105 ng m$^{-3}$, respectively). These differences in BC concentrations in the FT are considered to be dependent on both the spatial distributions of BC emissions and precipitation. The greater BC concentrations in the NC air parcels suggest that BC particles emitted from northern China may have exerted greater impacts on the regional-scale radiation budget during the A-FORCE period.

There remain large uncertainties in the representations of wet removal process of BC in current 3-D models. A number of vertical profiles of BC over East Asia obtained by the A-FORCE aircraft campaign can be used for model validation of the spatial distribution of BC in this region. In particular, the observed $TE_{BC}$ values will provide a good constraint for accurate calculations of wet removal of aerosols in 3-D models.

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