



Transport of CO in a middle latitude cyclone

C. A. Klich and H. E. Fuelberg

The role of horizontal model resolution in assessing the transport of CO in a middle latitude cyclone using WRF-Chem

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Abstract

We use the Weather Research and Forecasting with Chemistry (WRF-Chem) online chemical transport model to simulate a middle latitude cyclone in East Asia at three different horizontal resolutions (45, 15, and 5 km grid spacing). The cyclone contains a typical warm conveyor belt (WCB) with an embedded squall line that passes through an area having large surface concentrations (> 400 ppbv) of carbon monoxide (CO). Model output from WRF-Chem is used to compare differences between the large-scale CO vertical transport by the WCB (the 45 km simulation) with the smaller-scale transport due to its convection (the 5 km simulation). Forward trajectories are calculated from WRF-Chem output using HYSPLIT. At 45 km grid spacing, the WCB exhibits gradual ascent, lofting surface CO to 6–7 km. Upon reaching the warm front, the WCB and associated CO ascend more rapidly and later turn eastward over the Pacific Ocean. Convective transport at 5 km resolution with explicitly resolved convection occurs much more rapidly, with surface CO lofted to altitudes greater than 10 km in 1 h or less. We also compute CO vertical mass fluxes to compare differences in transport due to the different grid spacings. Upward CO flux exceeds $110\,000\text{ t h}^{-1}$ in the domain with explicit convection when the squall line is at peak intensity, while fluxes from the two coarser resolutions are an order of magnitude smaller. Specific areas of interest within the 5 km domain are defined to compare the magnitude of convective transport to that within the entire 5 km region. Although convection encompasses only a small portion of the 5 km domain, it is responsible for $\sim 40\%$ of the upward CO transport. We also examine the vertical transport due to a short wave trough and its associated area of convection, not related to the cyclone, that lofts CO to the upper troposphere. Results indicate that fine-scale resolution with explicitly resolved convection is important when assessing the vertical transport of surface emissions in areas of deep convection.

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1 Introduction

Anthropogenic emissions are transported at atmospheric scales ranging from turbulence to global phenomena. Where and how these emissions are transported can have major impacts on air quality (e.g., Ellingsen et al., 2008) and can affect radiative forcing and other physical and dynamical processes that impact Earth's climate (e.g., Park et al., 2001). Accurately modeling pollution transport is crucial to understanding atmospheric chemistry and its effects on the environment.

Surface-based pollutants are transported most efficiently when they are lofted to the middle or upper troposphere where wind speeds generally are stronger than near the surface. Three primary mechanisms can loft pollutants from the boundary layer to the free troposphere: 1) airstreams such as the warm conveyor belt (WCB) that are associated with middle latitude cyclones (Bey et al., 2001; Cooper et al., 2002; Hannan et al., 2003; Kiley and Fuelberg, 2006), 2) orographic lifting (Henne et al., 2004; Chen et al., 2009; Ding et al., 2009), and 3) deep convection (Lawrence et al., 2003; Liu et al., 2003; Hess, 2005; Halland et al., 2009; Zhao et al., 2009). Although all three transport mechanisms are important, middle latitude cyclones and deep convection are the most variable and difficult to simulate accurately using meteorological or chemical transport models (CTMs).

Although middle latitude cyclones and their associated WCBs can occur virtually anywhere in the middle and polar latitudes (Stohl et al., 2002), distinct maxima are located off the east coasts of North America and Asia (Stohl, 2001; Eckhardt et al., 2004). The long-range synoptic scale WCB-related transport of atmospheric pollutants has been studied across both the Pacific Ocean (e.g., Jaffe et al., 1999; Yienger et al., 2000; Bey et al., 2001; Carmichael et al., 2003; Heald et al., 2003; Jacob et al., 2003; Liu et al., 2003; Cooper et al., 2004) and Atlantic Ocean (Stohl and Trickl, 1999; Cooper et al., 2001, 2002; Naja et al., 2003; Li et al., 2005; Fehsenfeld et al., 2006; Singh et al., 2009). Anthropogenic emissions across eastern Asia have been increasing rapidly (Richter et al., 2005; Ohara et al., 2007; Zhang et al., 2007, 2009), and their transport

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to the western United States (US) has been shown to increase local surface ozone concentrations (Jacob et al., 1999; Fiore et al., 2002; Jaffe et al., 2004; Oltmans et al., 2008; Zhang et al., 2008; Reidmiller et al., 2009; Lin et al., 2012). Asian emissions sometimes extend as far as Europe (Stohl et al., 2007; Fiedler et al., 2009).

A great deal of transport modeling has been done using global CTMs whose typical grid spacing is approximately 2° latitude \times 2.5° longitude (Park et al., 2001; Liu et al., 2003; Liang et al., 2004; Li et al., 2005). However, such coarse grid resolution can produce deficiencies in the results. Large spatial gradients of tracer concentrations result from complex interactions between emissions, meteorological conditions, and nonlinear atmospheric chemistry (Constantinescu et al., 2008; Srivastava et al. 2001a, b). Coarse grids artificially diffuse these large gradients, and coupled with highly nonlinear chemistry, may adversely affect the predicted pollutant concentrations. Furthermore, coarse resolution models excessively diffuse the concentrations of atmospheric pollutants, thereby underestimating their amounts in the upper troposphere (Heald et al., 2003; Lin et al., 2010).

In terms of resolving specific meteorological phenomena, coarse resolution models capture large-scale transport processes such as the WCBs (Harrold, 1973) that originate in the warm sector of a cyclone and ascend ahead of the surface cold front (Carlson, 1980). These cyclones and their WCBs are thought to be the dominant form of synoptic scale transport in the middle latitudes (Harrold, 1973), and the primary mechanism for rapidly exporting air pollution from Asia to North America (Cooper et al., 2002, 2004; Stohl et al., 2002)

Conversely, the meteorological conditions that influence the development of deep convection, such as mixing depth, local thermodynamic variability, and wind velocity are best resolved by models having much higher resolution than CTMs (Lin et al., 2009). Furthermore, once deep convection forms it cannot be adequately resolved at coarse grid spacings. Since the scale of the convection ranges from a few kilometers for small isolated cells to tens or hundreds of kilometers for organized systems such as squall lines or other mesoscale convective systems (Markowski and Richardson, 2010),

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the explicit resolution of convection requires a horizontal grid spacing less than ~ 5 km. Lackmann (2011) recently recommended a grid spacing less than 4 km to achieve explicit convective resolution. This recommendation is supported by recent studies such as Baldauf et al. (2011) and Weisman et al. (2008). We will use the term “explicit resolution” with the understanding that even a 4–5 km grid spacing is insufficient to resolve all details of convection; cloud resolving models with grid spacings of a few meters are needed for that purpose.

Since convection is not “explicitly” simulated in coarse resolution models, it must be parameterized using various cumulus parameterization schemes. There is uncertainty and subjectivity in selecting the best cumulus parameterization for a specific study. However, the important point is that no parameterization scheme will provide the detailed representation provided by higher resolution models in which parameterization is not employed. Each scheme varies in its assumptions and methodology and may produce considerably different results (Doherty et al., 2005; Taylor, 2011). Furthermore, it is not advisable to “turn off” convective parameterization and attempt to simulate convection explicitly at coarse resolution because results have revealed slow forming, large convective storms that are not observed in nature (Weisman et al., 1997; Sato et al., 2008).

Many middle latitude cyclones have deep convection embedded in their WCBs. This embedded convection enhances the overall WCB transport, and may even exceed the synoptic scale component in weaker cyclones (Purvis et al., 2003; Kiley and Fuelberg, 2006). During periods of summertime deep convection, as much as 50 % of pollutants in the boundary layer is transported to the free troposphere by thunderstorms (Thompson et al., 1994). Convection with strong updrafts can loft surface emissions to the tropopause (~ 9 –15 km altitude) as quickly as 30 min (Thompson et al., 1997). Values of carbon monoxide (CO) downwind of deep convection near tropopause altitude have been observed to be three times greater than background values (Pickering et al., 1996).

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Few studies have quantified the differences in atmospheric chemical simulations that result from using relatively high versus lower resolution models. Lin et al. (2010) compared results from two high-resolution regional atmospheric chemistry models: the global Model for Ozone and Related Tracers (MOZART v2) (Horowitz et al., 2003) run at $1.9^\circ \times 1.9^\circ$ grid spacing, with the Weather Research and Forecasting with Chemistry (WRF-Chem) model (Grell et al., 2005) and the Community Multiscale Air Quality model (CMAQ) (Byun and Schere, 2006) run at 36×36 km grid spacing. Deep convection was parameterized in both models. Results showed that the vertical lifting and outflow of Asian pollution were enhanced in the regional models throughout their study period. Nonetheless, 36 km grid spacing still is too coarse to resolve most aspects of deep convection. Therefore, it is important to investigate vertical transport using even higher resolution models. A model grid spacing of 12 km produced the best agreement between simulated and measured concentrations of O_3 and O_3 precursors in Mexico City (Tie et al., 2010), with poorer performance at both larger and smaller spacing. Chock et al. (2002) concluded that coarse resolution leads to faulty representations of ozone and nitrogen oxide chemistry. Similarly, simulated concentrations of particulates over central Europe agreed best with observations when the model grid spacing was 7 km instead of 28 km (Wolke et al., 2012). Using a high resolution (hundreds of meters) regional scale two-dimensional (2-D) model, Halland et al. (2009) found that a single squall line during its 8 h lifetime could transport as much as 96 metric tons (t) of CO upward and 67 t downward at 300 hPa.

This study focuses on the transport of CO within a middle latitude cyclone containing deep convection. We employ the WRF-Chem model run at horizontal grid spacings of 45, 15, and 5 km. Unlike Halland et al. (2009), we employ three-dimensional (3-D) simulations to capture the full scope of convective transport. We compare transport based on convective parameterization (45 and 15 km) with that using explicitly resolved convection (5 km), i.e., no convective parameterization. Results will show the importance of high resolution modeling when examining convective transport. Section 2 provides information on the data used, WRF-Chem configurations, and calculation methods.

Section 3 presents a synoptic overview, Sect. 4 the results, and Sect. 5 the summary and conclusions.

2 Data and methodology

2.1 Study domain

5 Our study domain encompassed a typical middle latitude cyclone in East Asia (Figs. 1–2). A detailed synoptic overview is provided in Sect. 3. The simulations began at 00:00 UTC 22 May 2008 and ended five days later at 00:00 UTC 27 May, spanning the entire life cycle of the cyclone. Output from the first 12 h of the runs is not used in the discussions that follow to allow sufficient time for model spinup.

10 Model simulations were made using three nested domains (Fig. 1). The outer domain (D1) covered most of eastern Asia with a horizontal grid spacing of 45 km. This relatively coarse spacing simulated large scale features of the cyclone such as its WCB. An embedded nested domain (D2) had a grid spacing of 15 km, and the innermost nested domain (D3) had a 5 km spacing that provided explicit resolution of deep convection embedded within the WCB. The wave cyclone was completely contained within
15 D2 during its life cycle (Fig. 2), and all important meteorological features were located far from the boundaries of D1 to reduce undesirable boundary effects (Warner et al., 1997). All three domains contained 50 sigma levels, most tightly packed in the boundary layer and near the tropopause.

2.2 Model specifics

We used WRF-Chem Version 3.4, an online chemical transport model coupled with the Advanced Research WRF (ARW) dynamical core (Grell et al., 2005). WRF-Chem is a fully compressible, non-hydrostatic, terrain-following model that is integrated on the Arakawa C-grid (Skamarock et al., 2008). It includes Runge-Kutta 2nd and 3rd
25 order time integration schemes and advection schemes up to the 6th order. Damping

is included near the model top of 50 hPa to prevent gravity waves from reflecting off the upper boundary.

Meteorological initial and boundary conditions were obtained from the National Centers for Environmental Protection (NCEP) final analysis on a $1^\circ \times 1^\circ$ grid at 26 pressure levels (<http://dss.ucar.edu/datasets/ds083.2/>). The boundary conditions were updated every 6 h. Four-dimensional data assimilation (nudging) was used at 6 h intervals on the outer two domains. The recommended nudging coefficient (0.0003 s^{-1} , Gilliam and Pleim, 2010) helped the simulations represent real-world conditions without being overly constrained. Other model settings and parameterizations are given in Table 1. An especially important choice was the Grell-3D cumulus parameterization (ARW WRF User's Guide, 2012) that is an updated version of the earlier Grell-Devenyi scheme (Grell and Devenyi, 2002). This parameterization, used only in D1 and D2, allows coarsely resolved subsidence to be spread to neighboring columns. Without this scheme, the downward motion between convective clouds is not well represented (Tzella and Legras, 2011). No cumulus parameterization was used in the 5 km domain, i.e., the convection was explicitly resolved to the extent possible at 5 km grid spacing.

WRF-Chem contains several options for simulating chemical interactions. We used the Regional Atmospheric Chemistry Mechanism (RACM) for gas phase chemistry that is based on the older Regional Acid Deposition Model, version 2 (RADM2, Stockwell et al., 1997). Although RACM simulates over 100 chemical species and their reactions, we focus on CO due to its long residence time of approximately 1–2 months (e.g., Novelli et al., 1998). The Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) aerosol parameterization (Chin et al., 2000) was used. GOCART is a bulk aerosol scheme that does not contain specific information about particle size or concentration, but instead calculates changes in total aerosol mass.

We performed three WRF-Chem simulations. The first utilized all three domains in Fig. 1 and included two-way nesting. Since we also wanted to examine larger-scale meteorological features without the influence of the fine-scale resolution provided by D3, we performed two other simulations, one using only D1 and D2 with two-way nest-

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ing, and the other only D1. These three simulations permitted an examination of the WCB, its convection, and the relationship between the two at both coarse and fine resolutions.

The Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT; Draxler and Hess, 1998; and <http://ready.arl.noaa.gov/HYSPLIT.php>) was used as a complement to WRF-Chem to describe the different modes of transport over time. Using the WRF-Chem simulations as input, HYSPLIT allowed us to create forward and backward trajectories at any starting location from any of the three simulations.

2.3 Emissions data

Several sets of emissions data were used to model the transport of CO. WRF-Chem was initialized with chemical data from MOZART-4 (Emmons et al., 2010). MOZART-4 also was used to update WRF-Chem's lateral boundary conditions at 6 h intervals. The archived data have a horizontal resolution of $1.9^{\circ} \times 2.5^{\circ}$ with 56 vertical levels. Initial conditions also included biomass burning emissions from the Fire INventory from NCAR (FINN, Wiedinmyer et al., 2011). Anthropogenic source emissions were from the Reanalysis of the TROpospheric dataset (RETRO, <http://retro.enes.org>). RETRO is a dataset of chemical composition at a resolution of $0.5^{\circ} \times 0.5^{\circ}$ that is mapped onto the WRF-Chem grid using a program created by the Brazilian Centro de Previsão de Tempo e Estudos Climáticos (CPTEC). Biogenic emissions were imported into WRF-Chem using the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006). MEGAN is a high-resolution ($\sim 1 \text{ km}^2$) dataset that includes leaf area index, vegetation type, and specific emission factors.

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intensified to create a well defined westerly jet stream. The low and associated jet weakened by 18:00 UTC 24 May (Fig. 2h).

The 500 hPa pressure level exhibited a second feature of interest (Fig. 2e–h). A short wave trough separate from the middle latitude cyclone began to develop over northeast China at 00:00 UTC 23 May with a wind maximum between 30°–35° N and 115°–120° E. The trough slowly moved eastward, gradually increasing in strength. It approached the Pacific Coast by 18:00 UTC 23 May (Fig. 2g), and by 07:00 UTC 24 May winds east of the trough exceeded 25 m s⁻¹ near southern Japan (Fig. 2h). The deep convection associated with the trough, as well as its flow patterns that produced CO transport are discussed in Sect. 4.

Our simulations closely represent the natural atmosphere. Observed infrared imagery (Fig. 3) from the Multifunctional Transport Satellite (MTSAT-1R) is consistent with the simulations in Fig. 2. The cyclone exhibited a classic comma shaped cloud pattern. As the system intensified and eventually became occluded (Fig. 3b–d), the band of clouds ahead of the cold front became better defined, signifying the location of the WCB described in Sect. 4. Similarly close agreements were found between other observed and simulated parameters, e.g., locations of the cyclone, its central pressure, and associated fronts (not shown).

3.2 Mesoscale features

Two major areas of deep convection occurred during the study period: a squall line ahead of the surface cold front, and a multi-cellular complex over southeast China near the southern boundary of D1 (Fig. 3). The squall line ahead of the cold front formed near 02:00 UTC 23 May and dissipated just after 12:00 UTC 23 May. Although much of this modeled convection dissipated overnight (local time = UTC + 8 h), some remnants of the squall line continued. Thunderstorm clusters re-generated ahead of the cold front along the coast late in the morning of 24 May (Fig. 3d, 4d). Although this convection did not constitute a squall line, the individual cells continually reformed over the same areas for several hours (Fig. 3d).

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Lightning occurs when there is a deep layer of mixed phase hydrometeors, generally signifying tall clouds with vigorous updrafts. We used data from the World Wide Lightning Location Network (WWLLN) to locate lightning flashes during the time of most intense convection. WWLLN has a detection efficiency of approximately 20% in our study region (Rodger et al., 2009) and a location accuracy of 5–10 km (Rodger et al., 2004). Despite this limited detection, previous studies have successfully used the data to locate deep convection (e.g., Abarca et al., 2010). WWLLN-derived lightning flashes were summed over 1 h periods between a half hour before and half hour after each satellite image and superimposed on the images (pink dots in Fig. 3). There was no lightning early in the morning of 23 May (Fig. 3a); however, high cloud tops and lightning developed within a few hours (Fig. 3b). The convection and its associated lightning continued for several hours (Fig. 3c) and dissipated late on 23 May. Less intense, disorganized convection was simulated northwest of the surface low in the far northwest part of D3, consistent with the satellite imagery (Fig. 3b–c).

Comparisons of Figs. 3–4 reveal that locations of the WRF-Chem simulated deep convection agree closely with those observed by satellite, again confirming the quality of the simulations. The squall line within D3 (Fig. 4) was located ahead of the surface cold front and exhibited simulated reflectivities exceeding 55 dBZ (Fig. 4b–c) when the maximum number of lightning flashes was observed (Fig. 3b–c). The line propagated from near the border of China and Mongolia to the border of China and Russia, a distance of approximately 700 km. Strong updrafts and downdrafts accompanying the squall line produced vertical transport of boundary layer pollution that is described in Sect. 4.

The second major area of deep convection, multi-cell storms in southeast China just south of 30° N, began near 04:00 UTC 23 May (Fig. 3a) and persisted through 14:00 UTC 24 May (not shown), producing nearly 36 h of continuous convection. These storms were associated with large-scale ascent east of the upper level short wave trough (Fig. 2f–h).

3.3 Surface CO

Eastern Asia contains some of the world's greatest anthropogenic surface emissions (Richter et al., 2005; Ohara et al., 2007; Zhang et al., 2007, 2009). The greatest CO during our period (> 400 ppbv) was located in domain D1, just south of D3 in eastern China (Fig. 5e). Some of the enhanced CO was advected into D3 by southerly winds (Fig. 2), producing concentrations that exceeded 400 ppbv over the extreme southern part of that smaller domain (Fig. 5a). The cyclonic flow transported CO northward, wrapping around the center of the low (Fig. 5a–c) after occlusion (Fig. 5d). The cold front was marked by a strong horizontal CO gradient (Fig. 5b–c), with air behind the front containing as much as 250 ppbv less CO than ahead. The horizontal gradient of surface CO concentration across the warm front was not as great as with the cold front.

The convection in southeast China within D1 (Fig. 3) occurred where CO concentrations exceeded 400 ppbv over a wide area (Fig. 5e). Since this area was located in the domain having 45 km grid spacing, the convection was parameterized, and Section 4 will show that its vertical transport of surface CO is relatively weak compared to that of the explicitly resolved convection in D3.

The squall line in D3 influenced surface concentrations of CO (Fig. 5). Since convection rapidly lofts polluted boundary layer air to the free troposphere, surface CO beneath strong updrafts often is smaller than in surrounding areas. Additionally, downdrafts typically transport relatively small values of CO from the mid troposphere to the surface. The effects of these convective scale vertical motions are apparent in Fig. 5b (40° N, 120° E) and Fig. 5c (44° N, 125° E) as isolated areas of surface CO as small as 150 ppbv, compared to surrounding environmental values exceeding 300 ppbv. When WRF-Chem was run at grid spacings greater than the 5 km in D3 (not shown), the fine-scale variations in CO concentration are not evident. Thus, high resolution models can reveal at least some localized surface variations in CO that are due to a storm's updrafts and downdrafts. As noted previously, even finer scale details of the CO field would require even smaller grid spacing.

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4 Results

4.1 Parameterized convection at 45 km resolution

Simulations performed using only outer domain D1 (45 km resolution, Fig. 1) reveal synoptic scale aspects of the WCB and its transport of CO. We used WRF-Chem output from this single-domain simulation to calculate forward trajectories using HYSPLIT. Areas of southwesterly surface winds far south of the parent low and just in advance of the cold front were considered the initiation area of the WCB, i.e., between 40° N–46° N and 117°–126° E (Fig. 6a). Trajectories were launched at each model grid point (45 km spacing) at 500 m above ground level. The trajectories were initialized at 06:00 UTC 23 May and continued for 48 h or until they left the model domain (Fig. 6a,c), the period when the middle latitude cyclone and associated WCB were best developed (Fig. 5b).

From this large number of trajectories, we isolated those representing the WCB by using criteria from Eckhardt et al. (2004). They required 48 h trajectories to travel eastward a distance exceeding 10° longitude, northward at least 5° latitude, and vertically at least 60 % of the average tropopause height at the ending latitude. The typical tropopause height in our simulation region was 11–12 km, yielding a 48 h ascent criterion of at least 6.5 km. Applying these criteria isolated approximately 100 trajectories that represented the WCB (Fig. 6a,c).

The WCB derived from the 45 km grid spacing exhibits gradual ascent from the boundary layer to the free troposphere (Fig. 6c). It remains in the boundary layer ahead of the cold front for the first 6 h (Fig. 6c) and then begins to ascend through the troposphere, rising ~3 km. When it reaches the warm front near the border of eastern China and southeast Russia (Fig. 6a), the ascent becomes more rapid, approximately 3 km during the next 6 h period. Once reaching the middle troposphere, the WCB turns more easterly (Fig. 6a). Maximum altitudes never exceed ~8 km (Fig. 6c).

The starting trajectory locations are in an area of enhanced CO (> 400 ppbv, Fig. 5). A cross section of CO (Fig. 7) along the axis of the WCB (line E–F in Fig. 6a) during the

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period of maximum ascent (03:00 UTC 24 May to 12:00 UTC 24 May) reveals values of ~300 ppbv gradually being lofted to 6–8 km.

Plan views of CO at an altitude of 5 km (Fig. 8) also reveal the evolution of the WCB related plume. As the WCB reaches the warm front between 12:00 UTC and 18:00 UTC 24 May (Fig. 2d), it begins to transport CO eastward (teal colored arrows in Fig. 8b–d). This transport is evident by an area of enhanced CO exceeding 275 ppbv in southeast Russia. Enhanced CO also wraps around the center of the low in northeast China, probably due to splitting of the WCB into clockwise and counterclockwise components. A portion of the CO plume then begins to advect eastward over the Sea of Okhotsk and eventually over the Pacific Ocean (Fig. 8c–d).

Cross sections perpendicular to the trajectories in Fig. 6 were prepared at selected times. Before 24 May (Fig. 9a), most of the CO along axis A–B in Fig. 6a is confined to the boundary layer. However, as the middle latitude cyclone and accompanying cold front move eastward, the WCB becomes much more efficient at transporting CO aloft. Just 16 h later (Fig. 9b), a large amount of CO has been transported from the boundary layer to 7–8 km. The enhanced CO remains near this altitude while transported eastward (Fig. 8). As the WCB-related CO plume moves farther east over the Pacific Ocean, it becomes diffuse both vertically (cross section C–D in Fig. 9c) and horizontally (Fig. 8c).

The CO pattern at 5 km (Fig. 8) exhibits another notable feature that is unrelated to the WCB. Synoptic scale ascent in the middle troposphere over southeast China due to the short wave trough (Fig. 2e–f) produces widespread, persistent deep convection east of the trough line beginning near 04:00 UTC 23 May. This ascent causes surface based CO to be transported into the middle troposphere south of 35° N, producing a northeast to southwest band of enhanced CO (red arrows in Fig. 8). Trajectories were launched at 500 m between 23°–30° N and 105°–120° E at 00:00 UTC 23 May, before the first lightning flashes in the area (Fig. 10), allowing parcels to encounter the convection as it forms. The criteria used to define a WCB (Eckhardt et al., 2004) also

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of storm tops, and therefore a broader range of horizontal wind directions and speeds (not shown). Since wind speeds in the upper troposphere are much faster than at lower altitudes, the pollutants quickly are advected eastward. Few trajectories from D1 reach the edge of the domain within 48 h; however, a majority of D3 trajectories do so.

5 Most of the D3 trajectories begin near an area of CO exceeding 400 ppbv (Fig. 5). The combination of strong vertical motion and large CO concentrations leads to strong vertical transport of CO. The simulated squall line exhibits a long, linear structure and passes between 120° E and 130° E (Fig. 4a–c) between 07:00 UTC and 12:00 UTC. A time sequence of east to west cross sections of CO through this region along 47° N
10 (Fig. 11) shows the evolution of the convective transport. Before the squall line develops (Fig. 11a), there is little vertical transport, with most of the CO confined to the boundary layer. An hour later as the convection forms (Fig. 11b), CO already has been lofted to ~11 km in a narrow region that corresponds to a convective “cell” as simulated at 5 km resolution. By 09:00 UTC (Fig. 11c), when the squall line is longest and near peak in-
15 tensity, a large amount of CO is being transported from the boundary layer to 12 km. As the air reaches the tropopause near 11 km altitude, the CO spreads horizontally, both eastward and westward. This spreading is consistent with the structure of squall lines observed in nature (e.g., Markowski and Richardson, 2010). The squall line continues to propagate eastward between 10:00 UTC and 11:00 UTC 23 May (Fig. 11d–e), lofting
20 additional CO into the upper troposphere. By 12:00 UTC (Fig. 11f), the squall line begins to exit the region of the cross section but continues to develop further south (not shown).

25 Cross sections of CO along the axis of the squall line at 09:00 UTC 23 May reveal the cellular nature of the simulated convection (Fig. 12). Since the squall line is bow shaped at this time, two cross sections were prepared, along I–J and J–L (axes shown in Fig. 13b). The southern portion of the squall line (axis I–J) consists of relatively young, developing “cells” (as depicted at 5 km resolution) that are typical of many squall lines occurring in nature (e.g., Markowski and Richardson, 2012). Conversely, the more mature cells are located farther north (axis J–K). The southern, younger, por-

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tion of the squall line exhibits regions of strong upward CO transport (Fig. 12a) that are surrounded by regions of diminished transport where the convection either is weaker or completely absent. The greatest vertical transport is located on the right side of cross section I–J, in the central portion of the squall line, where CO values reaching 350 ppbv have been lofted above 10 km. The northern, older section of the squall line (axis J–K in Fig. 12b) also has transported large amounts of CO upward, but the upper tropospheric CO concentrations are somewhat weaker, and the vertical plumes less defined, especially in the right half of the cross section. This is evidence of the weakening convection, and the transport of CO downwind of the squall line and away from the axis of the cross section.

The upward CO transport continues as the squall line passes through its life cycle. This quasi-continuous transport produces a large area of enhanced CO in the upper troposphere (10 km, Fig. 13) that originated near the surface. When the squall line first was forming (04:00 UTC 23 May, Fig. 13a), relatively weak values of CO are evident at 10 km. However, at 09:00 UTC 23 May (Fig. 13b), CO at 10 km has increased greatly in the areas of convection. By 12:00 UTC (Fig. 13c), even more CO has been lofted to 10 km, and the area of enhanced CO has grown quite large, with embedded values exceeding 275 ppbv. The shape of the enhanced CO feature is similar to that of the squall line that produced it during the previous several hours (Fig. 4a–c). The two-way nesting of the three-domain WRF-Chem simulation (Fig. 1) allows the convectively transported CO at 10 km to be advected downwind, out of D3 and into D1 (not shown), passing over the Sea of Okhotsk and Pacific Ocean.

The day after the squall line saw the development of multi-cell storms along the frontal boundary (Figs. 3d, 4d). Although the multi-cells produce distinct areas of convective lofting to 12 km (Fig. 12c), the CO pattern at that altitude is not as continuous or as intense as produced by the squall line the previous day (Fig. 12a, b). This occurs because the coastal multi-cells exhibit weaker updrafts and because surface CO concentrations in the multi-cell storm area are less, e.g., over North Korea, China, and Russia (Fig. 5d).

4.3 Vertical fluxes of CO

To quantify the vertical transport of CO due to our differing model resolutions, we computed vertical CO mass fluxes for several altitudes and areas. CO mass flux was computed following the technique of Halland et al. (2009) that is given by

$$M = \frac{MC \times \omega \times A \times t}{1000} \quad (1)$$

where M is the total CO mass transported in metric tons (t) per time increment, MC is the mass concentration of CO (kg m^{-3}), ω is vertical velocity, A is the area of the model grid box, and t is the time increment (hourly in our simulations). Using output from WRF-Chem, MC can be calculated using the ideal gas law:

$$MC = \frac{P_L \times M \times C}{R \times T} \quad (2)$$

where P_L is the pressure level of the flux computation, M is the molecular weight of CO, C is the model-derived CO concentration, R is the universal gas constant, and T is the model-derived temperature.

To compare fluxes from the three different WRF-Chem simulations (45, 15, 5 km), we computed areal sums within D3. That is, grid point values within the area of D3 were added, and we “zoomed in” on D1 and D2 to encompass only the D3 model domain. Although D1 and D2 contained fewer grid points than D3 itself, all three areas were the same ($\sim 4,106,200 \text{ km}^2$). We computed CO mass flux at 500 hPa at hourly intervals over a 36 h period, starting at 00:00 UTC 23 May. The 500 hPa pressure level often is near the altitude of maximum atmospheric vertical motions.

D3 produces substantially greater upward and downward CO flux than D1 or D2 even before the convection begins. At 00:00 UTC 23 May (Fig. 14) upward values are $\sim 50\,000 \text{ t h}^{-1}$. Downward flux is $-40\,000 \text{ t h}^{-1}$, creating a net flux of $\sim 10\,000 \text{ t h}^{-1}$. The enhanced mesoscale forcing that ultimately leads to squall line development begins

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near 03:00 UTC (Fig. 14), causing a rapid increase in both upward and downward CO flux. The squall line itself begins to form shortly after 07:00 UTC. Between 09:00 UTC and 10:00 UTC, when the squall line reaches peak intensity, the total area upward mass flux of CO exceeds $105\,000\ t\ h^{-1}$, with a maximum grid point value of $460\ t\ h^{-1}$. Values of total area downward mass flux are $-75,000\ t\ h^{-1}$, creating a net upward flux of $\sim 30,000\ t\ h^{-1}$, with a minimum grid point value only $-73\ t\ h^{-1}$. The updrafts at 5 km resolution are more localized and intense than the downdrafts, and CO concentrations in the middle troposphere generally are weaker than those near the surface. Thus, the upward and downward fluxes do not balance each other and some of the CO remains at 500 hPa, thereby increasing the local concentration. As the squall line begins to dissipate after 12:00 UTC 23 May (Fig. 14), the 5 km data exhibit a gradual decrease in upward, downward, and net CO vertical flux. Later, as the low pressure system and associated cold front approach the Pacific Coast, the weaker multi-cellular convection ahead of the cold front begins at 03:00 UTC 24 May. This produces a secondary increase in the three types of mass flux, with peaks near 06:00 UTC. While not as great as the squall line related fluxes, upward values exceed $85\,000\ t\ h^{-1}$ and downward flux exceeds $-65,000\ t\ h^{-1}$, producing a net vertical flux of $\sim 20\,000\ t\ h^{-1}$. Finally, as the convection slowly begins to dissipate after 06:00 UTC, the mass fluxes decrease.

There are important differences (Fig. 14) between the CO vertical fluxes from D3 and those from D1 and D2. D3 exhibits two distinct peaks at times of maximum convection; however, D1 and D2 exhibit much weaker peaks. Since convection is parameterized in these two coarser resolution simulations, their convectively induced vertical motions are much weaker than those explicitly resolved in D3.

The vertical fluxes from D1 and D2 exhibit little difference in magnitude (Fig. 14), with values separated by only a few thousand tons per hour. This small difference indicates that simply decreasing the horizontal grid spacing from 45 km to 15 km does not adequately simulate deep convection in the real atmosphere. Instead, one must explicitly resolve the convection at even smaller grid spacing (e.g., 5 km) to approximately simulate real-world convective scenarios. Due to computing requirements, explicitly re-

solving convection in global models may not be feasible, but including it in regional studies over areas of deep convection appears crucial.

Profiles of CO vertical mass flux at 09:00 UTC 23 May (Fig. 15) reveal that the contrasts seen at 500 hPa (Fig. 14) occur throughout the troposphere. The profile was computed over the same area (D3) as Fig. 14, but at the time of peak squall line intensity. Although the greatest vertical velocities are in the middle troposphere, the largest CO fluxes are near the surface, due to the much greater CO concentrations in the boundary layer. The 5 km simulation produces the greatest upward, downward, and net CO vertical transport at all levels. D1 and D2 again exhibit little difference in their fluxes. Maximum upward fluxes based on D1 and D2 resolution are located somewhat higher in the atmosphere than those from D3 resolution, and their values are approximately an order of magnitude smaller than from D3. All three resolutions produce maximum downward flux in the lower troposphere (~ 800 hPa).

We examined the vertical transport in greater detail between 04:00 UTC 23 May and 07:00 UTC 24 May (Table 2). Each hour we defined an area of interest (AOI) in the 5 km domain (D3, Fig. 4) based on simulated radar reflectivity. There was some subjectivity in defining the AOIs. They encompassed the main convective areas of the squall line each hour, including all regions of updrafts and downdrafts. The northern extent of each AOI was located where the convection began to have a more west-to-east than north-to-south orientation. The southern boundary of each AOI extended to the southern end of the continuous squall line. In the example at 09:00 UTC (Fig. 4b), the convection in the far southern portion of the box was not considered part of the squall line, but within the next 1–2 h, it merged with the convection to the north and was considered part of the line (Fig. 4b–c). Due to these criteria, the sizes of the AOIs vary considerably during the life cycle of the squall line. The area of each AOI is given in Table 2. We also defined an AOI over the multi-cell coastal convection that occurred near 07:00 UTC 24 May (Fig. 4d). We then computed CO vertical fluxes within each AOI.

We also computed upward CO transport within the entire domain of D3 (Table 2) and compared the vertical flux in this entire domain to that of the convection embedded in

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the AOIs. As the squall line begins to form at 04:00 UTC, the area of the AOI is only 2.6 % of the area of the total D3 region. However, the upward transport due to the squall line AOI is 13.4 % of the total at 700 hPa, 15.7 % at 500 hPa, and 9.8 % at 300 hPa. This difference becomes even greater as the squall line develops further. At 09:00 UTC

5 when the squall line is near peak intensity and area coverage, the area of the AOI is approximately 8.4 % of D3, but the transport within the AOI compared to D3's total is 34.1 % at 700 hPa, 36.9 % at 500 hPa, and 41.5 % at 300 hPa (Table 2). Magnitudes are similar at 12:00 UTC, when the area of the AOI is 7.7 % that of D3.

At the time of multi-cell convection at 07:00 UTC 24 May, the AOI comprises 10.2 % of the total domain (Table 2). However, at 300 hPa, the mass flux of CO is approximately 45.7 % the value of the entire domain, the greatest percentage of any altitude or time. Corresponding percentages at 500 and 700 hPa are 36.7 % and 29.1 %, respectively. Even though the convection is weaker than the squall line of the previous day, it still

10 lifts a large amount of CO into the upper troposphere.

5 Summary and conclusions

The horizontal and vertical transport of atmospheric pollutants depends greatly on synoptic scale airstreams such as warm conveyor belts within middle latitude cyclones, mesoscale deep convection that is embedded within them, as well as any deep convection that is located elsewhere. Chemical transport models (CTMs) with a horizontal grid spacing of tens to a few hundred kilometers usually faithfully simulate synoptic-scale meteorological systems. However, they cannot resolve details of convective systems because their relatively coarse grid spacing requires that the convection be parameterized at the resolution of the model. As a result, many facets of the convection are not faithfully represented, including the strong mesoscale vertical motions that can rapidly

20 transport surface based pollutants high into the atmosphere. Grid spacing less than ~5 km with explicitly resolved convection is required for this purpose.

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This study used the WRF-Chem CTM to simulate a middle latitude cyclone on 23–24 May 2008 that contained a typical WCB with an embedded squall line, as well as deep convection located away from the cyclone. Simulations were made over three domains (grid spacings of 45, 15, and 5 km) to examine CO transport at the differing resolutions. The model domains were centered over eastern Asia (Fig. 1), completely encompassing the middle latitude cyclone, its associated frontal systems, and flow patterns aloft. Convective parameterization was used in the outer two domains, while convection was explicitly resolved in the innermost domain.

The simulated middle latitude cyclone began near 12:00 UTC 22 May and lasted several days (Fig. 2a–d). A WCB was located ahead of the surface cold front with an embedded squall line on 23 May, followed by multi-cellular convection on 24 May (Fig. 4). Locations of these simulated features agreed closely with satellite imagery and WWLLN lightning data (Fig. 3), thereby supporting the validity of the simulations. The cyclone's cold front and associated deep convection passed through an area of surface CO concentrations exceeding 400 ppbv (Fig. 5), producing vertical transport into the free troposphere where horizontal wind speeds are stronger than near the surface.

Forward trajectories based on the WRF-Chem simulations were calculated using the HYSPLIT model. Comparisons were made between the coarse resolution (D1, 45 km) and finer resolution (D3, 5 km) simulations. Trajectories were launched at 06:00 UTC 23 May at 500 m above ground and run for 48 h or until they left model domain D1. These trajectories simulated the WCB, with gradual ascent occurring between the surface to 6–7 km altitude (Fig. 6a, c). Trajectories also were launched at a grid spacing of 5 km (domain D3) encompassing the simulated squall line ahead of the cold front. These trajectories vividly revealed the strong convective transport embedded within the WCB. Rapid ascent to heights > 10 km occurred within 1 h of convective initiation (Fig. 6b, d). Both resolutions of trajectories passed through areas of large surface CO concentrations, allowing us to compare differences in the vertical CO transport.

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The WCB at 45 km grid spacing lofted CO values exceeding 275 ppbv from the boundary layer to altitudes of 6–7 km (Fig. 7). The lofted CO plume passed over the cyclone's warm front and then was transported eastward toward the Pacific Ocean. Conversely, CO transported by the much stronger explicitly resolved convection was rapidly lifted to 10 km. The plume spread horizontally upon reaching the tropopause (Figs. 11–13).

The vertical transport of CO by the squall line on 23 May was much greater than by the multi-cell convection that reformed ahead of the cold front on 24 May. Cross sections of CO parallel and perpendicular to the squall line illustrated the large magnitude of the transport. Along the squall line, CO was transported to altitudes of 10–12 km (Fig. 12a–b). The multi-cell convection along the coast on 24 May (Fig. 12c) lofted CO to 10–12 km, but the areas of upward transport were more isolated, and surface CO concentrations were smaller. Although these weaker and more separated areas of ascent still transported CO upward and then spread it horizontally, the magnitude of the transport was not as great.

An area of deep convection over southeast China, unrelated to the WCB, also lofted CO to altitudes exceeding 10 km (Fig. 10). However, this transport occurred relatively slowly because the convection was located in the coarse domain where it was parameterized. Transport to the upper troposphere occurred because the convection persisted for 36 h over the same area (Fig. 3), allowing the convective parameterization to produce long lasting but weaker ascent.

CO vertical mass fluxes were computed at several altitudes over the area encompassed by D3, using WRF-Chem data at each of the three resolutions. Results showed that the simulation at 5 km grid spacing produced substantially greater upward, downward, and net CO flux when compared to fluxes based on the output from the coarser D1 and D2 simulations (Fig. 14). Values of upward flux based on the 5 km grid spacing and explicit convection were as large as $50\,000\ t\ h^{-1}$ just before convection began and exceeded $110\,000\ t\ h^{-1}$ during peak intensity of the squall line. Conversely, upward flux from the 45 km simulation (D1) and 15 km simulation (D2) never exceeded $25\,000\ t\ h^{-1}$.

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There were similar differences in magnitudes for the downward and net fluxes during the periods of convection. During a lull in the convection, magnitudes of net fluxes from the three domains were more similar. These resolution differences were evident at all altitudes examined (Fig. 15). The maximum net flux from the 5 km data occurred at a higher altitude than observed with the D1 or D2 data. However, all three resolutions yielded maximum upward flux near the surface, due to large CO values in the boundary layer.

We further examined the importance of convective transport by defining specific areas of interest (AOIs) within the 5 km domain (Fig. 4). These AOIs encompassed the simulated squall line and the multi-cell storms that formed later. When the squall line was most intense (09:00 UTC 23 May), the area of the AOI comprised only 8.4 % of the total D3 domain, but was responsible for 36.9 % of the vertical transport within the domain at 500 hPa (Table 2). Similar contrasting magnitudes were found at other altitudes and times. During the multi-cellular coastal convection on 24 May, the AOI comprised the greatest percentage of the total domain (10.2 %) and contained 45.7 % of the transport within the domain. Although the convection was multi-cellular, and less intense than the squall line of the previous day, it continuously passed over the same area, lofting large amounts of CO into the atmosphere.

These WRF-Chem simulations reveal major differences in vertical CO transport between parameterized and explicitly resolved convection. Simply decreasing the resolution from 45 km to 15 km did not produce major differences in vertical CO transport because parameterization is required at both resolutions. However, the transport was greatly enhanced by increasing the resolution to 5 km which permitted explicit resolution of the convection. Parameterized convection produces weak ascent that is hoped to represent conditions at the coarse resolution of the model. However, this convection must persist to produce significant vertical transport. Conversely, high resolution simulations with explicit convection begin to loft CO almost immediately after convective initiation. This contrasting development of convective lifting may greatly affect the patterns of long range CO transport.

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Computing requirements may prohibit the use of high resolution simulations with explicit convection over large or global domains. However, limited area, high resolution domains could be embedded within a coarse domain to encompass areas of deep convection (Wang et al., 2004), and they could move with the convection to better simulate the rapid vertical transport that occurs there. Alternatively, one could apply an adaptive mesh refinement (AMR) (e.g., Constantinescu et al., 2008) in which the grid adapts dynamically during the simulation, changing the grid resolution at certain intervals (regridding frequency), with the purpose of controlling the numerical spatial discretization error.

Although our findings are for a single case study, we hypothesize that they are applicable to many middle latitude cyclones that contain deep convection, and also to areas of convection not associated with wave cyclones. The results demonstrate the importance of explicitly resolving convection and why it should be used whenever possible. Additional studies must be performed for cyclones and convection of varying strengths and area coverage and in other areas of the globe to better understand convective transport and its influence on atmospheric chemistry.

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Table 1. WRF-Chem configurations used in the model simulations.

Option	Configuration
Cumulus Parameterization	Grell-3D, Grell and Devenyi (2002)
Shortwave Radiation	Goddard Chou et al. (2001)
Longwave Radiation	Rapid Radiative Transfer Model, Mlawer et al. (1997)
Microphysics	Lin Microphysics, Lin et al. (1983)
Planetary Boundary Layer	Yonsei University, Hong et al. (2006)
Land Surface	Noah Land Surface Model, Ek et al. (2003)
Chemical Scheme	RACM, Stockwell et al. (1997)
Aerosol Scheme	GOCART, Chin et al. (2000)
Photolysis	Madronich, Madronich (1987)

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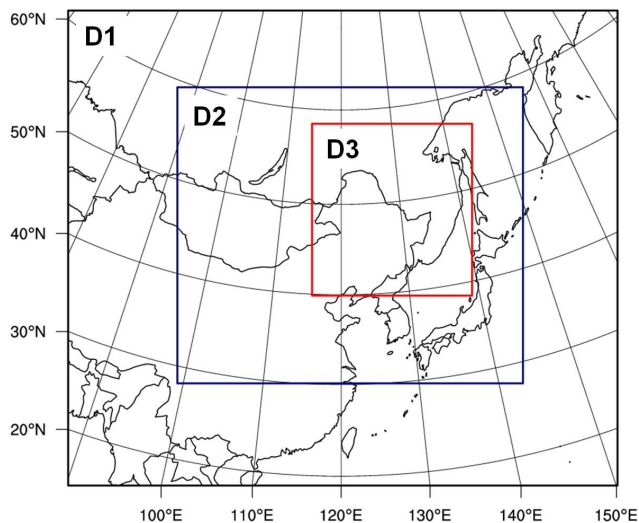


Fig. 1. The three domains used in the WRF-Chem simulations. Domain one (D1) has a horizontal grid spacing of 45 km, domain 2 (D2) 15 km, and domain 3 (D3) 5 km.

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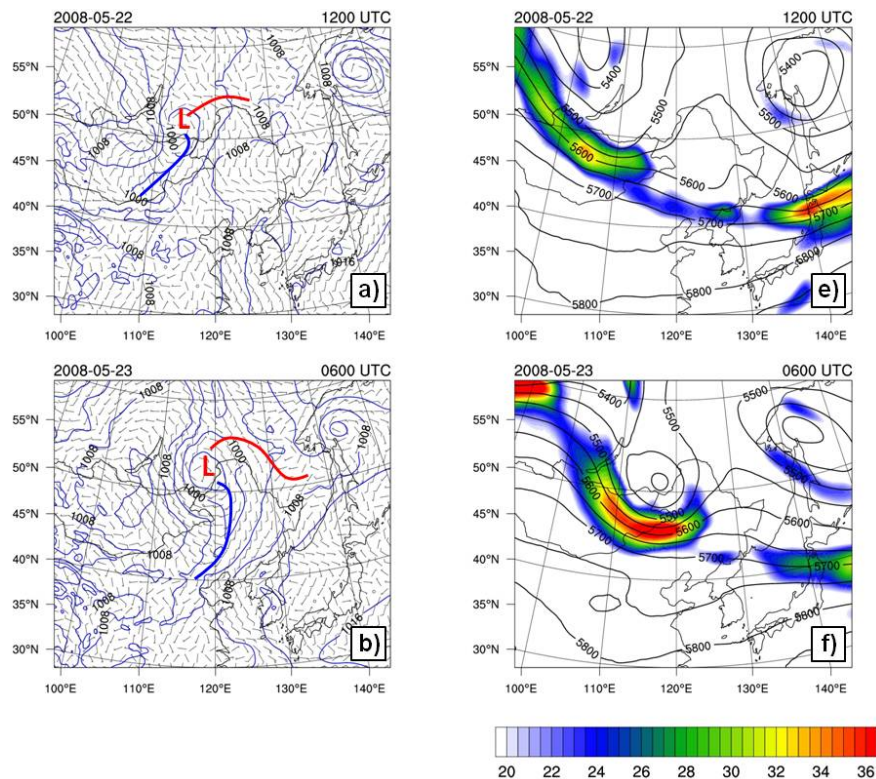


Fig. 2. Domain 2 of the WRF-Chem simulation. **(a–d)** Sea level pressure (hPa, blue contours) and 10 m winds (m s^{-1}) during the life cycle of the middle latitude cyclone. The surface location of the cyclone is shown (red “L”), as are its cold front (thick blue line), warm front (thick red line), and occluded front (thick purple line). **(e–h)** 500 hPa heights (m, black contours) and windspeed (m s^{-1} , color filled contours) at the same times as **(a–d)**.

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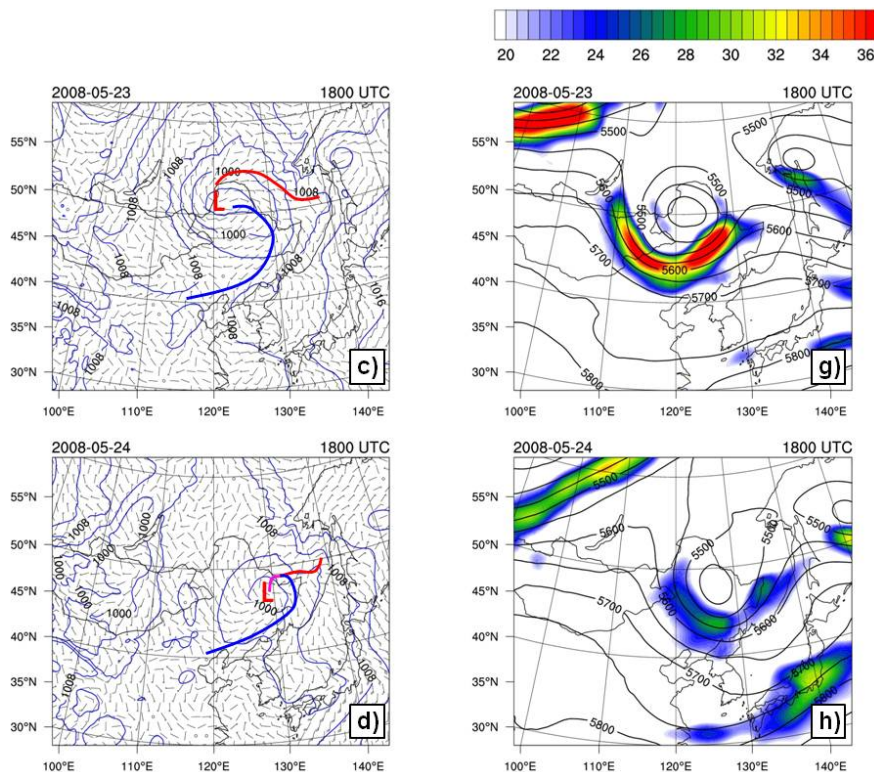


Fig. 2. Domain 2 of the WRF-Chem simulation. (a–d) Sea level pressure (hPa, blue contours) and 10 m winds (m s^{-1}) during the life cycle of the middle latitude cyclone. The surface location of the cyclone is shown (red “L”), as are its cold front (thick blue line), warm front (thick red line), and occluded front (thick purple line). (e–h) 500 hPa heights (m, black contours) and windspeed (m s^{-1} , color filled contours) at the same times as (a–d).

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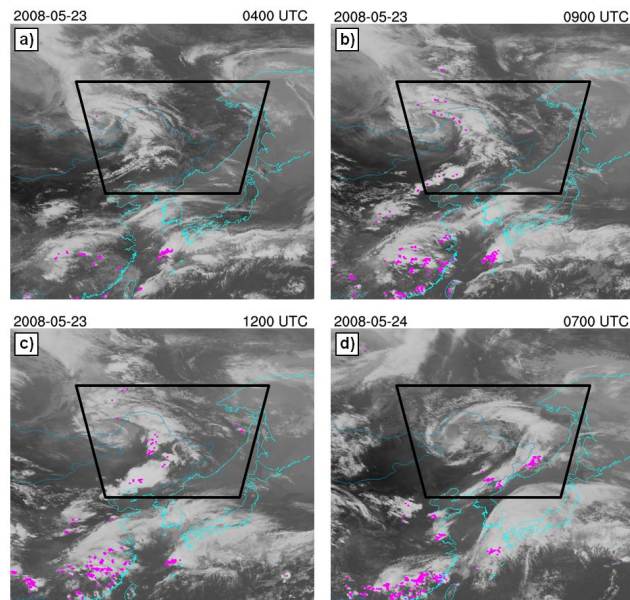


Fig. 3. Infrared imagery from MTSAT-1R and lightning data from WWLLN (pink) at four times of deep convection during the WRF-Chem simulation. The black box outlines the area of domain D3 at 5 km resolution.

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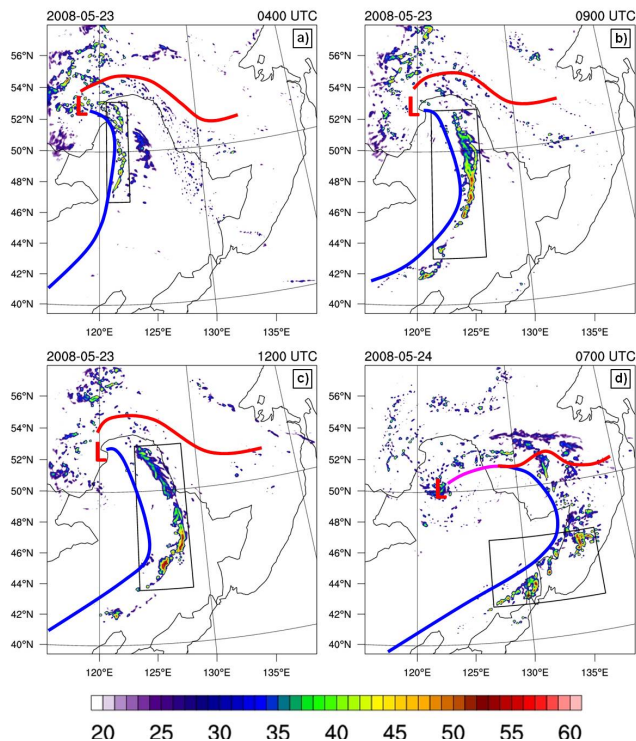


Fig. 4. WRF-Chem simulated composite reflectivity (dBZ) within D3 from formation to dissipation of the squall line (**a–c**) and coastal convection (**d**). The times shown are the same as those in Fig. 3. Locations of the surface low, cold front (blue), warm front (red) and occluded front (pink) also are shown. Black boxes around the convection are Areas of Interest (AOIs) used for flux calculations in Sect. 4.

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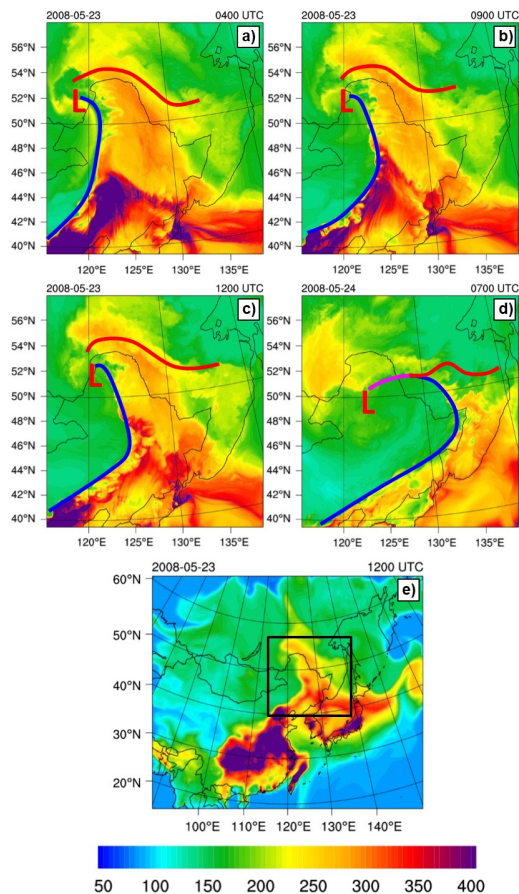


Fig. 5. Surface CO concentrations (ppbv) in D3 (a–d) and D1 (e) of our WRF-Chem simulation. The surface low, cold front (blue), warm front (red), and occluded front (purple) are shown for D3. The black box in (e) is the area of D3.

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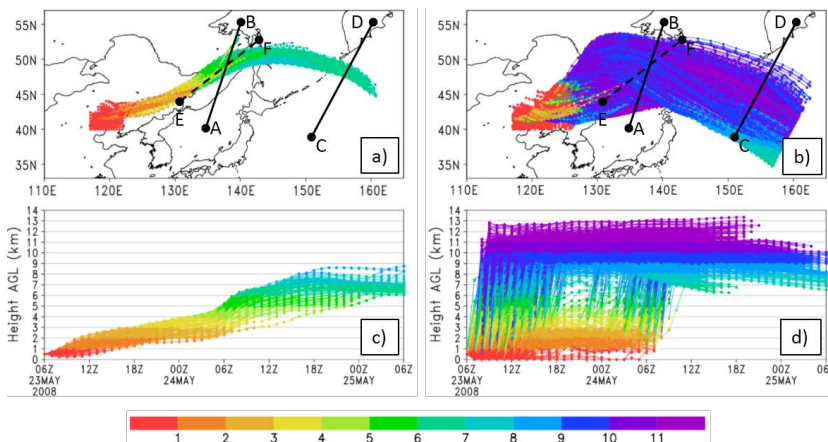


Fig. 6. HYSPLIT trajectories from D1 (**a, c**) and D3 (**b, d**) using WRF-Chem output. Axes for cross sections used in Figs. 7 and 10 also are shown (black lines). Colors indicate the heights of trajectories (km).

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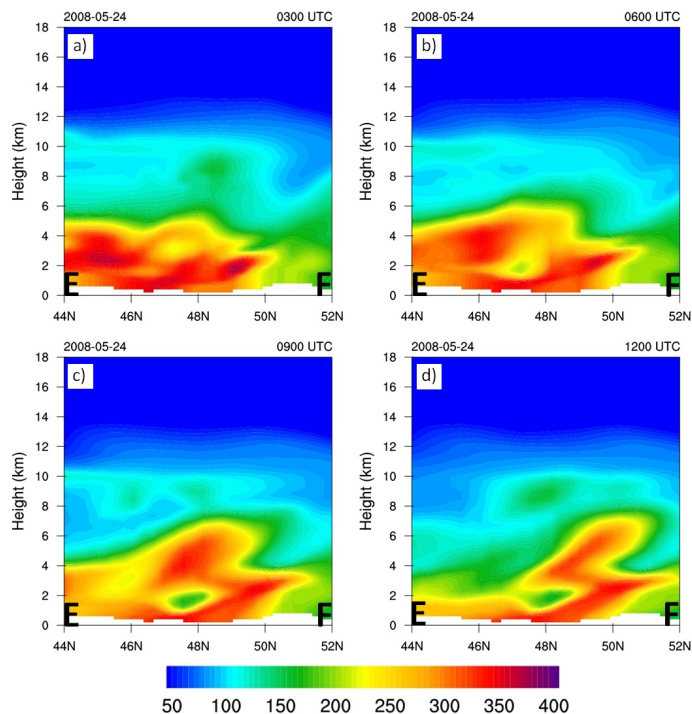


Fig. 7. Vertical cross sections of CO (ppbv) along axis E–F in Figs. 6a, b and 8a, c. Values were obtained from WRF-Chem using output from D1 only.

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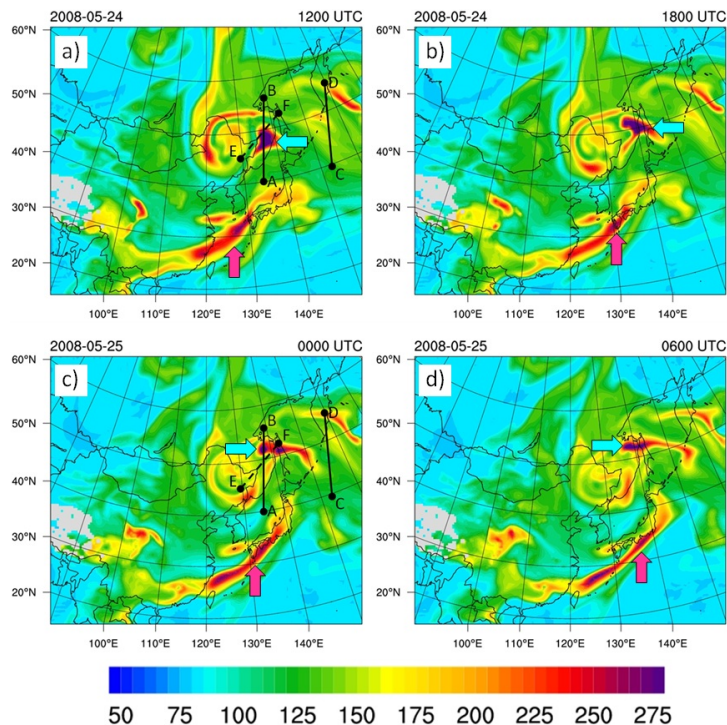


Fig. 8. CO concentration (ppbv) at 5 km altitude for the single-domain run. Axes of cross sections are shown in (a) and (c). The pink and red arrows refer to specific CO features described in the text.

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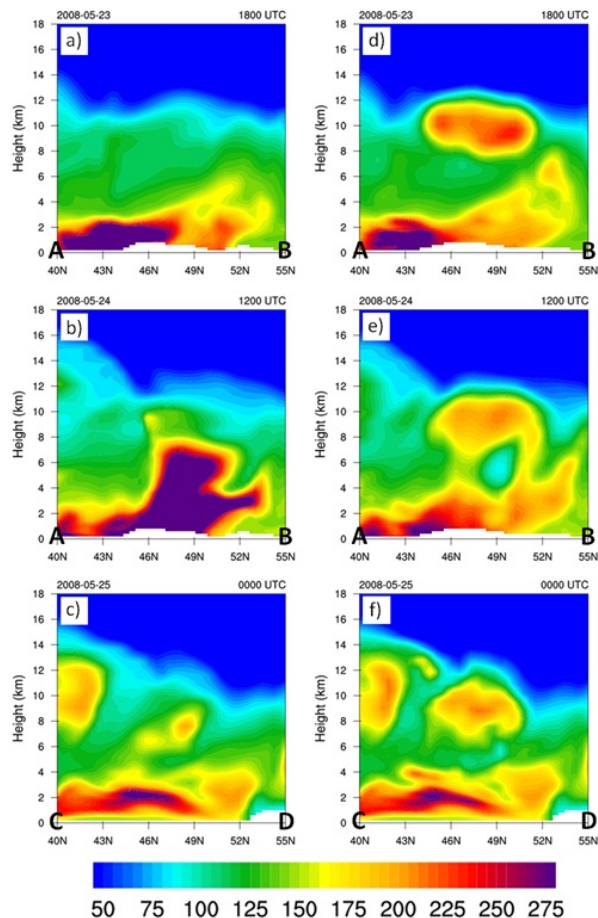


Fig. 9. Vertical cross sections of CO (ppbv) perpendicular to different locations along the trajectories in Fig. 6 for both the single-domain run (a–c) and three-domain run (d–f). Locations of cross sections A–B and C–D are shown in Figs. 6 and 8.

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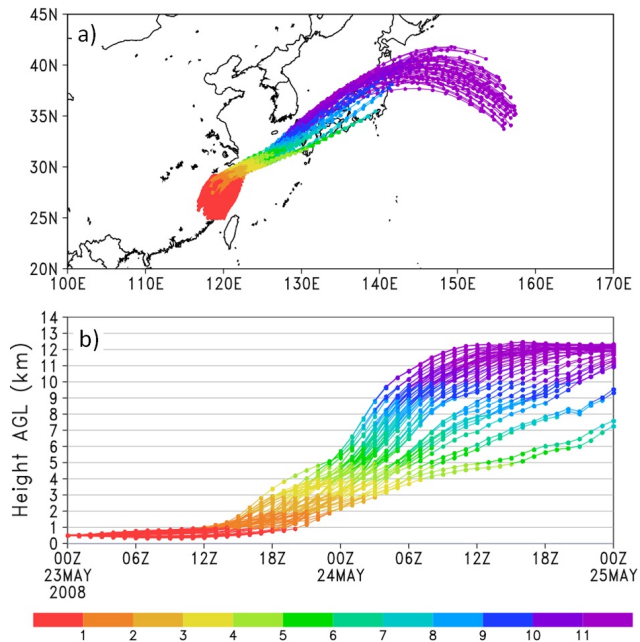


Fig. 10. HYSPLIT trajectories from D1 using WRF-Chem output as in Fig. 6a–b. Deep convection begins to form over southwest China near 04:00 UTC 23 May due to a short wave trough in the middle troposphere (Fig. 2e–h). This convection is un-related to the middle latitude cyclone or its WCB.

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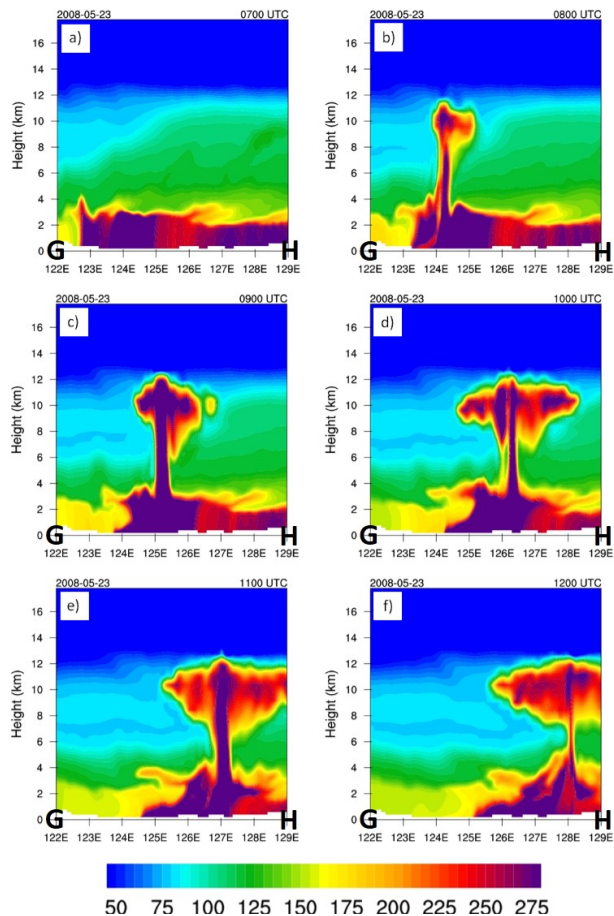


Fig. 11. Vertical cross sections of CO (ppbv) through the squall line in D3 (5 km grid spacing) at hourly intervals between 07:00–12:00 UTC 23 May. The axis of the west to east cross section is along 47° N, stretching from 122° E (G) to 129° E (H) (see Fig. 5).

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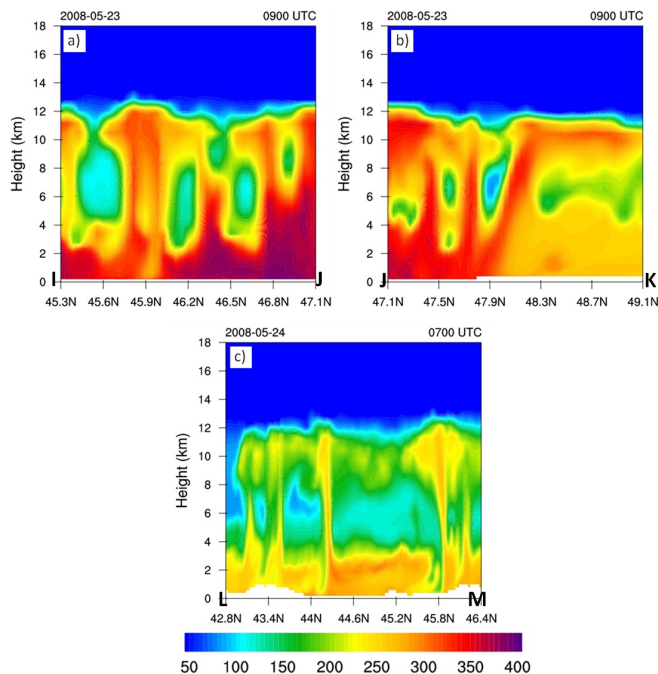


Fig. 12. Vertical cross sections of CO (ppbv) parallel to the squall line (**a, b**) at 09:00 UTC 23 May and through the multi-cell convective region (**c**) at 07:00 UTC 24 May using WRF-Chem output from D3. Axes of the two cross sections are shown in Fig. 13b, d.

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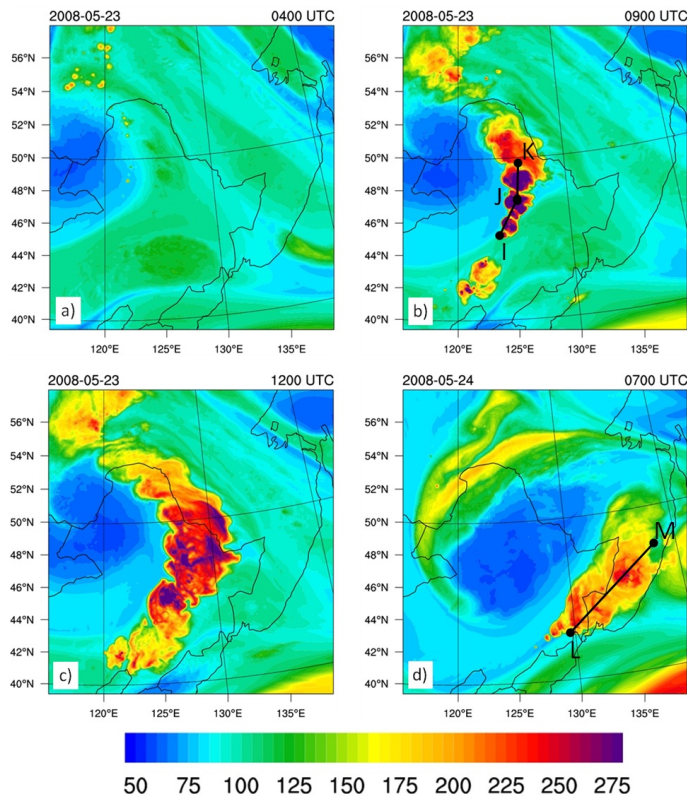


Fig. 13. CO concentration (ppbv) at 10 km at four different times using WRF-Chem output from D3. Axes of cross sections I–J at 09:00 UTC 23 May (**b**) and L–M at 07:00 UTC 24 May (**d**) that are used in Fig. 12 also are shown.

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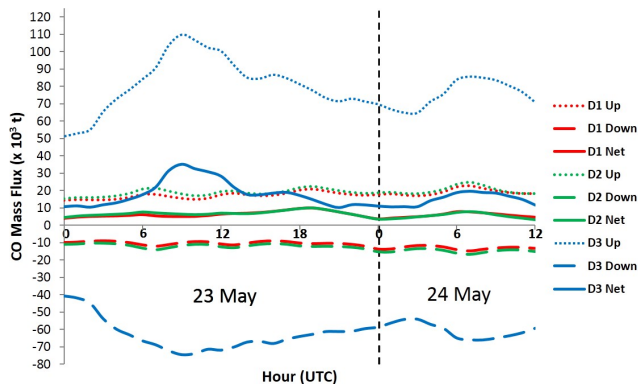


Fig. 14. Time series of hourly CO mass flux ($\times 10^3$ metric tons, t) at 500 hPa starting at 00:00 UTC 23 May and ending at 12:00 UTC 24 May (36 h duration) within D3. Results from D1 and D2 were computed only within the area of the D3 domain. Upward, downward, and net fluxes are shown.

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Transport of CO in a middle latitude cyclone

C. A. Klich and H. E. Fuelberg

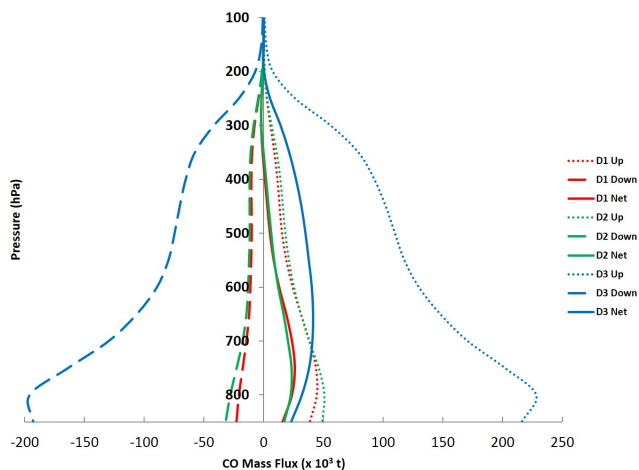


Fig. 15. Vertical profile of CO vertical mass flux ($\times 10^3 t$) within domain D3 at 09:00 UTC 23 May. Upward, downward, and net fluxes are shown for all three WRF-Chem horizontal resolutions as described previously in the text.

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