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Transpacific transport of benzo[a]pyrene emitted from Asia

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Abstract. A global-scale three dimensional atmospheric transport and chemistry model was applied to simulate transpacific transport of Benzo[a]pyrene (BaP) emitted from Asia. The model results were compared with observations at six monitoring sites. The annual mean and seasonal variation of transport patterns and the contributions of different Asian source regions to transpacific transport flux were investigated. The episodic nature of transpacific transport was also systematically explored. Interannual variability of transpacific transport of BaP was also assessed during the period of 1948–2007. Results showed that strong enhancements of modeled BaP occurred in an area bounded by 70–80° E and 100–120° E. Air containing these elevated BaP concentrations was then delivered eastward by westerly winds. When approaching the West Coast of North America, the descending atmospheric motion carried BaP-laden air into the lower atmosphere. The transpacific transport flux was 1.6 times higher in the winter than in the summer. East Asian emission dominates the transpacific transport flux with a contribution of about 97 %. Near ground concentration of BaP induced by Asian sources in North America varied between 1–20 pg m^{-3}. A case study for observation at Cheeka Peak Observatory during March 2002–May 2002 reveals the importance of warm conveyor belt for transpacific transport. The number of days with transpacific transport flux with a factor of 0.5, 1.0, 1.5, and 2.0 larger than the running mean were 9.4 %, 0.72 %, 0.06 % and 0.01 %, respectively, implying a mild contribution of episodic transport to the long-term mean transport flux. Significant interannual fluctuation of transpacific transport of BaP was found, including a general decreasing trend during 1948–2007, and especially after the 1970s. The transpacific transport was found to be positively correlated with the Southern Oscillation Index.

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1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) have been included in the Convention on Long range Transboundary Air Pollution Protocol on Persistent Organic Pollutants (United Nations Economic Commission, http://www.unece.org/env/lrtap/pops_h1.htm) and linked with human health (Armstrong et al., 2004). PAHs are mainly generated from incomplete combustion of carbonaceous bio- and fossil fuels. Despite the drastic decrease of PAH emissions in developed countries due to PAH emission reduction efforts (Pacyna et al., 2003), PAH deposition on the ice sheet in Greenland showed the global trend of PAH emission was nearly constant from the beginning of the industrial period to the early 1990s (Masclet et al., 1995). One of the reasons is increasing PAH emission due to rapid economic development and population increases in Asia which compensates for the emission reduction in developed countries (Zhang et al., 2008b). According to a global emission inventory of PAHs, the total emission of sixteen PAH compounds listed in the United States Environmental Protection Agency (USEPA) priority control list from Asia was about 290 Gg y^{-1}, accounting for more than half of total global emissions (Zhang and Tao, 2009). Given considerable long range transport potential of many

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PAH compounds (Lang et al., 2008; Primbs et al., 2008; Klasmeier et al., 2006), higher emissions of PAHs from Asia might threaten ecological environments and human health on an intercontinental or even global scale.

A number of observations of transpacific transport of PAHs have been documented in the literature. Surface concentration measurements combined with backward trajectories have shown that semi-volatile organic compounds (SVOCs), including PAHs, can be transported across the Pacific Ocean in about a week under certain meteorological conditions (Primbs et al., 2007; Primbs et al., 2008; Killin et al., 2004). For example, Killinet al. (2004) observed significantly elevated concentrations of PAHs and other SVOCs during 15–16 March, 27–28 March and 22–23 April 2002, when backward trajectories passed over Asia, suggesting Asia as the potential source of these compounds.

Several modeling studies have also addressed the transpacific transport of particulate and gaseous pollutants derived from incomplete combustion of carbonaceous fuels including CO and Hg (Liu et al., 2003; Liang et al., 2004, 2005; Strode et al., 2008), and SVOCs such as Polychlorinated Biphenyls (Huang et al., 2007). In these studies, the warm conveyor belt (WCB) was identified as an important pathway for transpacific transport. For instance, Liang et al. (2004) investigated transpacific transport events occurring from March 2001 to late May 2002. They found that lifting from the surface to the free troposphere ahead of surface cold fronts in WCBs was the main pathway ($\sim$70 %) of the transpacific transport events occurring in this time period; other processes, including deep convection and transport in the atmospheric boundary layer behind cold fronts, accounted for remaining $\sim$30 %. After the lifting of pollutants from the surface to the free troposphere where the wind is stronger and fast transpacific transport is possible, single or multiple succeeding WCBs are also strongly associated with this advection process (Cooper et al., 2004). Additionally, significant seasonal variation of transpacific transport due to the frequency of cold surge events and the East Asian Monsoon system has been reported in the literature (Liu et al., 2003).

Owing to the strong affinity of BaP to aerosols and subsequently subjected to stronger dry and wet deposition, the overall lifetime of BaP is much shorter than Hg and CO (Liu et al., 2003; Strode et al., 2008). However, modeling studies of transpacific transport of PAHs have not been carried out and reported in the literature except for the study by Lang et al. (2008). In that investigation, Lang et al. (2008) applied a forward trajectory method to assess the outflow of pyrene emitted from China to different regions from 1996 to 2005, and found a mean transit time of $>$9 days and an outflow mass of $\sim$0.5 tons for North America. The trajectory approach is subject to substantial uncertainty due to the large uncertainty in extensive trajectory calculations as well as a highly simplified chemistry scheme (Kahl and Samson, 1986). Other PAH modeling studies either mainly focus on the regional air quality and fate (e.g. Matthias et al., 2009; Zhang et al., 2009; Sehili and Lammel 2007) or the importance of the gas/particle partitioning scheme (e.g. Prevedouros et al., 2008; Lammel et al., 2009).

The objectives of this study were to (1) model the transpacific transport of PAHs using a state-of-the-art model with complete atmospheric chemistry and multi-compartment fate and exchanges, (2) identify background transport patterns and episodic transport events, with a special focus on WCB transport, (3) highlight the contribution of different Asian regions to transpacific transport, (4) quantify the concentration of PAHs in North American induced by Asian sources, and (5) investigate the seasonal cycle and interannual variations of transpacific transport of PAHs. Due to its high emission rate and extremely strong carcinogenic effect (Zhang and Tao, 2009; Zhang et al., 2009; Nisbet and Lagoy, 1992), benzo[a]pyrene (BaP) was chosen as the representative compound for PAHs in this study.

2 Methodology

2.1 Model description

The global-scale three dimensional atmospheric transport and chemistry model CanMETOP (Canadian Model for Environmental Transport of Organochlorine Pesticides) (Ma et al., 2003; Zhang et al., 2008a, 2010) was modified and applied in this study. Other modifications to a regional scale version of this model, which was successfully used in modeling the transport of PAHs in China (Zhang et al., 2009, 2011), were also merged into this global model. The CanMETOP model was successfully used to model the transpacific transport of lindane (Zhang et al., 2008a) and the long range transport of hexachlorocyclohexane to the Arctic region (Zhang et al., 2010).

Briefly, the atmospheric transport module of this model is based on the atmospheric advection-diffusion equation, which was discretized to the model grids at $1^\circ \times 1^\circ$ latitude/longitude and 14 vertical levels (with 6 below 1 km), and then solved numerically. The details of the numerical schemes are described elsewhere (Ma et al., 2003; Zhang et al., 2008a). In order to better handle the transport over high latitudes and especially over the Polar Regions, the horizontal advection was solved by a multi-dimensional flux-form semi-Lagrangian transport scheme (Lin and Rood, 1996). The meteorological data (including wind, pressure, temperature, precipitation, etc.) were obtained from the National Centers for Environmental Prediction (NCEP) reanalysis (Kalnay et al., 1996), and interpolated at each 30-minute model time step. A non-local and higher order turbulence closure scheme was used in this model to estimate the momentum and heat fluxes and to characterize the sub-grid scale process caused by the coarse meteorological data and surface heterogeneity (Ma et al., 2003). The dry deposition velocity is calculated by a resistance-in-series scheme following Ma

et al. (2003). As BaP can partition between gaseous and aerosol phases, the wet deposition in both phases are considered. For the gaseous phase, the wet deposition flux is proportional to the precipitation intensity and the solubility of BaP in rain water. The temperature-dependent solubility of BaP is calculated by its Henry's Law constant. The fraction of wet scavenging in the aerosol phase is calculated as exponential dependent on the precipitation intensity (Ma et al., 2003).

Similar to the regional-scale version of CanMETOP for PAHs (Zhang et al., 2009), a level-IV fugacity model is coupled with the atmospheric module. Fugacity is defined as the tendency of a chemical to leave a certain phase compartment (Mackay 2001), and the corresponding concentration can be calculated as the product of the fugacity and the chemical capacity. In this model, the soil/air and ocean/air exchange at the bottom of the atmosphere, gas/particulate partition of semi-volatile PAH compounds, degradation by OH radicals in the atmosphere, biodegradation and leaching in soils, and degradation and exchange between surface and deep ocean are considered and simulated by a fugacity approach (Zhang et al., 2009, 2011). These processes are briefly described below.

We treat the transport and fate of PAHs in soil largely following Ma et al. (2003) and Harner et al. (1999). The fugacity of PAHs in the fast exchanging layer (f_{s1} 0.1 cm), buffer layer (f_{s2}, 1 cm), and reservoir soil layers (f_{s3}, 10 cm) are modeled by the following set of equations:

$$\begin{cases} \frac{d(f_{s1}Z_{s1})}{dt} = \left[f_a\left(D_q + D_d + D_m\right) + f_{s2}D_{v1,2} - f_{s1}\left(D_{v1,2} + D_L + D_R\right)\right]/v_{s1} \\ \frac{d(f_{s2}Z_{s2})}{dt} = \left[f_{s1}\left(D_L + D_{v1,2}\right) + f_{s3}D_{v2,3} - f_{s2}\left(D_{v1,2} + D_{v2,3} + D_L + D_R\right)\right]/v_{s2} \\ \frac{d(f_{s3}Z_{s3})}{dt} = \left[f_{s2}\left(D_L + D_{v2,3}\right) - f_{s3}\left(D_{v2,3} + D_L + D_R\right)\right]/v_{s3} \end{cases} \quad (1)$$

where f_a is the fugacity in the atmosphere above, f_{s1}, f_{s2} and f_{s3} are the fugacity in the three soil layers, and Z and v are the chemical capacity and volumes of the soil, respectively. The D values describe the rates of transport and transformation processes in the soil, which include vertical diffusion between layers ($D_{vn,n+1}$), biological reduction (D_R), leaching (D_L), dry deposition (D_q), wet deposition (D_d), and rain dissolution (D_m). The details of these D values are given by Harner et al. (1999).

The ocean is considered as a series of independent slabs without lateral advection and diffusion. These slabs have the same horizontal spatial resolution as the atmospheric grid boxes above and are divided into surface (with a depth of 30 m) and deep ocean boxes (depth 4000 m) following Wania (1995). The fugacity of PAHs in the two layers (f_{w1} and f_{w2} for the surface and deep layers, respectively) are predicted by Eq. (2):

$$\begin{cases} \frac{d(f_{w1}Z_{w1})}{dt} = \left[f_a\left(D_q + D_d + D_m\right) + f_{w2}D_{v1,2} - f_{w1}\left(D_{v1,2} + D_S + D_R\right)\right]/v_{w1} \\ \frac{d(f_{w2}Z_{w2})}{dt} = \left[f_{w1}\left(D_S + D_{v1,2}\right) - f_{w2}\left(D_{v1,2} + D_S + D_R\right)\right]/v_{w2} \end{cases} \quad (2)$$

where the D, Z and v are similar to those for the soil compartment. Besides, we also consider the settling of PAHs associated with organic matter (D_S), which is calculated by Eq. (3) based on Dachs et al. (2002):

$$D_s = \mathrm{BCF} \times F_{om} Z_{w1} \quad (3)$$

where BCF and F_{om} are the bioconcentration factor of PAHs between ocean water and phytoplankton and the settling velocity of organic matter in the surface ocean water, respectively.

The exchange fluxes ($F_{soil/ocean-air}$) of PAHs between the atmosphere and the underneath soil/water are then calculated based on the difference of fugacity across the interface between compartments:

$$F_{soil/ocean-air} = D_v(f_{soil/ocean} - f_{air}) \quad (4)$$

where the f-values are the fugacity of PAHs in different compartments, while the transport coefficients (D_v) was described by Ma et al. (2003). The initial values of fugacity in the soil and ocean water are generated by an iteration approach with constant PAH emissions until a steady state is achieved.

A certain fraction of the PAHs is assumed to adsorb onto the surface of aerosols in the atmosphere (Pankow et al., 1987), and the fraction of particulate associated (θ) is calculated according to the surface area of aerosol (S_{tot}) and the vapor pressure of PAH compounds (P_s), as shown in Eq. (5):

$$\theta = \frac{cS_{tot}}{cS_{tot} + P_s} \quad (5)$$

where c (17.2 pa cm) is an independent constant.

The reaction of atmospheric PAHs with OH radicals is considered for gaseous phases, whereas the degradation in particulate phase is neglected in this study. The reaction coefficient (k_{OH}) is temperature (T) dependent and calculated by Eq. (6):

$$k_{OH} = k^0_{OH} \exp(a(1/T_0 - 1/T)) \quad (6)$$

where K^0_{OH} is the preexponential factor and a is the active energy. We used the monthly archived data from GEOS-Chem model for the total aerosol surface area and OH radical concentrations (ftp://ftp.as.harvard.edu/gcgrid/data). The aerosol-adsorbed BaP was assumed to be non-react to oxidants in gas phase because of the lack of experimental data and much smaller reaction coefficient (Estève et al., 2006). Based on the reaction estimation by Schauer et al. (2003), the residence time of particulate phase reduction of BaP is on the order of a couple of days due to heterogeneous reaction with O_3 and other photo-oxidants in the atmosphere. The neglecting of this effect could cause an overestimation of the residence time of BaP in the atmosphere for ~30 % in this study.

2.2 Emission inventory

Zhang and Tao (2009) have developed a global emission inventory based on a compiled emission factors database

and emission activity data collected by the International Energy Association and other related sources. This inventory estimates the total PAH emission for each country in 2004, and also provides data on the socio-economic status of these countries including the total population, the Gross Domestic Product and the per-capita income level (Zhang and Tao, 2009). Regression models were also built between the PAH emissions and these parameters (Zhang and Tao, 2009). Based on these relationships, the national PAH emissions over Asian countries were interpolated into the model grid, using the population statistics of each grid cell as a weighting factor. The national population data were obtained from the LandScan™ global population dataset with a spatial resolution of 1 km×1 km which was further resampled and merged into a 1°×1° latitude/longitude grid (Oak Ridge National Laboratory, Geographic Information Science and Technology, the database can be accessed via: http://www.ornl.gov/sci/landscan/index.shtml). For simplicity, we cut off the BaP emissions west of 60° E due to their small portion compared with that from the whole continent.

The BaP emissions other than the year 2004 are not available in the emission inventory developed by Zhang and Tao (2009). We therefore adopted the emission inventory developed by Zhang et al. (2008b) for China. This inventory investigated the interannual variation of PAH emissions from 1948–2007. The BaP emissions over other countries were scaled by the historical population data by assuming that the emission is proportional to the population. The population data for each country during 1948–2007 were obtained from the Statistics Division of the United Nations (http://unstats.un.org/unsd/demographic/sconcerns/popsize/default.htm).

2.3 Reference compound and year

The model was run with 2004 as the reference year and full BaP emissions over the entire Asian continent (east of 60° E). The interannual variation of transpacific transport of BaP was also investigated from 1948–2007 with the population-scaled emissions.

2.4 Comparison with observations

The model results were compared with the observations at six monitoring sites including Changdao, Gosan, Kanazawa, Okinawa, Cheeka Peak Observatory (CPO), and Mount Bachelor Observatory (MBO) during different time periods when observations were actually conducted (Feng et al., 2007; Lee et al., 2006; Tamamura et al., 2007; Primbs et al., 2007; Killin et al, 2004; Primbs et al., 2008). A map of the location of these sites as well as a comparison of their measured concentrations is shown in Fig. 1a. The total PAH concentrations, instead of BaP concentration, were used as a surrogate to compare with our model results, when individual compound concentrations were not reported or not detected (Fig. 1c, d, f and g). Changdao is an island near Shandong, China. Figure 1b shows modeled and measured seasonal mean BaP concentrations at the Changdao site during 2003–2004. The model underpredicted the concentrations in winter, partly because of underestimation of the seasonal variation of PAH emissions in this region (Zhang and Tao, 2008). At the Gosan site, modeled concentrations show a good correlation with monitoring data (Pearson correlation coefficient $r = 0.67$), but the model tends to underestimate BaP concentrations in most winter cases from 2001–2004. Generally, the predictive power decreases as the distance between observation site and source areas increases, due to the weaker contribution from Asian sources as compared with local sources that were not represented in the model. Although a good agreement between modeled and measured concentrations was achieved in the western Pacific Ocean (Okinawa in the spring of 2004, $r = 0.37$, Kanazawa in 2003–2004, $r = 0.80$), the model produces larger bias in predicting the concentrations in the western seaboard of North America, as illustrated by Fig. 1f. This figure shows that the model does not entirely capture the observed data before May 2002, when the local sources may dominate. Nevertheless, the model successfully simulated the concentrations after 3 May 2002 and satisfactorily captured the concentration peak on 12 May 2002 ($r = 0.98$ if only this time period was considered). During this period the Intercontinental Transport and Chemical Transformation (ITCT 2K2) experiment was also conducted, during which the concentrations of different pollutants were measured by an aircraft campaign over the West Coast of the United States (near Monterey, California). Results from this field campaign are discussed elsewhere (Allan et al., 2004; de Gouw et al., 2004; Goldstein et al., 2004; Nowak et al., 2004). At the MBO site, the modeled concentrations were poorly correlated with measured concentrations (r=0.09),likely because of the high altitude (2700 m a.s.l.) and strong influence by local North American emissions of this site (Primbs et al., 2008). At this site local wind patterns are strongly influenced by the local topography, which cannot be captured by course wind data in a global-scale transport model. Our model also underestimates the average BaP concentration (8 points were detected out of total observation of 69) by a factor of 3, indicating a dominate contribution from local sources. Nevertheless, the overall agreement between the model and the observations at CPO when Asian source dominates provides an acceptable validation of the model when it is used to quantitatively assess the large scale transpacific transport of PAHs emitted from Asia.

3 Results and discussion

3.1 Annual mean transport pattern

Figure 2a presents the annual mean BaP concentrations and wind vectors at the height of 3 km across the Pacific. The vertical profile of the meridional mean concentration of BaP

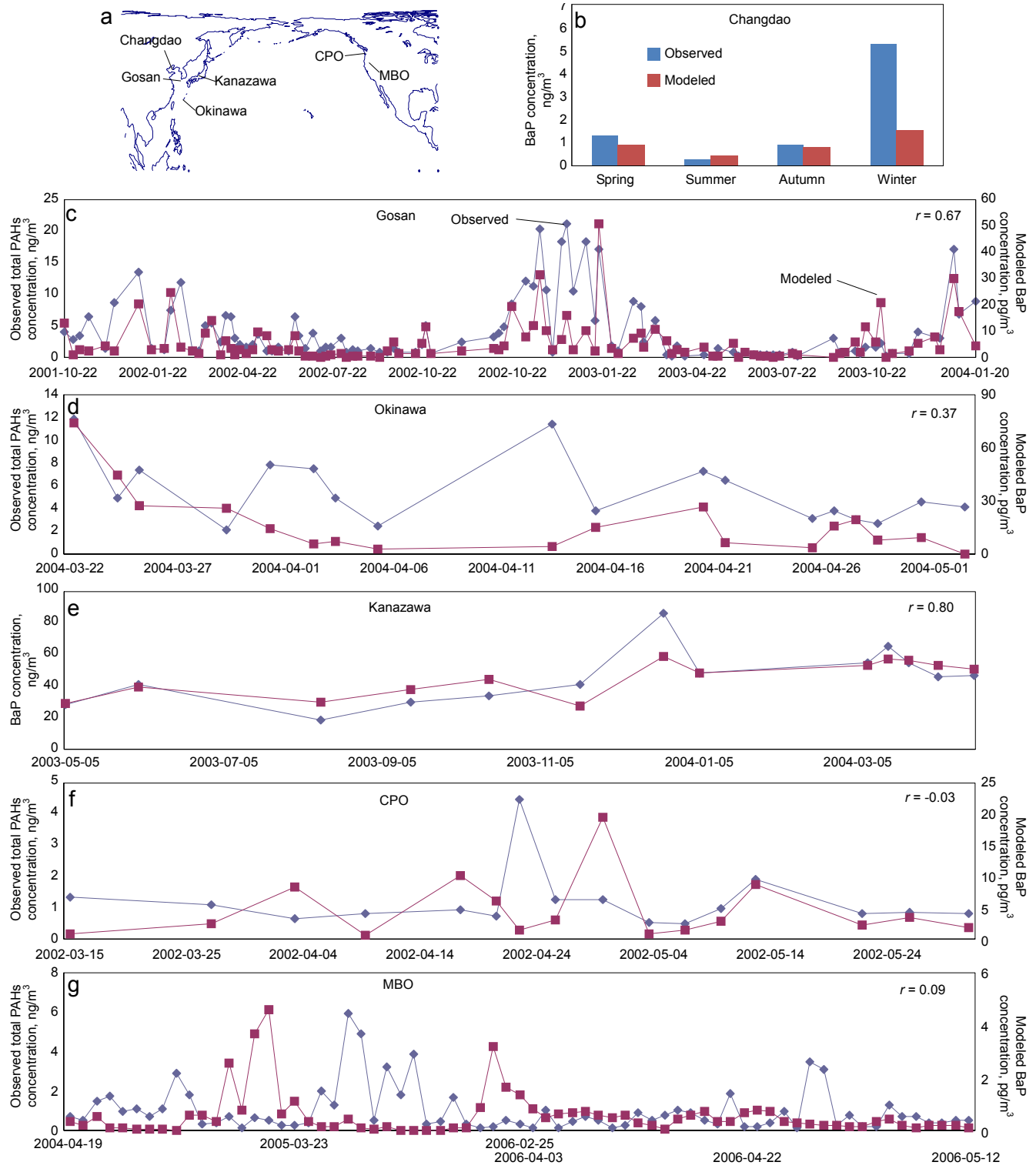

Fig. 1. Comparison of the modeled BaP concentrations with observations at different sites: **(a)** a map of the locations of these sites; **(b)** seasonally averaged total BaP concentration at Changdao, China during 2003–2004; **(c)** comparison of weekly averaged particulate phase total PAH concentration at Gosan, Korea with modeled total BaP concentration during 2001–2004; **(d)** comparison of total PAH concentration at Okinawa, Japan with modeled total BaP concentration during May, 2004; **(e)** comparison of total BaP concentration at Kanazawa, Japan with modeled total BaP concentration during 2003–2004; **(f)** comparison of the total PAH concentration at CPO, US with modeled total BaP concentration during March–May, 2002; **(g)** comparison of total PAH concentrations over MBO, US with modeled BaP concentration during 2004–2006.

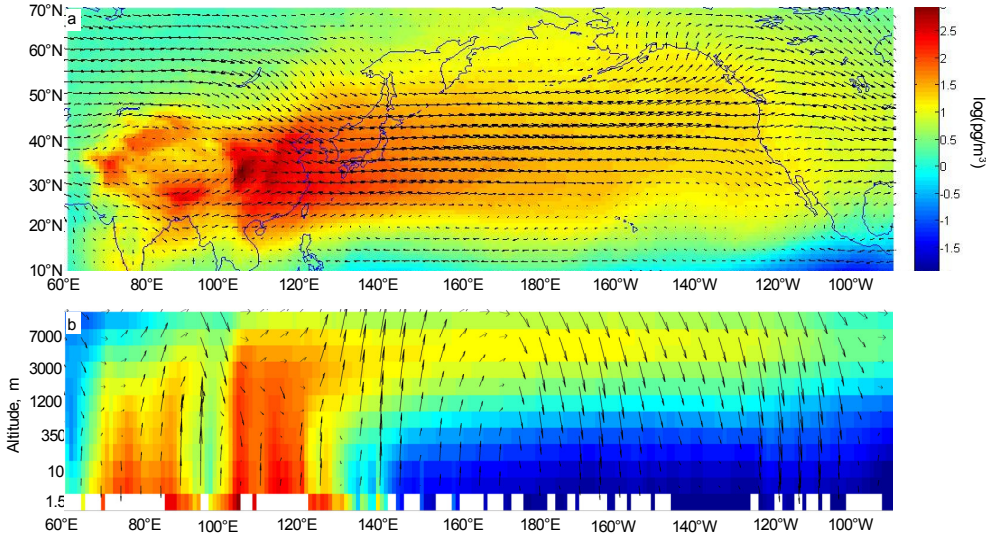

Fig. 2. Annual mean total BaP concentration (pg m^{-3}) over the northern Pacific Ocean in 2004: **(a)** horizontal distribution of total BaP concentration at 3 km above ground; **(b)** a cross section showing the meridional mean concentration between 25–55° N.

averaged over 25–55° N is also shown in Fig. 2b. Significant enhancements of modeled BaP concentrations occurred between 70–80° E and 100–120° E (Fig. 2b), corresponding to West Asia and East Asia, respectively. Over East Asia, under the influence of convective injection and mid-latitude cyclones, BaP emitted from Siberia, Mongolia, and North and Northeast China were lofted to the free atmosphere and transported southeast to the western Pacific Ocean. The mechanism of this transport pattern has been extensively explored by several modeling studies (Liang et al. 2004, 2005; Zhang et al., 2011). Over West Asia (70–80° E), the upward transport of BaP is weaker and largely contained under 3 km, corresponding to the low concentration levels in this region, as shown in Fig. 2a. The mean ascending motion over this zone is largely forced by the Tibetan Plateau. The divergence of air parcels delivered by westerly flows produces an upward motion over the upslope of the plateau, whereas on the south of the plateau the winter India monsoon and the plateau interact to form a strong descending motion in the winter (Zhang, 1991). Therefore, despite the high emissions of PAHs in South and Southeast Asia, the descending air parcels over this region inhibit the lofting of pollutants from these regions (Wallace and Hobbs, 2005; Shrestha et al., 2000).

The BaP-laden air parcels were transported eastward by the westerly winds after they rose high in the atmosphere. Liang et al. (2004) summarized the main mechanisms of transpacific transport in two categories: advection in the free troposphere by westerly flow and direct boundary layer transport. Although under some episodic conditions transpacific transport occurs predominantly below 3 km (Jaffe et al., 1999), our model results show that the largest transport flux occurs at a height of 3–5 km each year. When approaching the western coast of North America, the descending air parcels associated with dry air streams behind cold fronts deliver BaP to a lower atmosphere (Liang et al., 2004). The downward wind vectors near 120° W and the slightly enhanced BaP concentrations near the surface as seen Fig. 2b also demonstrate this point. The magnitude and spatial pattern of BaP concentration induced by Asian emission will be discussed in Sect. 3.4 with more detail.

3.2 Seasonal variation of transpacific transport

Significant seasonal variations of transpacific transport of different pollutants and tracers have been reported in the literature (Liang et al., 2004; Liu et al., 2003). Figure 3 shows the seasonal variation of the transpacific transport of BaP. Modeled mean column concentrations of BaP at North America (130° W–105° W, 20° N–60° N, as shown the black box in Fig. 3c) induced by Asian source vary between 5.3×10^4 and 3.3×10^5 pg m^{-2} in different seasons. The higher transport flux in winter is partly caused by the higher BaP emission in Asia. With bio- and fossil fuel consumptions as the major sources, the emission of BaP is much higher during the cold season than the warm season in the mid- and high-latitude areas of Asia. The emission flux over the Asian continent varied between ∼160 and ∼320 t month^{-1}. Another important factor attributable to the lower transport flux in summer is the stronger OH radical degradation of BaP. The degradation flux was ∼120 t month^{-1} in July, but only 70 t month^{-1} in January.

Besides the influence of emissions and removal processes, significant seasonal displacements and variations of the semi-permanent pressure systems and the westerlies across the North Pacific Ocean also contribute to the seasonal variation of transpacific transport (Liang et al., 2005; Wallace and

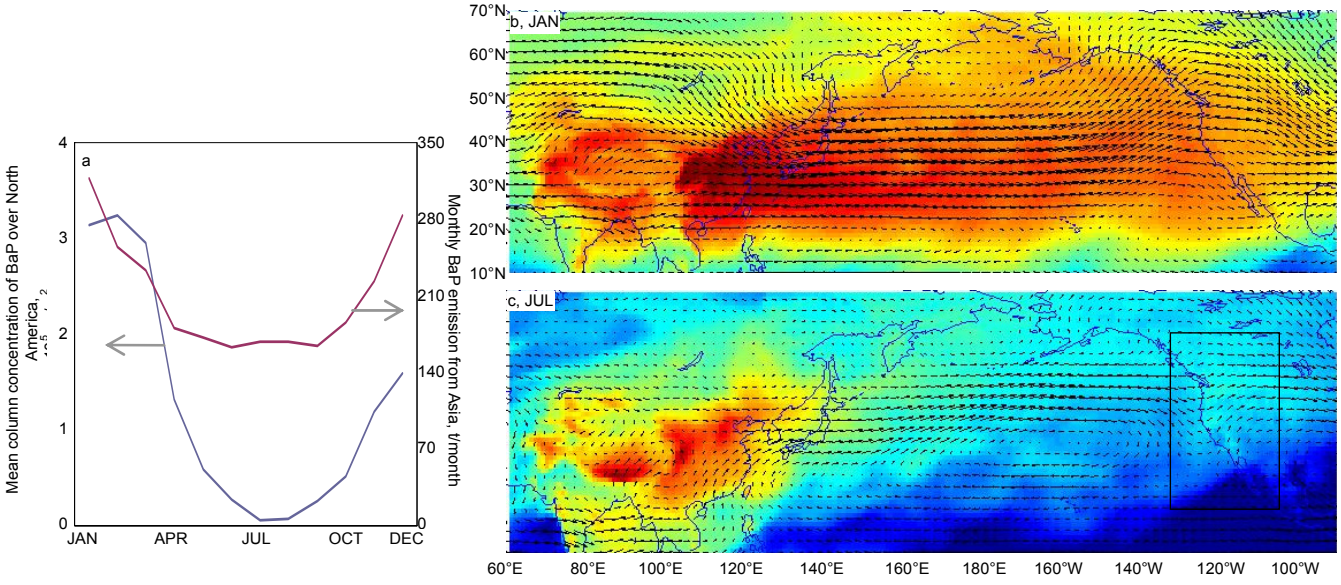

Fig. 3. Seasonal variation of transpacific transport of BaP in 2004: **(a)** monthly mean column BaP concentration over North America (blue curve, the region was shown as the black rectangle in **(c)**), which was chosen as an indicator of transpacific transport of BaP; the monthly emission flux over Asia (brown curve) is also shown; **(b)**, **(c)** monthly mean total BaP concentration in January and July, 2004 at a height of 1200 m above ground over the northern Pacific Ocean; the color bar is the same as that in Fig. 2.

Hobbs, 2005). Figure 3b, c presents the concentrations and the corresponding wind vectors for January and July 2004, respectively. In winter, the westerly flow and winter monsoon may carry BaP emitted from northern China to the western Pacific where the westerly winds reinforced by counterclockwise air flows on the south of the Aleutian Low further deliver the pollutant eastward. Over the eastern Pacific, the eastward flow reinforced between the Aleutian Low and the Pacific High finally pushes the BaP-laden air parcels to the western seaboard of North America. However, with the onset of the East Asian summer monsoon – which features southeasterly winds extending from the western Pacific to East China (Tian et al., 2009) and the northward displacement of the semi-permanent high pressure system over the North Pacific Ocean – the outflow of BaP from North China to the western Pacific becomes almost unlikely at lower atmospheric levels. Although an eastward returning flow at the top (about 3000 m) of the East Asian summer monsoon system could transport the pollutants emitted from Asia to the western Pacific, the strength and spatial scale of this returning flow is much less than the outflow flux in winter. The northward movement of the Pacific High grows more intense in summer and extends from 15 to 60° N, which blocks the eastward transport of air masses over the North Pacific Ocean (Wallace and Hobbs, 2005).

3.3 Contribution from different Asian sources

The modeled annual mean BaP concentrations at 1200 m altitude over North America originated from different regions of Asia are shown in Fig. 4a–f. As shown in Fig. 2, the model has predicted significant outflow flux from the Asian continent, illustrated by a plume crossing over 25° N and 45° N and extending to the western Pacific Ocean. The main contributors to this plume are emissions over East Asia (Fig. 4b). Despite of the high emission density in South Asia (Fig. 4c) and the Indochina Peninsula (Fig. 4e), the contributions from these two sources are much smaller compared with that from East Asia due to the general descending movement of air parcels in the downdraft of the Hadley cell (20–30° N). The dominant easterly winds in the subtropics further constrain the transpacific transport from this part of Asia. On the contrary, although BaP emission from West Asia can be effectively lofted to a higher atmospheric level (Fig. 2b), the contribution of BaP emissions from East Asia is less significant due to a longer travel distance (Fig. 4f). The contribution from North and Southeast Asian BaP emissions is minimal due to considerably lower emissions compared with emissions in East and South Asia (Fig. 4a, d). If the mean concentration of BaP over the western coast of North America (defined as a box between 30–50° N and 120–130° W) was taken into consideration, the East Asian emissions contributed about 97 % to BaP mixing ratios, while other regions contributed at the following percentages: South Asia at 1.3 %, West Asia at 1.1 %, North Asia at 0.68 %,

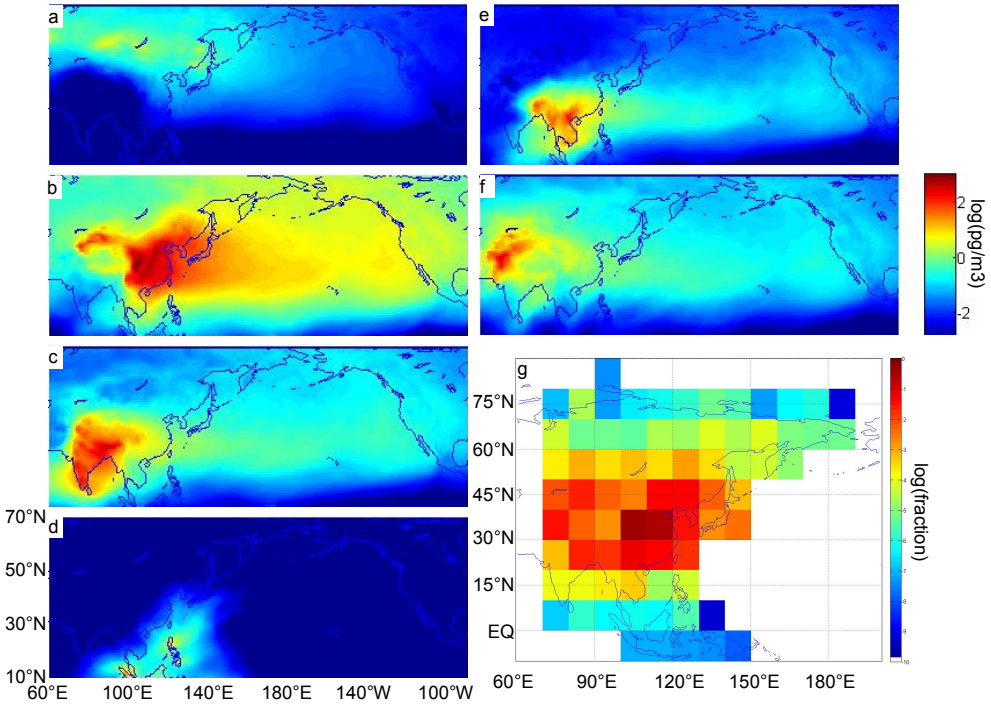

Fig. 4. Annual mean concentration of BaP induced by emissions from different Asian sources: **(a–f)** North Asia, East Asia, South Asia, Southeast Asia, the Indochina Peninsula, and West Asia, respectively; **(g)** contribution of the Asian BaP emission sources to the average BaP concentration over North America in 2004; each box represents a 10 degree by 10 degree area.

the Indochina Peninsula at 0.33 %, and Southeast Asia at 0.0052 % (Fig. 4). We further divided Asia into grids of $10° \times 10°$ latitude/longitude and estimated the contributions of BaP emissions from these grid boxes to the mean BaP concentration over North America due to transpacific transport. Results are shown in Fig. 4g. It is evident that the emission from East Asia between 20° N and 50° N made the largest contribution to the mean BaP mixing ratio over North America, primarily due to the high emissions over this region and favorable climate conditions for long-range transpacific transport. The contribution from other Asian regions becomes less important because of their lower BaP emissions and unfavorable climate and meteorological conditions.

3.4 Transport budget and near ground concentration in North America

Figure 5 shows the modeled annual mean BaP concentration at 10 m above ground level over North America. The concentration was enhanced by about $4\,\mathrm{pg\,m^{-3}}$ (with a range of $1{\sim}20\,\mathrm{pg\,m^{-3}}$), indicating a slight enhancement of the background BaP concentration over North America (which is $\sim 100\,\mathrm{pg\,m^{-3}}$, Naumova et al., 2002) due to Asian emissions. The BaP-laden air masses approaching North America spread over a wide latitudinal band between 25–70° N. The concentrations over the western coast of North America were the highest due to the subsidence of air parcels in this region (Fig. 2b), which entrain the BaP-laden air originating from Asian sources into the boundary layer. The surface concentrations induced by the Asian emissions were more than $6\,\mathrm{pg\,m^{-3}}$ in the western North America, much higher than the $\sim 2\,\mathrm{pg\,m^{-3}}$ in Central and Eastern America. The lowest concentration was modeled above seasonal or permanent ice surfaces (e.g. the center of Greenland) where the ground friction is minimal, and the weak eddy diffusion cannot efficiently transport the pollutants to the surface (Zellner et al., 2000). In 2004, the total mass of BaP is about 1.4 t within a box bounded by 30–70° N, 75–125° W and 0–7 km altitude. Of this BaP, about 6.7 t is degraded by OH radicals and 0.6 t is deposited onto the surface. We also calculated the fraction of atmospheric BaP emitted from North American soils, and found that the reemission from North American soils enhances the background concentration by only $\sim 10^{-4}\,\mathrm{pg\,m^{-3}}$, and therefore is negligible even though soil is the largest reservoir for BaP (Sehili and Lammel, 2007).

3.5 Episodic transport mechanism

Many studies dedicated to investigating the episodic nature of transpacific transport indicate stronger transport of pollutants than background level under certain meteorological conditions (Lee et al., 2006; Tamamura et al., 2007; Primbs et al., 2007; Killin et al., 2004; Primbs et al., 2008). On 5 May 2002, the Intercontinental Transport and Chemical

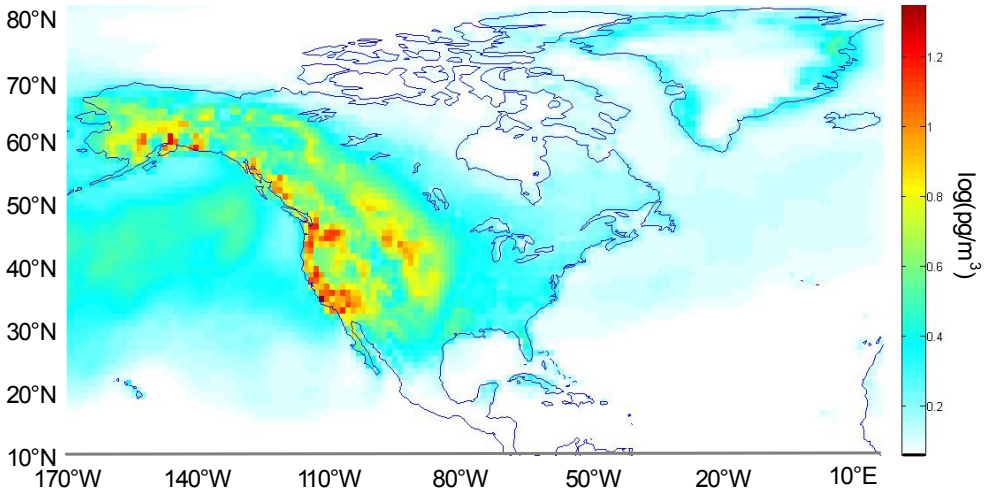

Fig. 5. Annual mean BaP concentration (pg m^{-3}) at 10 m above ground level induced by Asian emission sources in 2004.

Transformation (ITCT 2K2) experiment was conducted over the West Coast of North America, and air masses containing pollutants originating from Asia were measured by aircraft over a region bounded by 32–44° N, 122–125° W, and 0–8 km altitude (Allan et al., 2004). Weekly monitoring of PAH concentrations at CPO was also conducted during March 2002–May 2002 (Killin et al., 2004). A transpacific transport event was captured on 12 May 2002 (Fig. 1f). In the present study, CanMETOP also captured this transpacific transport event. This event is closely associated with the ITCT 2K2 experiment carried out over the region about 1000 km south of CPO, which has been discussed in the literature (Allan et al., 2004; Cooper et al., 2004; de Gouw et al., 2004; Goldstein et al., 2004; Nowak et al., 2004;). The role of warm conveyer belt (WCB) was also identified and discussed by using satellite imagery, trajectory ensembles and in situ aircraft measurements (Cooper et al., 2004). Therefore, this event will be discussed as a representative case to investigate the role of WCB in transpacific transport of BaP.

BaP concentrations and wind vectors during this transport event at a height of 1200 m above ground are shown in Fig. 6. In later April, a low pressure system was centered in Northeast China and the associated warm conveyer belt (WCB) helped to elevate air parcels to a relatively higher atmospheric level. This low pressure system moved eastward and headed to 170° E on 3 May and to the International Dateline on 4 May. Correspondingly, a typical comma-shaped distribution of elevated BaP can be seen in Fig. 6b (circled region). This is also a typical cold front type distribution. With further eastward movement, this low pressure system weakened considerably due to (sea) surface cooling, so the surface front associated with this low pressure system was no longer identifiable (Fig. 6c). On the same day, it can be seen that BaP concentrations extended from the central North Pacific (150° W) to the West Coast of Oregon State (near 45° N, 120° W), and some of the air parcels originating from eastern Asia even penetrated inland up to 100° W. This transport event was captured by the aircraft monitoring on 5–6 May on the western coast of North America (near 36° N, 120° W). However, the plume did not extend to the CPO monitoring site because the PAH concentration measured at this site was not enhanced. This phenomenon has been clearly modeled in this study (Fig. 6c).

Likewise, another low pressure system moved to the east coast of China, forming a southwesterly flow over the western North Pacific (Fig. 6c), just ahead of the cold front associated with this low pressure system. This southwesterly flow moved to the central North Pacific (about 180° E and 135° W on 7 May), resulting in another cold-front-type distribution of BaP, as shown in Fig. 6d. Because of the northward extension of a North Pacific high pressure system, this southwesterly flow was forced to turn west (Fig. 6e). Responding to the interaction of these changing meteorological conditions, BaP-laden air parcels were pushed eastward to the west coast of Oregon (Fig. 6e and f). Meanwhile, these air parcels descanted as the decay of this low pressure system (Cooper et al., 2004) resulting in the enhanced BaP concentrations measured at the CPO site. On 12 May, higher concentrations were observed over the western coasts of Washington State and British Columbia (circled in Fig. 6f), and a significant peak of PAH concentrations was also observed at the CPO site (Fig. 1f).

We also conducted a systematic study to further demonstrate the episodic nature of the transpacific transport. As shown in the aforementioned case, the episodic transpacific transport occurred over approximately one week. We calculated the eleven-day running mean of the BaP concentration in North America averaged over a box bounded by 32–44° N, 122–125° W and 0–8 km altitude between 1948 and 2007. The BaP concentration anomaly was calculated

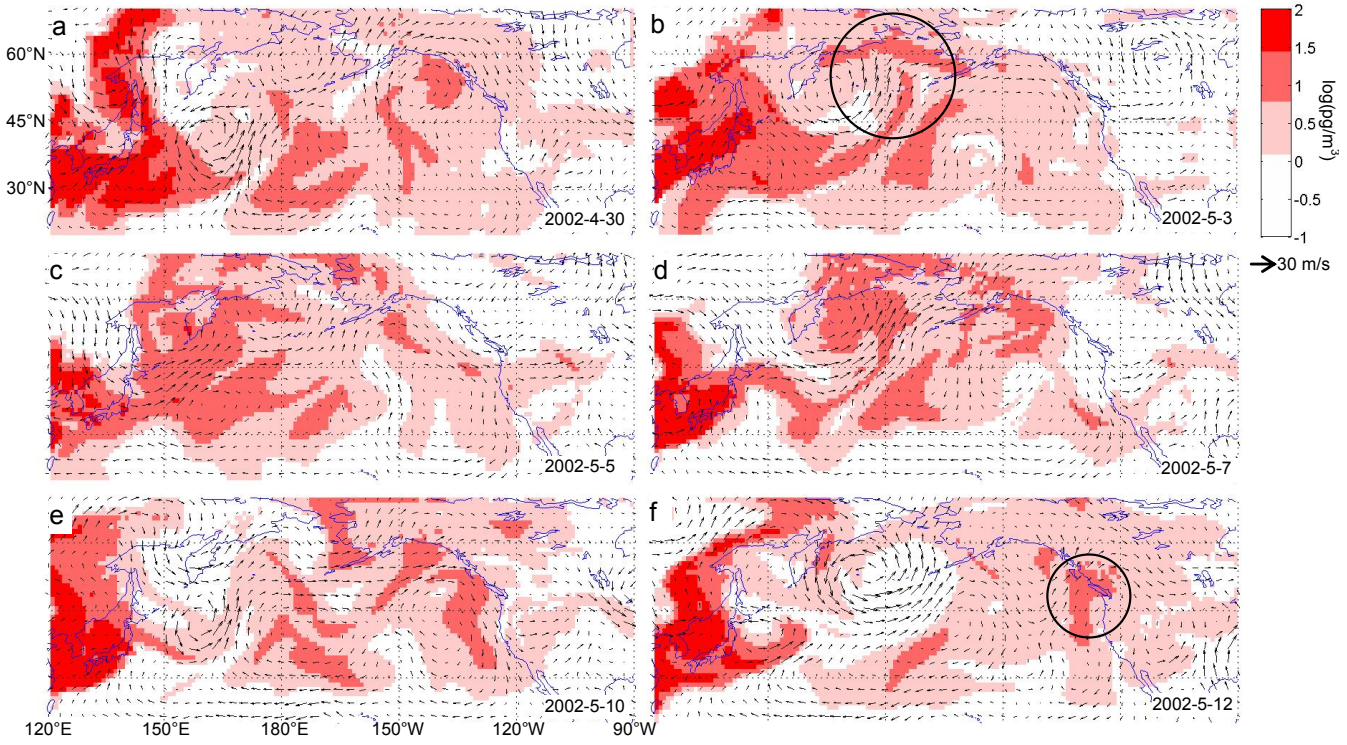

Fig. 6. BaP concentration (background color) and wind field (arrows) at 1200 m during the transport event on 12 May 2002; **(a–f)** show 30 April, 3 May, 5 May, 7 May, 10 May and 12 May, respectively.

by subtracting this running mean from the daily mean concentration to filter out the low frequency variations (a.k.a. High-pass filter, e.g. Liang et al., 2005). Results reveal that the frequencies of daily concentrations at factors of 0.5, 1.0, 1.5, and 2.0 higher than the 11 day running mean concentrations were 9.4 %, 0.72 %, 0.06 % and 0.01 %, respectively. This suggests that although episodic transport can significantly enhance the concentration at a specific site or region for a short time period, its contribution to the long-term mean transpacific transport flux does not appear to be very significant.

3.6 Interannual variation of transport from East Asia

Owing to the significant contribution of trans-Pacific transport of East Asian BaP emissions to North American BaP concentrations (97 %, Fig. 4a), the interannual variability of such a transport merits further investigation. Figure 7a shows the interannual variability of the mean BaP concentrations averaged over North America (brown line) and corresponding total BaP emissions in East Asia (blue bar). As shown, the average BaP concentration over North America has a significant positive correlation with the East Asian emission flux ($r = 0.98$). The North American mean concentration attributable to transpacific transport has increased rapidly since the 1950s and slightly decreased during the last decade due to the declining trend in East Asian emissions. We further defined a transpacific transport index (TPI), calculated by dividing the modeled mean concentration over North America (the black box shown in Fig. 3c) by the Asian emission flux for 1948–2007. Figure 7b illustrates the annual anomaly of the TPI as departures from its mean averaged over 1948–2007 (blue points). The decadal running mean of the TPI was also calculated and shown in this figure (pink points). Despite of the intense annual fluctuations, a decreasing trend in both annual ($p = 0.012$) and average TPI ($p = 8\times10^{-17}$) was modeled, particularly since the 1970s ($p = 6\times10^{-13}$ for annual TPI). Although no measurement extends long enough to directly observe this trend, we hypothesis this TPI trend is associated with changes in interannual or interdecadal atmospheric circulations related to transpacific transport.

Based on extensive studies on the outflow of pollutant tracers from East Asia to the western Pacific Ocean, the East Asian winter monsoon is a key factor influencing the outflow potential and strength (Liu et al., 2003; Zhang et al., 2011). The decreasing trend in TPI is partially caused by the continuous weakening of the East Asian winter monsoon since the late 1980s (Nakamura et al. 2002). Another important reason is the decadal weakening of the Pacific jet, which lengthens the transport time of PAHs over the North Pacific Ocean (Nakamura et al., 2002). Based on a recent review on the effect of global climate change on atmospheric transport potential, the future climate is expected to be more stagnant, with a weaker global atmospheric circulation and

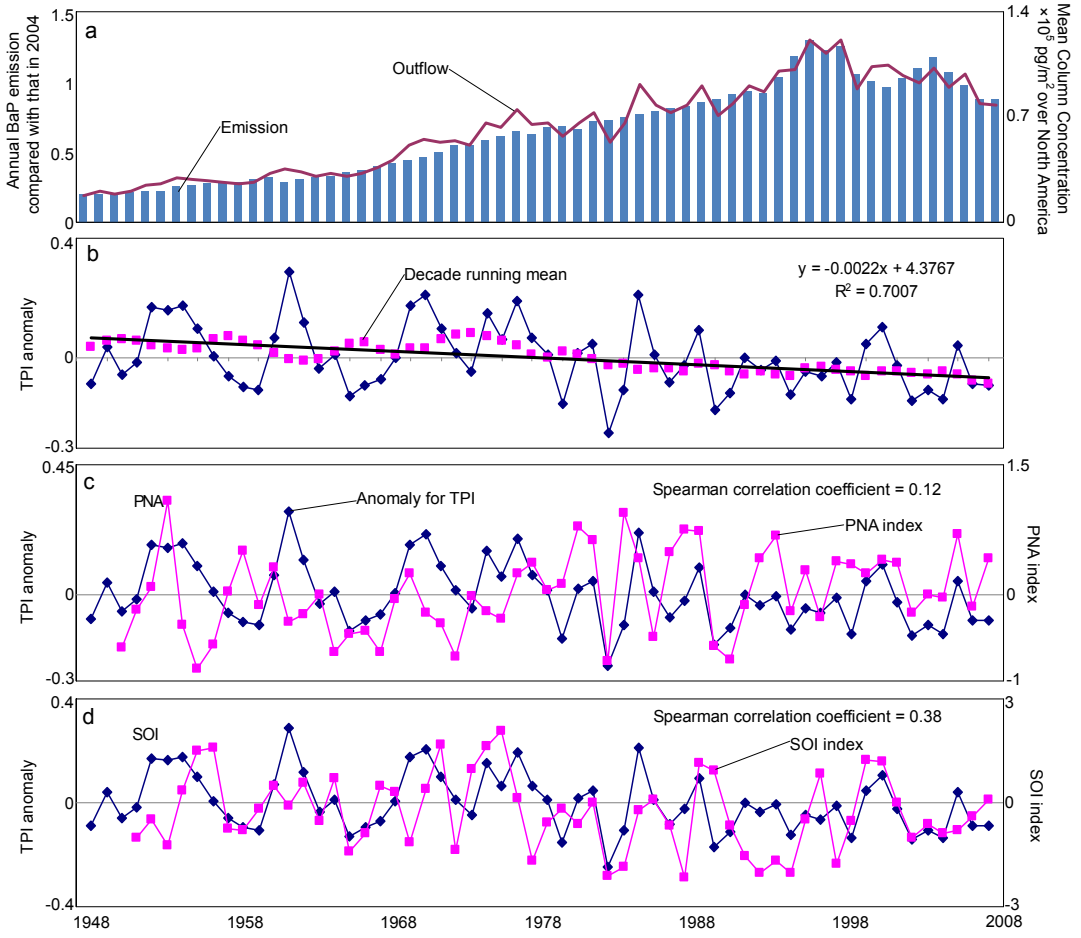

Fig. 7. Interannual variability of transpacific transport of BaP during 1948–2007. **(a)** interannual variation of BaP emission over East Asia and mean BaP column concentration over the region of North America within the boundaries of 30–70° N, 75–125° W, and 0–7 km altitude; **(b)** interannual variation of transpacific transport index (TPI) anomaly, and its decadal running mean; **(c)** the relationship between TPI anomaly and the anomaly of the Pacific North America Index (PNA); **(d)** the relationship between TPI anomaly and the anomaly of the Southern Oscillation Index (SOI).

a decreasing frequency of mid-latitude cyclones (Jacob and Winner, 2009). Therefore, the decreasing trend in TPI would continue in the future. Although the energy consumption and PAH emissions are projected to increase in the next several decades (Zhang et al., 2008b), the decreasing TPI would mitigate this increase.

The Pacific North American index (PNA) and the Southern Oscillation Index (SOI) are indicators for the North-South and East-West atmospheric pressure dipole patterns, respectively, and used to link interannual climate variability with changes in transpacific transport in this study. Lang et al. (2008) found a positive correlation between PNA and transpacific transport for pyrene. Liang et al. (2005) also reported a similar result. In the present study we further examined the relationship between PNA and TPI, but no significant correlation between the two indices was observed (Fig. 7c, Spearman correlation coefficient = 0.12). However, this correlation could be improved (Spearman correlation coefficient = 0.518, $p = 0.009$) if only the recent 14 yr (same as the time period considered by Lang et al., 2008) were taken into account. On the other hand, a significant positive correlation exists between TPI and SOI (Fig. 7d, Spearman correlation coefficient = 0.38, $p = 0.004$). This result contradicts our first intuition because negative (positive) phase SOI is associated with El Niño (La Niña), during which zonal circulation and thus transpacific transport is stronger (weaker) (Wang 2002). However, according to Zhang et al. (1997), positive (negative) SOI was also associated with higher (lower) frequency of cold episodes and intense (weaker) boundary outflow behind fronts. This explains the positive correlation between TPI and SOI found in this study. The positive correlation, instead of negative, between TPI and SOI also implies that the outflow flux of BaP from Asia is a stronger controlling factor for the strength of transpacific transport, compared with the later advection process over the North Pacific.

4 Conclusion

The present modeling study showed strong enhancements of PAH over the region bounded by 70–80° E and 100–120° E, and these PAH were transported eastward by the westerly winds. When the air parcels approached the western coast of North America, the general descending motion carried the PAH-laden air parcels into the lower atmosphere. The transpacific transport flux is ∼1.6 times higher in the winter than in the summer. The strength of East Asian emissions dominates the transpacific transport flux, which contributed 97 % of the modeled average BaP concentrations over North America. The near ground concentration of BaP in North America induced by Asian sources varied between 1–20 pg m^{-3}.

An episodic transport event measured at the CPO site near the west coast of North America on 12 May 2002 reveals the importance of BaP enhancement due to transpacific atmospheric transport, facilitated by the WCB and low pressure systems. The frequencies of daily BaP fluxes from transpacific transport that were a factor of 0.5, 1.0, 1.5, and 2.0 larger than the 11 day running mean were 9.4 %, 0.72 %, 0.06 % and 0.01 %, respectively, implying a mild contribution of episodic transport to the long-term mean transport flux. Significant interannual fluctuations of transpacific transport of PAHs were found. A general decreasing trend was modeled during 1948–2007, especially after the 1970s. The transpacific transport was found to be positively correlated to the Southern Oscillation Index, likely due to the higher frequency of cold episodes and intense boundary outflow within the positive phase of the SOI.

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