

Influence of monsoon system on α -HCH fate in Asia: A model study from 1948 to 2008

Yue Xu,^{1,2} Chongguo Tian,¹ Gan Zhang,² Lili Ming,² Yan Wang,² Yingjun Chen,¹ Jianhui Tang,¹ Jun Li,² and Chunling Luo²

Received 26 November 2012; revised 20 March 2013; accepted 23 March 2013.

[1] The Chinese Gridded Pesticide Emission and Residue Model was applied to simulate long-term environmental fate of α -hexachlorocyclohexane (HCH) in Asia spanning 1948 to 2008. The simulation identifies the southern Tibetan Plateau and the margin of the Asian monsoon regime from the northeast area of the Tibetan Plateau to the Northeast China as sink areas. This spatial pattern of sink distribution is closely related to the summer monsoon activities in Asia. Wet deposition associated with the summer monsoon (June to August) is responsible for at least half of the α -HCH mass entering into the sink areas. After deposition, relatively low temperature and high soil organic matter in the sink areas are favorable for α -HCH detention. Chinese and Indian sources are the major contributors to α -HCH contamination over the monsoon margin and the Tibetan Plateau, driven by the East Asian summer monsoon and South Asian summer monsoon, respectively.

Citation: Xu, Y., C. Tian, G. Zhang, L. Ming, Y. Wang, Y. Chen, J. Tang, J. Li, and C. Luo (2013), Influence of monsoon system on α -HCH fate in Asia: A model study from 1948 to 2008, *J. Geophys. Res. Atmos.*, 118, doi:10.1002/jgrd.50368.

1. Introduction

[2] The fate of persistent organic pollutants (POPs) is a global concern. Many studies combined knowledge of source emission rates with the quantification of environmental reservoirs and sink fluxes to illustrate their environmental transport behaviors [Lohmann *et al.*, 2007]. The migration of POPs from source to sink largely depends on atmospheric transport and deposition, which is sensitive to temperature, precipitation, wind field, oceanic current, and land cover characteristics [Ma and Cao, 2010; Lamon *et al.*, 2009]. The climate in Asia is characterized by South and East Asian monsoon, the largest seasonal abnormality of the global climate system accompanied by huge water vapor transport and latent heat release [Ueda *et al.*, 2003]. This monsoon system could not only affect volatilization, degradation, wet deposition, and runoff processes of POPs but also provide an effective pathway to deliver airborne pollutants [Tian *et al.*, 2009b].

[3] Knowledge in the source-sink relationship of POPs in Asia and its association with the Asian monsoon is of critical importance to understand their regional dynamics and global budget. Countries in this region were large producers and consumers of some POPs, such as technical HCH (hexachlorocyclohexane) [Li, 1999a; Li and Macdonald, 2005]. It has been estimated that the historical usage in China and India was 4464 and 1057 kt, ranking as the top two countries with the highest technical HCH use in the globe [Li, 1999a; Li and Macdonald, 2005]. China prohibited technical HCH usage in 1983, whereas India and other Asian countries restricted or banned the substance since the 1990s [Li and Macdonald, 2005]. Although the legal use of technical HCH has been discontinued for a long period of time in these countries and regions, the environmental residues of HCH isomers are still high [Liu *et al.*, 2009; Zhang *et al.*, 2008]. Technical HCH contains mainly five isomers: α -HCH (53–70%), β -HCH (3–14%), γ -HCH (11–18%), δ -HCH (6–10%), and ϵ -HCH (3–5%). Due to the adverse effects on human health and ecosystem, three of them (α -HCH, β -HCH, and γ -HCH) were added to Annex A of the Stockholm Convention on POPs with no exemptions for ongoing production in 2009. Among the five isomers, α -HCH is commonly selected as a representative to investigate the environmental fate of POPs due to its strong long-range atmospheric transport potential and well-documented measurement data. A number of monitoring campaigns have demonstrated that α -HCH in Asia can be transported to the Arctic and North America via atmospheric movement [Bailey *et al.*, 2000; Primbs *et al.*, 2008; Xiao *et al.*, 2010], and recent studies have linked its atmospheric transport and deposition to seasonal monsoon systems [Kang *et al.*, 2009; Tian *et al.*, 2009b].

[4] Soil is a more suitable environmental matrix to explore the long-term environmental behavior of a persistent chemical

Additional supporting information may be found in the online version of this article.

¹Key Laboratory of Coastal Zone Environmental Processes and Ecological Remediation and Shandong Provincial Key Laboratory of Coastal Zone Environmental Processes, Yantai Institute of Coastal Zone Research (YIC), Chinese Academy of Sciences (CAS), Yantai, China.

²State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, China.

Corresponding author: C. Tian, Key Laboratory of Coastal Zone Environmental Processes and Ecological Remediation and Shandong Provincial Key Laboratory of Coastal Zone Environmental Processes, Yantai Institute of Coastal Zone Research (YIC), Chinese Academy of Sciences (CAS), 17 Chunhui Road, Laishan District, Yantai, Shandong 264003, China. (cgtian@yic.ac.cn)

©2013. American Geophysical Union. All Rights Reserved.
2169-897X/13/10.1002/jgrd.50368

because it is a nonfloating (or low floating) and principal storage medium for POPs [Wöhrenschiimmel *et al.*, 2012]. With the help of a well-established multimedia model, the environmental fate of POPs in the soil compartment can be well explored [Gong *et al.*, 2007; Wöhrenschiimmel *et al.*, 2012]. Due to historical usage, Asia was previously considered as a source rather than a sink [Wania and Mackay, 1999; Toose *et al.*, 2004]. Little work has addressed the influence of monsoon [Tian *et al.*, 2011]. In the present study, the Chinese Gridded Pesticide Emission and Residue Model (ChnGPERM) was applied to (i) identify the potential source and sink regions of α -HCH in Asian soils, (ii) explore the causes of sink formation, and (iii) assess the association between sink formation subjected to China and India sources and monsoon system.

2. Materials and Methods

2.1. Model and Model Input

[5] The ChnGPERM applied in this study is a gridded mass balance model developed to simulate transport and transfer of pesticides in Asia. Details of this model and the input data have been described elsewhere [Tian *et al.*, 2009a; Tian *et al.*, 2011]. The soil compartment is categorized into four types: cropland, forestry, grassland, and uncultured land.

[6] The model domain spans from 0°N to 60°N and from 50°E to 150°E with a horizontal resolution of $\frac{1^\circ}{4} \times \frac{1^\circ}{4}$ latitude/longitude. Based on previous work [Li, 1999b], the α -HCH inventory from 1948 to 1999 with the same horizontal resolution has been developed [Tian *et al.*, 2011]. Accumulated technical HCH usage until 1999 in the model domain is presented in Figure S1 in the auxiliary material. As Figure S1 illustrates, large amounts of technical HCH were applied mainly over southeast China and India. China was the major contributor of total HCH usage before the 1980s. From the 1980s to 1990s, Indian usage became significant (see Figure S2 in the auxiliary material). Table S1 in the auxiliary material lists the physicochemical properties of α -HCH used in the modeling investigation.

[7] The linearity of the governing equations of the model enables us to quantify the relative contribution of different source regions of α -HCH to its burden in receptors. To do so, numerical simulations have been performed for three model scenarios accounting for different sources as adopted in a previous study [Tian *et al.*, 2011]. These three scenarios were (1) all the sources over the model domain (scenario 1), (2) China sources only (scenario 2), and (3) India sources only (scenario 3).

[8] Since mass exchange of α -HCH between different model grid cells was only accomplished by atmospheric transport and deposition in the model, the net loading due to atmospheric movement can reflect geographical source-sink distribution of the pesticide. The mass conservation of the model (see the next subsection) can be used to distinguish source and sink regions of α -HCH in the model domain [Tian *et al.*, 2011]. The mass balance from 1948 to the n th year within each grid cell (i) can be written as

$$\sum_{1948}^n \text{flow}_i = \sum_{1948}^n U_i - \text{Re}_i^n - \sum_{1948}^n D_i - \sum_{1948}^n L_i \quad (1)$$

[9] $\sum_{1948}^n \text{flow}_i$ is the net mass flow due to atmospheric input

and output; Re_i^n , $\sum_{1948}^n U_i$, $\sum_{1948}^n D_i$, and $\sum_{1948}^n L_i$ are the mass of

α -HCH due to soil residue, the accumulated usage, and the losses by degradation and leaching in the soil within each grid, respectively. Positive $\sum_{1948}^n \text{flow}_i$ represents the

net input, suggesting the net inflow to the grid cell due to atmospheric transport and deposition, and negative $\sum_{1948}^n \text{flow}_i$ indicates the net outflow from the grid cell.

[10] A fugacity method developed by Harner *et al.* [Harner *et al.*, 2001; Tian *et al.*, 2012] was used to assess the response of α -HCH degradation, deposition, and reemission to environmental factors.

2.2. Model Evaluation

[11] Figure S3 in the auxiliary material shows the overall mass balance of the pesticide over the model domain for the entire 61 year modeling integration. The total mass of usage during 1948–1999, the residues in 2008, and the accumulated degradation, soil leaching, and air advection from 1948 to 2008 were 4.06×10^9 , 1.73×10^7 , 3.76×10^9 , 1.12×10^7 , and 1.87×10^8 kg, respectively. The latter four terms were summed and compared with the total usage to assess the mass conservation of the model. The relative error of the comparison is $\sim 2\%$ (see Figure S3). This indicates that the model achieved well the mass conservation.

[12] The model performance was also evaluated by comparing the observed soil concentrations to simulated values and a peat core record to modeled deposition fluxes derived from model scenario 1. Historical soil concentrations across the model domain from 1979 to 2008 collected from literatures and corresponding modeled values were listed in Table S2 in the auxiliary material. Surface soil (0–10 cm) samples were generally taken around small areas. Therefore, the monitored concentrations were obtained from arithmetical mean values in soil samples, and the modeled concentrations used soil concentrations in the third soil layer (from 1.1 to 21.1 cm) [Tian *et al.*, 2011]. The simulated concentrations are generally comparable to those monitored values (Figure S4 in the auxiliary material). The Pearson correlation coefficient was 0.81 ($p < 0.01$) for the paired data set, suggesting that the mean soil concentration distribution pattern is well captured by the model. In addition, the monitored to modeled mean values in Table S2 ranged from 0.02 to 9.6, with an average of 1.4. It indicated that the two sets of data were mostly in the same order of magnitude.

[13] A monitoring campaign has been conducted to reconstruct the historical records of α -HCH in a peat core from Zoigê-Hongyuan, Tibetan Plateau, China (32°46.774'N, 102°30.964'E; 3510 m above sea level), a remote site located on the eastern edge of the Tibetan Plateau. Due to high organic carbon content, the peat core achieved long-term settling of α -HCH. Since peat cores represent an almost ideal medium for recording temporal changes in organic contaminant deposition rates [Rapaport and Eisenreich, 1988], the modeled deposition flux was compared with peat core recorded data. After deducting α -HCH volatilization from the soil, the modeled annual atmospheric depositions at a grid most adjacent to the sampling site were compared with the peat core observations (Figure S5 in the auxiliary material). Overall, the modeled depositions to a large extent

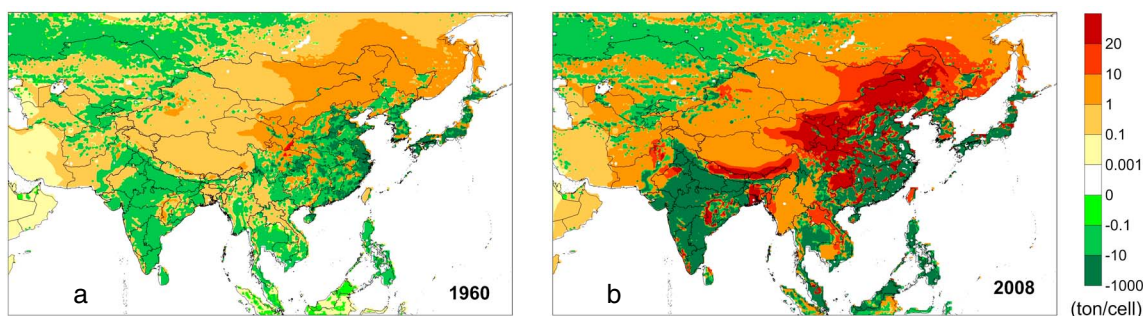


Figure 1. The net mass flow (unit: ton/cell) according to the mass balance at each grid until (a) 1960 and (b) 2008.

reproduce the temporal trend of peat core recorded α -HCH (Pearson correlation coefficient $r=0.80$; $p < 0.01$), while in some cases (from the late 1960s to the 1970s), the modeled values show a lag to the concentration recorded in the peat core. High concentrations in the peat core occurred earlier than the corresponding elevated modeled values. It was possibly due to the downward leaching process.

2.3. Monsoon Index and Monsoon Margin

[14] To quantify the seasonal variation of East and South Asia monsoon, a unified dynamical index of monsoon (MI) was calculated following [Li and Zeng, 2002]

$$\delta = \frac{\|\bar{V}_1 - V_i\|}{\|\bar{V}\|} - 2 \quad (2)$$

where \bar{V}_1 and V_i are the January climatological and monthly wind vectors at a grid and \bar{V} is the mean of the January and July climatological wind vectors at the same grid. The norm

$\|A\|$ is defined as $\|A\| = \left(\int \int_S |A|^2 dS \right)^{\frac{1}{2}}$, where S denotes

the domain of integration. In the model domain, $\delta > 0$ represents summer monsoon. The monthly average monsoon index in the monsoon sectors of East Asia (112.5–125°E, 25–42.5°N) and South Asia (72.5–90°E, 8–27.5°N) was calculated to represent East and South Asia monsoon, respectively.

[15] Monsoon activities also exhibit annual and spatial variations. To outline the monsoon-dominated areas, the concept of monsoon margin was introduced in many studies [Li et al., 2012; Zhao and Yu, 2012]. Monsoonal margin is a broad transitional zone representing the most northern areas where a monsoon regime can extend to. Shi [1996] defined the monsoon margin as an area with annual precipitation that ranged from 200 mm to 450 mm. Here we further constrained the areas in a temperate monsoon region (the rainfall from June to August exceeds 50% of the annual precipitation), in order to exclude some areas with temperate continental climate. Due to the annual variation of the monsoon margin, the probability of the margin at each grid can be presented by the frequency of the margin that occurred at a grid during the modeling period (Figure S6 in the auxiliary material).

3. Results and Discussions

3.1. Soil Concentrations and Sink Distribution

[16] Figure S7 in the auxiliary material presents the annual average of modeled soil concentrations of α -HCH in 1960,

1983, 1996, and 2008, respectively. During the application period of technical HCH, α -HCH soil residues were higher in southeast China and other source regions (see Figure S1) than in those nonsource areas over the model domain, indicating that fresh emission of α -HCH dominated the spatial and temporal pattern of soil residues (see the soil residues in 1960 and 1983 shown in Figure S7). After the ban of technical HCH in the model domain, the spatial distribution of α -HCH soil residues changed substantially. The soil burden in some nonsource areas was comparable with, or even higher than, that in source regions where a large amount of technical HCH was used.

[17] In order to identify the sink distribution, the accumulated net mass of α -HCH among different model grid cells due to atmospheric input and output from 1948 to 1960

$\left(\sum_{1948}^{1960} \text{flow}_i \right)$ and to 2008 $\left(\sum_{1948}^{2008} \text{flow}_i \right)$ was calculated using

equation (1). The results are illustrated in Figure 1. Figure 1a shows that the region extending from Northeast China (referred to as the northern sink) was the major sink when the usage was low in India (see Figure S2) before 1960. Although the southern areas in the model domain were generally characterized as sources, the Tibetan Plateau was not marked as a sink area before 1960 because of the low use of the substance in India (see Figure S2). After the 1970s, the source/sink distribution pattern remained almost the same as the pattern in 2008 (see Figure 1b), in which the southern Tibetan Plateau became another sink region in the plateau (the red areas in Figure 1b). As the atmosphere is a major pathway to transport α -HCH in the model, the development of a sink region indicates favorable transport pathways from source regions to the sink through reemission and deposition as well as other environmental factors influencing the fate of α -HCH in the sink regions.

3.2. Monsoon-Associated Sink Formation

3.2.1. General Comments

[18] Sink formation has been linked to the deposition, volatilization, and degradation processes, which are commonly determined by environmental factors [Lohmann et al., 2007] such as wind, precipitation, temperature, and soil organic carbon (SOC). The connection between each process and environmental factor has been documented (Figure S8 in the auxiliary material). Wet deposition of α -HCH was generally enhanced by monsoon precipitation

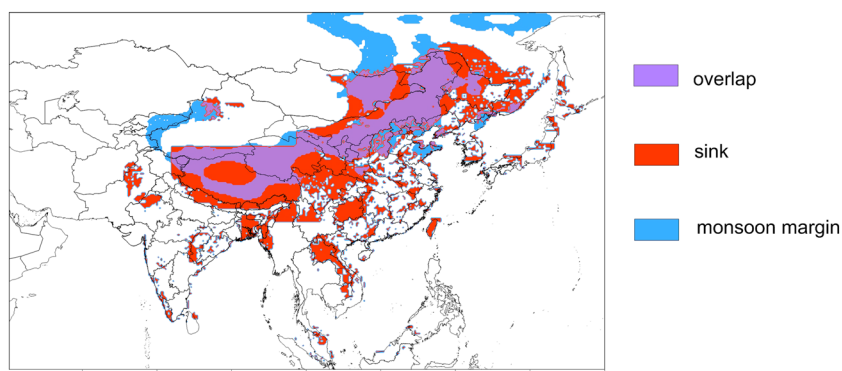


Figure 2. The overlap areas between the monsoon margin and sink areas.

[Ramesh *et al.*, 1989] and increased significantly at lower temperature [Wania and Haugen, 1999]. On the other hand, volatilization can be reduced with low temperature [Bethan *et al.*, 2001] and high SOC [Nam *et al.*, 2008]. According to the Arrhenius equation, slower degradation will take place at lower temperatures. Having those taken into account, we assumed that a sink could be formed in those areas where deposition is high and emission and degradation are low. That is, a sink can be formed under relatively higher precipitation and SOC and lower temperatures.

[19] Changes in wind field, precipitation, temperature, and SOC (via long-term response to changes in vegetation [Fang *et al.*, 1999]) in the model domain have been found to be all related to Asian monsoon. Those regions beyond the monsoon region are under relatively cold and dry climate conditions where atmospheric transport is weak during the monsoon season [Sato, 2009]. The monsoon region is characterized by heavy seasonal precipitation and strong thermal atmospheric circulation. The latter leads to higher temperature and forms an effective northward atmospheric transport pathway [Tian *et al.*, 2011]. As noted before, high rainfall and low temperature are favorable for sink formation. Therefore, between monsoon and nonmonsoon regions, there likely exists an area where major environmental factors may favor sink formation. We found that there was an ~60% overlap between the monsoon margin and sink areas (Figure 2). The sink region was resided in the southern side of the monsoon margin, suggesting the sink formation processes could be associated with those environmental factors within the monsoon areas. Large discrepancy only occurred in the Tibetan Plateau, where a large slope of up to 5000 m in the altitude (the Greater Himalayas) could result in more strong deposition of POPs from the atmosphere to the surface by mountain cold trapping effect [Wania and Westgate, 2008].

3.2.2. Precipitation

[20] Our results have shown that the modeled soil residues of α -HCH correlated well with (Pearson correlation coefficient: 0.6–0.7) its accumulated deposition amount. In this study, the modeled wet and dry deposition fluxes are illustrated in Figure S9 in the auxiliary material. Wet deposition contributed to a significant part of the net mass input of α -HCH. Precipitation mainly influences the wet deposition process. If the monthly wet deposition fluxes were standardized by their annual average, the correlation coefficient

between standardized wet deposition and monthly precipitation was generally >0.6 over the monsoon region (Figure S10 in the auxiliary material). Temporally, accumulated dry and wet depositions of α -HCH from June to August accounted for 47.3% and 52.2% of the annual total deposition fluxes in the sink areas, respectively, although the summer monsoon only extends for several months. This implies that the summer monsoon season is an important period for the annual deposition. Geographically, atmospheric α -HCH mainly deposited to monsoon regions (Figure S9) where annual rainfall >400 mm (Figure S11 in the auxiliary material). Before the prohibition in the 1980s, the atmospheric α -HCH deposited primarily to the source areas in the model domain, the Northeast China, and the Tibetan Plateau. After the 1980s, the areas with high atmospheric deposition were still located within the monsoon Asia, especially 105°E (the orange areas in Figure S9, 2000, right panel) which delineates the South and East Asian monsoon regions [Wang *et al.*, 2003]. As a result, the summer monsoon and correspondent monsoon precipitation might be considered as the most effective and important factors contributing to the α -HCH input into sink/receptor regions which, as shown in Figure 1, superimposed with the monsoon margin.

3.2.3. Temperature

[21] Temperature influences degradation, deposition, and reemission processes [Ma and Cao, 2010]. Compared with that in other regions in the same latitude over the model domain, the average temperature in sink areas was relatively low (Figure S12 in the auxiliary material). Within the model domain, the average temperature was 7°C higher in source areas and 16°C and 1.4°C lower in the Tibet and northern sink areas than in other regions in the same latitude (Figure S13 in the auxiliary material); we then expect the stronger volatilization of α -HCH from surface soil and degradation in source regions and weaker volatilization and degradation in sink areas. According to the fugacity model (section 2), temperature mainly influences the surface exchange (Figure 3). Compared with those in other regions in the same latitude, the atmospheric degradation rate, deposition, and reemission fluxes in source regions were 9%, 38%, and 95% higher, respectively. Although, as aforementioned, wet deposition in source areas was higher, the significant increase of reemission and faster degradation due to higher temperature in source areas may buffer the mass input and cause a net output of α -HCH.

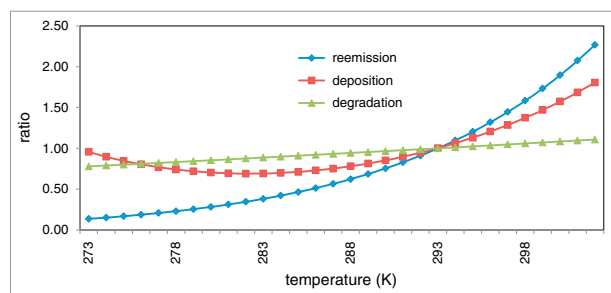


Figure 3. Ratio of α -HCH reemission from soil, atmospheric deposition, and degradation at different temperatures to that at 293 K.

On the other hand, it was found that the relatively low temperature in the Tibet resulted in 65% higher deposition, 81% lower reemission, and 19% slower degradation than that in other areas at the same latitudes, thereby leading to the sink formation in the Tibet. In the northern sink region, decreasing reemission (14%), slowing degradation (2%), and increasing deposition (2%) were also favorable for HCH detention.

3.2.4. SOC

[22] SOC and monsoon are not linked directly. However, soil organic matter responds to vegetative change that has been linked to summer monsoon enhancements [Fang *et al.*, 1999]. Spatially, high SOC content also occurs along the summer monsoon margin, such as the southeast Tibet [Tian *et al.*, 2008] and Northeast China [Wang *et al.*, 2002] (see Figure S14 in the auxiliary material). The elevated SOC resulted in 40% decrease of α -HCH reemission in the Tibetan Plateau, ~5% decline in the northern sink areas, and 7% increase in the source areas, in comparison with that in other areas in the same latitude. In the model domain, spatial changes in environmental factors that drive the environmental fate of α -HCH led to significant accumulation of α -HCH within the monsoon margin.

3.3. Impact of East and South Asia Monsoon on α -HCH Sink

[23] Asian monsoon can be divided into East and South Asia monsoon systems: The former leads to northward outflow of α -HCH from a Chinese source, and the latter results in eastward extension from an Indian source (Figure S15 in the auxiliary material). It would be interesting to know the association of the formation of the sink area with the two monsoon systems. Given the intimate connection between the two monsoon systems, the association of atmospheric transport of α -HCH with individual source in both monsoon systems is not straightforward to be understood from bivariate correlation analysis. Partial correlation is a method to assess covariation of a pair of random variables from a collection of random variables in the case where the influence of the remaining variables is eliminated. For three variables, the partial correlation between two variables (X and Y) holding the third variables (Z) fixed is the correlation between the residuals X and Y resulting from the linear regression of X with Z and of Y with Z , respectively. Therefore, the partial correlation coefficient between total wet deposition in sinks and MI in each monsoon region was computed, in order to exclude influence from another monsoon sector. When a

meridional line (the blue line in Figure S16 in the auxiliary material) moves from 80°E to 125°E with an interval of every 5°, the monthly wet deposition within the western sink region (the red areas located on the west of the blue line) was summarized. The partial correlation coefficients between the summarized deposition and East and South Asia MI were calculated and are plotted in Figure 4, respectively. Western sink areas correlated more strongly with South Asia MI (red line) than East Asia MI (blue line). The decline of the correlation coefficients (red) in Figure 4 occurred at 105°E, the longitude that has been considered to be the border between East and South Asia monsoons [Wang *et al.*, 2003]. On the east of 105°E, the impact of South Asia monsoon on the α -HCH wet deposition gradually diminishes. However, there appeared no stronger association between α -HCH wet deposition and East Asia monsoon on the east of this border (namely, 105°E) as we expected. The correlations (the blue line in Figure 4) exhibit a largest value at 105°E and decrease thereafter. This is because, when the blue line moves from 90°E to 110°E, the western areas also included the α -HCH sink in Qinghai province, with the sink still attributable to eastern China sources and East Asia monsoon (Figure S17 in the auxiliary material).

[24] The analysis can be further supported by evaluating the relative contribution of Chinese and Indian sources to the sink formation of α -HCH. Since both eastern China and India are major source regions under Asian monsoon wind regimes, normalized soil concentrations can be used to eliminate the influence of the strength of α -HCH usage. Figure S17 shows normalized soil concentrations from Chinese sources and Indian sources, respectively. Chinese sources lead to accumulation of α -HCH in the Northeast China and Qinghai province, while Indian sources result in high concentration in the southern Tibet and the part of southwest China. This indicates that the Tibetan sink could be linked to α -HCH outflow from India and the northern sink could be connected to outflow from eastern China.

4. Conclusion

[25] The environmental fate of α -HCH from 1948 to 2008 in South and East Asia was simulated by ChnGPERM. The

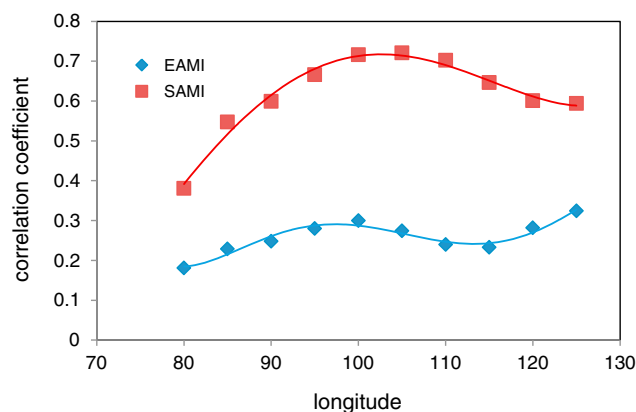


Figure 4. The change of correlation coefficient between the total wet deposition within the sink areas at the left side of a predefined meridional line in Figure S16 and the East and South Asian monsoon index when the line moves from 80°E to 125°E.

sink distribution generally resembles the Asian monsoon margin. According to the mass balance of the model, the sink formation in the model domain can be attributed to the increasing input and decreasing output of α -HCH in a certain region of the model domain. Both the increased atmospheric input and the persistence of the substance along the monsoon margin may contribute to the model simulated sink distribution. Significant amount of α -HCH through wet deposition in the summer monsoon season increased atmospheric input, whereas relatively high SOC and low temperature favored accumulation of α -HCH over the sink areas. Since the major areas of pesticides application were located in monsoon zones, the association between sink distribution and monsoon investigated in the present study postulates that the monsoon system is an important factor influencing the geographical distribution and transport of airborne pollutants, which should merit attention and further investigation. Geographically, the environmental fate of α -HCH in the eastern Tibetan Plateau was influenced by the atmospheric outflow from Chinese sources, whereas Indian sources played a major role for its accumulation in the southern Tibetan Plateau.

[26] **Acknowledgments.** This work was supported by the Knowledge Innovation Program of the Chinese Academy of Sciences (KZCX2-YW-GJ02) and the Natural Science Foundation of China (41101495 and 41125014). The authors gratefully acknowledge Global Soil Data Task and the National Centers for Environmental Prediction for providing the environmental data.

References

- Bailey, R., L. A. Barrie, C. J. Halsall, P. Fellin, and D. C. G. Muir (2000), Atmospheric organochlorine pesticides in the western Canadian Arctic: Evidence of transpacific transport, *J. Geophys. Res.-Atmos.*, *105*, 11805–11811, doi: 10.1029/1999JD901180.
- Bethan, B., W. Dannecker, H. Gerwig, H. Hühnerfuss, and M. Schulz (2001), Seasonal dependence of the chiral composition of α -HCH in coastal deposition at the North Sea, *Chemosphere*, *44*, 591–597, doi:10.1016/S0045-6535(00)00495-1.
- Fang, X.-M., Y. Ono, H. Fukusawa, P. Bao-Tian, J.-J. Li, G. Dong-Hong, K. Oi, S. Tsukamoto, M. Torii, and T. Mishima (1999), Asian summer monsoon instability during the past 60,000 years: Magnetic susceptibility and pedogenic evidence from the western Chinese Loess Plateau, *Earth Planet. Sci. Lett.*, *168*, 219–232, doi: 10.1016/S0012-821X(99)00053-9.
- Gong, S. L., P. Huang, T. L. Zhao, L. Sahsuvar, L. A. Barrie, J. W. Kaminski, Y. F. Li, and T. Niu (2007), GEM/POPs: A global 3-D dynamic model for semi-volatile persistent organic pollutants—Part 1: Model description and evaluations of air concentrations, *Atmos. Chem. Phys.*, *7*, 4001–4013, doi: 10.5194/acp-7-4001-2007.
- Harner, T., T. F. Bidleman, L. M. M. Jantunen, and D. Mackay (2001), Soil-air exchange model of persistent pesticides in the United States Cotton Belt, *Environ. Toxicol. Chem.*, *20*, 1612–1621, doi: 10.1002/etc.5620200728.
- Kang, J. H., S. D. Choi, H. Park, S. Y. Baek, S. Hong, and Y. S. Chang (2009), Atmospheric deposition of persistent organic pollutants to the East Rongbuk Glacier in the Himalayas, *Sci. Total Environ.*, *408*, 57–63, doi: 10.1016/j.scitotenv.2009.09.015.
- Lamon, L., M. Dalla Valle, A. Critto, and A. Marcomini (2009), Introducing an integrated climate change perspective in POPs modelling, monitoring and regulation, *Environ. Pollut.*, *157*, 1971–1980, doi: 10.1016/j.envpol.2009.02.016.
- Li, J. P., and Q. C. Zeng, (2002), A unified monsoon index, *Geophys. Res. Lett.*, *29*, 1274, doi: 10.1029/2001GL013874,2002.
- Li, Y. F. (1999a), Global technical hexachlorocyclohexane usage and its contamination consequences in the environment: From 1948 to 1997, *Sci. Total Environ.*, *232*, 12–158, doi: 10.1016/S0048-9697(99)00114-X.
- Li, Y. F. (1999b), Global gridded technical hexachlorocyclohexane usage inventories using a global cropland as a surrogate, *J. Geophys. Res.-Atmos.*, *104*, 23785–23797, doi: 10.1029/1999JD900448.
- Li, Y. F., and R. W. Macdonald (2005), Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: A review, *Sci. Total Environ.*, *342*, 87–106, doi:10.1016/j.scitotenv.2004.12.027.
- Li, Y., N. a. Wang, H. Chen, Z. Li, X. Zhou, and C. Zhang (2012), Tracking millennial-scale climate change by analysis of the modern summer precipitation in the marginal regions of the Asian monsoon, *J. Asian Earth Sci.*, *58*(0), 78–87, doi: 10.1016/j.jseas.2012.07.001.
- Liu, X., G. Zhang, J. Li, L. Yu, Y. Xu, X. Li, Y. Kobara, and K. C. Jones (2009), Seasonal patterns and current sources of DDTs, chlordanes, hexachlorobenzene, and endosulfan in the atmosphere of 37 Chinese cities, *Environ. Sci. Technol.*, *43*, 1316–1321, doi: 10.1021/es802371n.
- Lohmann, R., K. Breivik, J. Dachs, and D. Muir (2007), Global fate of POPs: Current and future research directions, *Environ. Pollut.*, *150*, 150–165, doi: 10.1016/j.envpol.2007.06.051.
- Ma, J., and Z. Cao (2010), Quantifying the perturbations of persistent organic pollutants induced by climate change, *Environ. Sci. Technol.*, *44*, 8567–8573, doi: 10.1021/es101771g.
- Nam, J. J., O. Gustafsson, P. Kurt-Karakus, K. Breivik, E. Steinnes, and K. C. Jones (2008), Relationships between organic matter, black carbon and persistent organic pollutants in European background soils: Implications for sources and environmental fate, *Environ. Pollut.*, *156*, 809–817, doi:10.1016/j.envpol.2008.05.027.
- Primbs, T., A. Piekarz, G. Wilson, D. Schmedding, C. Higginbotham, J. Field, and S. M. Simonich (2008), Influence of Asian and Western United States urban areas and fires on the atmospheric transport of polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and fluorotelomer alcohols in the Western United States, *Environ. Sci. Technol.*, *42*, 6385–6391, doi: 10.1021/es702160d.
- Ramesh, A., S. Tanabe, R. Tatsukawa, A. N. Subramanian, S. Palanichamy, D. Mohan, and V. K. Venugopalan (1989), Seasonal variations of organochlorine insecticide residues in air from Porto Novo, *South India. Environ. Pollut.*, *62*, 213–222, doi: 10.1016/0269-7491(89)90188-7.
- Rapaport, R. A., and S. J. Eisenreich (1988), Historical atmospheric inputs of high molecular-weight chlorinated hydrocarbons to eastern North-America, *Environ. Sci. Technol.*, *22*(8), 931–941, doi: 10.1021/Es00173a011.
- Sato, T. (2009), Influences of subtropical jet and Tibetan Plateau on precipitation pattern in Asia: Insights from regional climate modeling, *Quat. Int.*, *192*, 148–158, doi: 10.1016/j.quaint.2008.07.008.
- Shi, Z. T. (1996), Regional characters of natural disaster in marginal monsoon belt of China, *J. Arid Land Resour. Environ.*, *10*, 1–7 (in Chinese).
- Tian, C. G., Y. F. Li, H. L. Jia, H. L. Wu, and J. M. Ma (2009a), Modelling historical budget of alpha-hexachlorocyclohexane in Taihu Lake, China, *Chemosphere*, *77*, 459–464, doi: 10.1016/j.chemosphere.2009.07.061.
- Tian, C. G., L. Y. Liu, J. M. Ma, J. H. Tang, and Y. F. Li (2011), Modeling redistribution of alpha-HCH in Chinese soil induced by environment factors, *Environ. Pollut.*, *159*, 2961–2967, doi: 10.1016/j.envpol.2011.04.031.
- Tian, C. G., J. M. Ma, L. Y. Liu, H. L. Jia, D. D. Xu, and Y. F. Li (2009b), A modeling assessment of association between East Asian summer monsoon and fate/outflow of alpha-HCH in Northeast Asia, *Atmos. Environ.*, *43*, 3891–3901, doi: 10.1016/j.atmosenv.2009.04.056.
- Tian, C. G., J. M. Ma, Y. J. Chen, L. Y. Liu, W. L. Ma, and Y. F. Li (2012), Assessing and forecasting atmospheric outflow of α -HCH from China on intra-, inter-, and decadal time scales, *Environ. Sci. Technol.*, *46*, 2220–2227, doi: 10.1021/es202851n.
- Tian, Y. Q., H. Ouyang, X. L. Xu, M. H. Song, and C. P. Zhou (2008), Distribution characteristics of soil organic carbon storage and density on the Qinghai-Tibet Plateau (in Chinese), *Acta Pedologica Sin.*, *45*, 933–942.
- Toose, L., D. G. Woodfine, M. MacLeod, D. Mackay, and J. Gouin (2004), BETR-World: A geographically explicit model of chemical fate: Application to transport of α -HCH to the Arctic, *Environ. Pollut.*, *128* (1–2), 223–240, doi: 10.1016/j.envpol.2003.08.037.
- Ueda, H., H. Kamahori, and N. Yamazaki (2003), Seasonal contrasting features of heat and moisture budgets between the eastern and western Tibetan Plateau during the GAME IOP, *J. Climate*, *16*, 2309–2324, doi: 10.1175/2757.1.
- Wöhmschimmel, H., P. Tay, H. von Waldow, H. Hung, Y.-F. Li, M. MacLeod, and K. Hungerbühler (2012), Comparative assessment of the global fate of α - and β -hexachlorocyclohexane before and after phase-out, *Environ. Sci. Technol.*, *46*, 2047–2054, doi: 10.1021/es203109q.
- Wang, B., S. C. Clemens, and P. Liu (2003), Contrasting the Indian and East Asian monsoons: Implications on geologic timescales, *Mar. Geol.*, *201*, 5–21, doi: 10.1016/S0025-3227(03)00196-8.
- Wang, S. P., G. S. Zhou, Y. C. Lv, and J. J. Zou (2002), Distribution of soil carbon, nitrogen and phosphorus along Northeast China transect (NECT) and their relationships with climatic factors (in Chinese), *Acta Phytocologica Sin.*, *26*, 513–517.
- Wania, F., and J. E. Haugen (1999), Long term measurements of wet deposition and precipitation scavenging of hexachlorocyclohexanes in Southern Norway, *Environ. Pollut.*, *105*, 381–386, doi: 10.1016/S0269-7491(99)00038-X.
- Wania, F., and D. Mackay (1999), Global chemical fate of alpha-hexachlorocyclohexane. 2. Use of a global distribution model for mass

- balancing, source apportionment, and trend prediction, *Environ. Toxicol. Chem.*, 18(7), 1400–1407, doi: 10.1897/1551-5028(1999)018 < 1400:Gefohu > 2.3.Co;2.
- Wania, F., and J. N. Westgate (2008), On the mechanism of mountain cold-trapping of organic chemicals, *Environ. Sci. Technol.*, 42, 9092–9098, doi: 10.1021/es8013198.
- Xiao, H., S. C. Kang, Q. G. Zhang, W. W. Han, M. Loewen, F. Wong, H. Hung, Y. D. Lei, and F. Wania (2010), *Transport of Semivolatile Organic Compounds to the Tibetan Plateau: Monthly Resolved Air Concentrations at Nam Co*, *J. Geophys. Res.-Atmos.*, 115, D16310, doi:10.1029/2010jd013972.
- Zhang, G., P. Chakraborty, J. Li, P. Sampathkumar, T. Balasubramanian, K. Kathiresan, S. Takahashi, A. Subramanian, S. Tanabe, and K. C. Jones (2008), Passive atmospheric sampling of organochlorine pesticides, polychlorinated biphenyls, and polybrominated diphenyl ethers in urban, rural, and wetland sites along the coastal length of India, *Environ. Sci. Technol.*, 42, 8218–8223, doi: 10.1021/Es8016667.
- Zhao, Y., and Z. Yu (2012), Vegetation response to Holocene climate change in East Asian monsoon-margin region, *Earth-Sci. Rev.*, 113(1–2), 1–10, doi: 10.1016/j.earscirev.2012.03.001.