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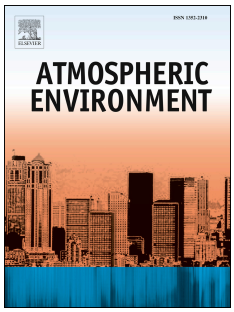

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Corresponding Author: Qingbo Li
ADDRESS: College of Environmental Science and Engineering, Dalian Maritime University, Dalian 116026, China
PHONE: 86-0411-84727771
FAX: 86-0411-84727771
E-MAIL: drliqb@gmail.com

Analysis for Sources of Atmospheric α- and γ-HCH in Gas and Particle-associated Phase in Dalian, China by Multiple Regression

QINGBO LI*†, YAN LI††, XIANYU WANG††, RUIQI ZHANG†††, JIANMIN MA††††, MENGXUE SUN†, XIAONING LV†, JIAOLING BAO†

† College of Environmental Science and Engineering, Dalian Maritime University, Dalian 116026, China

††National Research Centre for Environmental Toxicology, The University of Queensland, 39 Kessels Road, Coopers Plains, QLD, 4108, Australia

†††Shaanxi University of Science & Technology, Shaanxi 710021, China

††††Key Laboratory of Western China's Environmental System, Ministry of Education, College of Earth and Environment Sciences, Lanzhou University, Lanzhou 730000, China

*Corresponding author phone: +86 411 8472-7771; fax: +86 411 8472-7771; e-mail: drliqb@gmail.com

Abstract:

Atmospheric concentrations of α- and γ-hexachlorocyclohexanes were measured once a week in Dalian throughout 2008, using a high-volume air sampler, to estimate diurnal, monthly and seasonal variations. Multiple regression analysis was used to estimate the impact of selected meteorological conditions on atmospheric concentrations of hexachlorocyclohexanes and to identify the potential source regions. Overall, α- and γ-hexachlorocyclohexanes were mainly associated with the gas phase, with an annual mean gas-phase concentration of 36±30 and 10±9.8 pg m^{-3} respectively. On the other hand, mean particle (PM_{10}) associated concentrations throughout the year were 1.9±2.4 and 0.46±0.43 pg m^{-3} respectively. Gas-phase concentration of α- and γ-hexachlorocyclohexanes peaked in the autumn season whereas highest concentrations in the particle phase were measured in spring. Ratio of α-/γ-isomer ranged from 3.7 to 7.4 in the gas phase which was close to the ratio in technical hexachlorocyclohexanes (5~7). In the particle-associated phase this ratio ranged from 1.2 to 3.8, with the exception of daytime samples in spring (up to 16) and summer seasons (up to 14) and this exception could be due to the isomerization from γ- to α- in ambient air, at least partly resulted from the impact of sunlight. Regression analysis showed that, at the sampling site, concentrations of α- and γ-hexachlorocyclohexanes in the gas phase were both elevated with increasing temperature and wind speed, whereas in the particle-associated phase their concentrations tended to remain stable.

Keywords: Hexachlorocyclohexanes; Gas phase; Particle-associated phase; Multiple Regression; Sources

1. Introduction

Hexachlorocyclohexanes (HCHs) were used extensively between the 1960s and the 1980s and cumulative consumption of technical HCHs has been estimated as 6 million tons globally (Willett, Ulrich, & Hites, 1998). In China, HCHs were widely used for agricultural purposes and vector control and the amount of technical HCHs used in China and South Asia during the late 1970s was estimated at about 60,000 tons per year (Willett et al., 1998). During and after application, large amounts of HCHs can be released and condensed into environmental reservoirs such as soil, vegetation and cryosphere. Subsequently, the stored HCHs can re-volatilize from these secondary sources into environmental compartments and can result in exposure of humans and the whole ecosystem. Temperature is invoked as the major controller for semi-volatile organic compounds (SVOCs) such as HCHs to condense into/(re-)volatilize from environmental reservoirs, effectively accounting for their cycling nature between air and earth's surfaces (Halsall et al., 1999). Several studies have interpreted this phenomenon using the Clausius-Clapeyron equation, in which chemical concentrations in air, expressed as partial pressure, are plotted against ambient temperature (Hoff, Mulr, & Grift, 1992; Hornbuckle & Eisenreich, 1996; Venier & Hites, 2010). Besides, wind speed and direction are also considered as the major factors, which can be integrated into a multiple regression model including also reciprocal temperature, to interpret the impact of these meteorological parameters on the concentrations of SVOCs in ambient air. For example, Hillery et al. used this equation to relate atmospheric PCB concentrations to meteorological conditions, thereby to study the temporal and spatial trends of gas-phase PCB concentrations near the Great Lakes (Hillery, Basu, Sweet, & Hites, 1997).

Dalian is a seaside city where the distribution of atmospheric pollutants could be influenced by diel cycle of sea-land breeze. However, to our best knowledge, limited reports are available for diurnal and seasonal variations of organochlorine pesticides (OCPs) including HCHs (Li et al., 2012; Li et al., 2011) and for their source analysis within this city. This study aims to obtain diurnal and seasonal data for atmospheric HCHs in Dalian air and to analysis their sources by a multiple regression model both in the gas and particle-associated phase.

2. Materials and methods

2.1 Sample collection

The sampling site is located in Dalian (121°31'E, 38°52'N), in the southern tip of Liaodong peninsular in northeast China, adjacent to the Yellow Sea and Bohai Sea (the supplementary information (SI) Figure S1). The city locates in the temperate zone featured by a typical maritime continental monsoon climate. A sampling platform (altitude 12 m

above the ground) is mounted on the Technology Building of Dalian Maritime University and is ~10 km to the city center and ~1 km to the Yellow Sea and lacks of major proximate OCP sources. Detailed sampling method has been published elsewhere (Li et al., 2012). Briefly, a high-volume air sampler operating at a flow rate at about 1.0 m^3 min^{-1} was used to collect gas-phase and particle (PM_{10})-associated-phase samples simultaneously once a week throughout the year of 2008. This sampling platform is also equipped with a mini weather station (Watchdog 900ET) for monitoring and recording meteorological parameters continuously, including wind speed and direction, ambient temperature, relative humidity and solar radiance at the height analogues to that the air sampler was mounted. Data were recorded automatically every 6 seconds into a data-logger and output as hourly mean values.

2.2 Sample analysis

Sample extraction, cleanup and analysis were published in detail elsewhere (Li et al., 2012). Briefly, samples were extracted by *n*-hexane and cleaned up by a silica gel/neutral alumina column. Sample analysis was performed with a Shimadzu 2010 gas chromatograph (GC) equipped with a micro electron capture detector (*m*-ECD).

2.3 Quality control and quality assurance (QC/QA)

Detailed QC/QA procedure was reported elsewhere (Li et al., 2012). In brief, limits of detection (LOD) for α- and γ-HCH in the gas phase are 0.071 and 0.37 pg m^{-3} respectively and in the particle phase, LOD for α- and γ-HCH were 0.033 and 0.052 pg m^{-3}, respectively.

2.4 Multiple regressions

Measured concentrations (pg m^{-3}) of α- and γ-HCH were converted to partial pressures (atm) using the ideal gas law. Air temperature (*T* in K) and wind speed (WS in mph) were regressed into the logarithms of the atmospheric partial pressures (Hillery et al., 1997):

$$\ln P = b0 + b1\,(1/T) + b2\,WS + b3\,\sin WD + b4\,\cos WD, \quad (1)$$

where b0 is an intercept and b1 and b2 describes the dependence of the partial pressure on reciprocal *T* and WS respectively; b3 and b4 are coefficients of the sine WD (wind direction in degree) and the cosine WD terms.
Two different methods, enter and stepwise method, which are fitted using IBM SPSS statistics 19, are used to run the multiple regression model separately.

3. Results and discussion

3.1 Ambient concentrations of HCHs in Dalian atmosphere

149 *α-HCH*. As expected, α-HCH distributed mainly in the gas phase (ratio of $C_{gas}/C_{particle}$ shown in SI Table S3), where
150 a mean concentration of 34±27 and 32±29 pg m^{-3} was measured during daytime and nighttime respectively
151 throughout the year (SI Table S1). In contrast, in the particle-associated phase, one order of magnitude lower
152 concentration was observed as 1.6±1.8 and 1.6±2.1 pg m^{-3} during daytime and the nighttime, respectively (SI Table
153 S2). As seen in Figure 1, in the gas phase, in terms of median concentration value in each month, the highest during
154 daytime and nighttime was both observed in September at 72 pg m^{-3} respectively. Whereas for particle-associated
155 α-HCH, this value during daytime and nighttime was both measured in April, at 2.8 and 3.5 pg m^{-3} respectively
156 (Figure 2). Seasonally, concentration of gaseous and particle-associated α-HCH peaked in autumn and spring season
157 respectively both for daytime and nighttime (SI Table S1&S2).
158
159 *γ-HCH*. Similarly, dominant annual mean concentration was measured in the gas phase (SI Table S3), at 10±10 and
160 11±9.6 pg m^{-3} during the daytime and the nighttime respectively, compared to the one in the particle-associated
161 phase at 0.46±0.38 (daytime) and 0.58±0.56 pg m^{-3} (nighttime) (SI Table S1&S2). As shown in Figure 1, median
162 value of γ-HCH concentration in the gas phase was highest in October (daytime, at 18 pg m^{-3}) and September
163 (nighttime, also at 18 pg m^{-3}), respectively. In the particle-associated phase, on the other hand, this value was
164 observed highest in January for the daytime (at 0.59 pg m^{-3}) and March for the nighttime (at 1.0 pg m^{-3}) (Figure 2).
165 Seasonal variation of concentration of γ-HCH was similar to that of α-HCH, as presented in SI Table S1&S2.
166
167 **3.2 Isomer ratio ($C_{α\text{-HCH}}/C_{γ\text{-HCH}}$) and its indications**
168 The ratio of $C_{α\text{-HCH}}/C_{γ\text{-HCH}}$ is typically used as an indicator to trace origin and pathway of HCHs in the air (Iwata,
169 Tanabe, Sakai, & Tatsukawa, 1993). As seen in SI Table S1&S2, this ratio for gaseous HCHs in each season ranged
170 from 3.7 to 7.4, which was close to the ratio in technical HCHs (5~7), whereas in the particle-associated phase, this
171 ratio was measured as 1.2 to 3.8, with the exception of daytime samples in spring (at 16) and summer seasons (at
172 14). A possible reason for this higher ratio could be the significant isomerization of γ-isomer into α-isomer, which
173 has been experimentally demonstrated in the presence of ferrous salts and sunlight (Malaiyandi & Shah, 1984). That
174 is, during the daytime, γ-isomer sorbed on the surface of particles may be isomerized intensively into the α-isomer,
175 resulted at least partly from the impact of sunlight.
176
177 **3.3 Impact of meteorological conditions on atmospheric concentrations of HCHs**
178
179 *Gas phase.* Under the enter method, as shown in Table 1, during daytime, integrated impact of temperature, wind
180 velocity and wind direction on atmospheric concentrations of α- and γ-HCH was found at significant level (R^2=0.41

and 0.45 for α- and γ-HCH respectively, $p<0.05$), whereas during nighttime it was only the case for γ-HCH ($R^2=0.24$, $p<0.05$). Atmospheric concentration of α- and γ-HCH at the sampling site both elevated with increased temperature and wind speed ($b1<0$ and $b2>0$). The correlation between concentration and temperature was statistically significant at $p<0.05$ during daytime for α-HCH and both during daytime and nighttime for γ-HCH. For the correlation between concentration and wind speed, statistical significance ($p<0.05$) was observed only for α-HCH during daytime. The insignificant impact from wind direction for both isomers indicated that spatial variations of potential sources for atmospheric HCHs at the sampling site were probably not significant, i.e. re-emission from terrestrial and ocean surface does not cause significant variations of concentrations of HCHs in ambient air at the sampling site.

As seen in Table 1, for both α- and γ-HCH, the impact from temperature on concentrations was more significant ($|b1| >> |b2|$) compared to wind speed (although for α-HCH neither reached a significant level at 0.05 during nighttime). Both of b3 and b4 were lower than 0 (and were marginally insignificant) for α-HCH, indicating that atmospheric concentration at the sampling site may be influenced by α-HCH transported by southwesterly wind. The area southwest of the sampling site is the Shandong Peninsular, which is the top province in China in terms of agricultural activities. Therefore, it is possible that gaseous α-HCH in the atmosphere at the sampling site was (partly) from re-volatilization from the Shandong Peninsular and surrounding sea water reservoirs (presumably the historic residues) (the geographic information can be referred in the SI Figure S1). The results of the back trajectory analysis of the air masses during this sampling period also supported this interpretation (Li et al., 2012; Li et al., 2011). For both of α- and γ-HCH, $|b1|$ was higher during daytime as compared with nighttime, indicating that the emission from terrestrial surfaces/reservoirs during the daytime exerted stronger impacts on their atmospheric concentrations, i.e. the influence of proximate sources was stronger on the concentrations at daytime than that on nighttime. Besides, $|b1|$ for γ-HCH was greater than that for α-HCH during both of the daytime and nighttime, indicating that the influence of proximate sources was stronger on the atmospheric concentrations of γ-HCH compared to that of α-HCH. Multicollinearity diagnostics of independent variables within equation (1) under the enter method were also carried out by evaluating eigenvalue, condition index, variance proportions and variance inflation factor (VIF). As shown in the SI Table S4, eigenvalue of dimension 5 was ≤0.001 and condition index was over 70 for each group and a high (1.0) variance proportion was detected for inverse *T* against dimension 5, indicating a possibility of multicollinearity inherent in the regression model between cosWD and inverse *T*, although this was not supported by the low VIF (≈1) (Jeeshim, 2003).

Given the insignificances and possible multicollinearity issue observed above for the enter method, a stepwise method was then applied for modelling gaseous HCHs and the result showed that the variable temperature can be included for α- and γ-HCH during daytime and γ-HCH during night ($P<0.002$) and sinWD can be included for α-HCH during night ($P=0.026$) (SI Table S6), i.e. variations of concentrations of α- and γ-HCH during daytime and γ-HCH during night were mainly controlled by temperature (invoking re-volatilization from terrestrial and ocean surface). For α-HCH at night time, although the above result indicated that the concentration could be controlled by contaminants delivered by wind from north, B value of variable sinWD was nevertheless two orders of magnitudes lower than the one of constant, which implied that the impact from north wind was limited.

Particle-associated phase. As seen in Table 2, under the enter method, integrated impact of temperature, wind speed and wind direction on atmospheric concentrations of α- and γ-HCH was found at insignificant level ($p>0.05$), which implied that this model may be inadequate to explain/predict the variations of concentrations of particle-associated HCHs. Similarly to gaseous HCHs, possible multicollinearity was also observed for particle-associated HCHs under the enter method (SI Table S5). Thus the stepwise method was applied and modelling result showed that none of the parameters can be included, i.e. these parameters (temperature, wind speed and direction) exerted limited impact on the variation of concentrations of particle-associated HCHs. As seen in Figure 2, concentrations of particle-associated HCHs during daytime and night time were close to each other and no obvious monthly variations/differences can be observed (ANOVA P lies between 0.26 and 0.59). In addition, partial regression of concentrations of particle-associated HCHs (shown as ln P) against meteorological parameters, as seen in Figure 3&4, showed scattered relationship, indicating their concentrations tended to remain stable rather than varying with these parameters. This result was opposite to that of another pesticide, endosulfan, analyzed from the same sample set, where an obvious monthly trend was observed, which was partly attributed to the changes of temperature and air mass origins between months (Li et al., 2012).

4. Conclusions

HCHs distribute mainly in the gas phase in Dalian air and mean concentration of α-HCH is generally higher than that of γ-HCH. Concentration of gaseous and particle-associated HCHs peaks in autumn and spring respectively. The main controlling factor for gaseous HCH concentrations is ambient temperature and concentration of particle-associated HCHs tends to remain stable, i.e. temperature, wind speed and direction exert limited impact. Spatial variations of potential sources for atmospheric HCHs at the sampling site are probably not significant, i.e. re-emission from terrestrial and ocean surface does not cause significant variations of concentrations of HCHs in

ambient air at the sampling site. Emission characteristics from terrestrial and ocean surface should be further studied to illustrate spatial variations of potential sources for atmospheric HCHs.

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Table 1. Multiple regression (enter method) results for gaseous HCHs

		ln *P*=b0+b1 (1/*T*)+b2 WS+b3 sin WD+b4 cos WD					
		α-HCH			γ-HCH		
		UC	SE	Sig.	UC	SE	Sig.
Day (n=50)	b0	-19	4.2	1.0E-5	-15	3.6	1.2E-4
	b1	-4,400	1,200	0.011	-5,800	1,000	1.4E-6
	b2	0.13	0.06	0.042	0.083	0.062	0.14
	b3	-0.42	0.21	0.051	0.083	0.18	0.65
	b4	-0.40	0.21	0.052	0.064	0.18	0.76
Night (n=49)	b0	-27	5.3	5.9E-6	-22	4.4	1.4E-5
	b1	-1,800	1,500	0.23	-3,800	1,200	3.9E-3
	b2	0.064	0.093	0.94	0.012	0.073	0.89
	b3	-0.48	0.24	0.051	-0.25	0.19	0.22
	b4	-0.043	0.25	0.87	-0.052	0.21	0.82

***T* is temperature in K; WS is wind speed in mph; WD is wind direction in degree. UC is unstandardized coefficient; SE is std. error.**

α-HCH: Integrated correlation coefficient R^2=0.41 and Sig. (F-test)=0.04 during daytime. Integrated correlation coefficient R^2=0.13 and Sig. (F-test)=0.18 at night;

γ-HCH: Integrated correlation coefficient R^2=0.45 and Sig. (F-test)=2.38E-5 during daytime. Integrated correlation coefficient R^2=0.24 and Sig. (F-test)=0.02 at night.

Table 2. Multiple regression (enter method) results for particle-associated HCHs

		ln P=b0+b1 (1/T)+b2 WS+b3 sin WD+b4 cos WD					
		α-HCH			γ-HCH		
		UC	SE	Sig.	UC	SE	Sig.
Day (n=45)	b0	-44	5.8	2.7E-9	-42	3.9	3.0E-13
	b1	2,000	1,600	0.23	1,200	1,100	0.29
	b2	-7.0E-3	0.093	0.94	-0.033	0.064	0.63
	b3	0.11	0.29	0.71	-0.27	0.19	0.19
	b4	-0.093	0.29	0.75	-0.19	0.20	0.35
Night (n=43)	b0	-39	7.0	2.0E-6	-39	4.5	2.3E-10
	b1	640	2,000	0.75	260	1,200	0.84
	b2	-0.034	0.11	0.81	-0.082	0.072	0.25
	b3	0.33	0.33	0.32	-0.39	0.21	0.071
	b4	-0.27	0.36	0.46	-0.22	0.22	0.33

α-HCH: Integrated correlation coefficient R^2=0.06 and Sig. (F-test)=0.68 during daytime. Integrated correlation coefficient R^2=0.05 and Sig. (F-test)=0.79 at night;

γ-HCH: Integrated correlation coefficient R^2=0.08 and Sig. (F-test)=0.51 during daytime. Integrated correlation coefficient R^2=0.16 and Sig. (F-test)=0.17 at night.

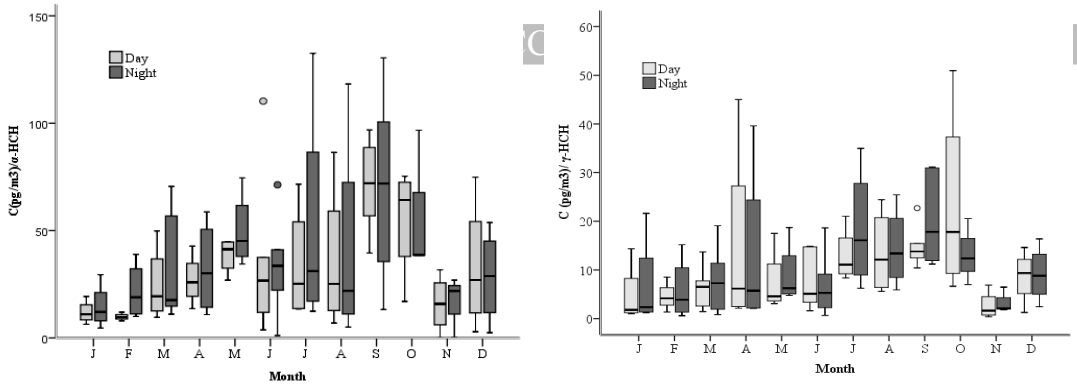

Figure 1. Monthly variations in α-HCH (left panel) and γ-HCH (right panel) concentrations in gas phase

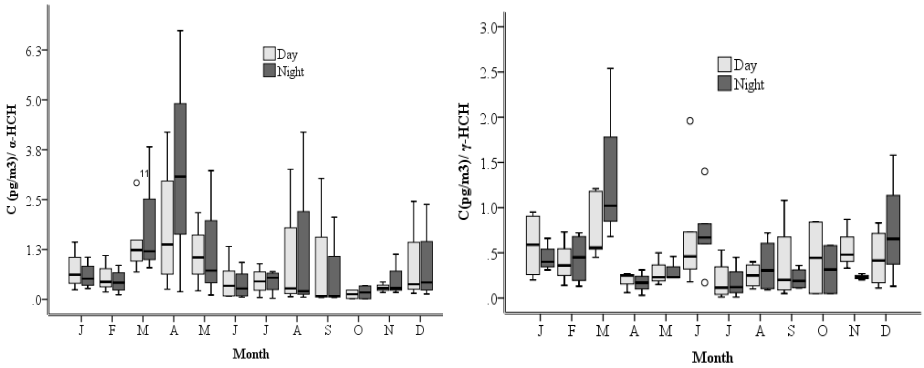

Figure 2. Monthly variations in α-HCH (left panel) and γ-HCH (right panel) concentrations in particle-associated phase

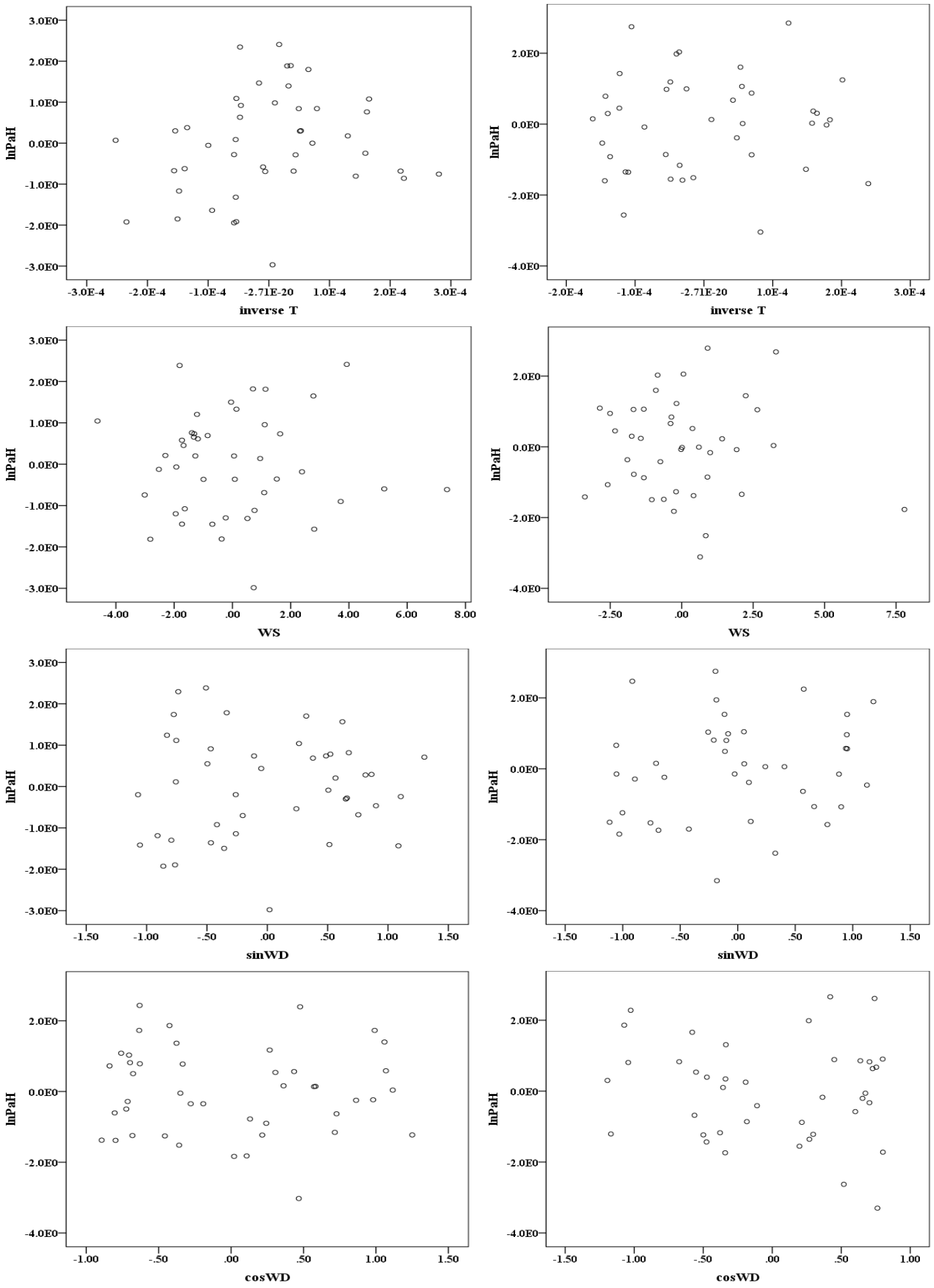

Figure 3. Partial regression of α-HCH (day on the left and night on the right) in particle-associated phase against meteorological

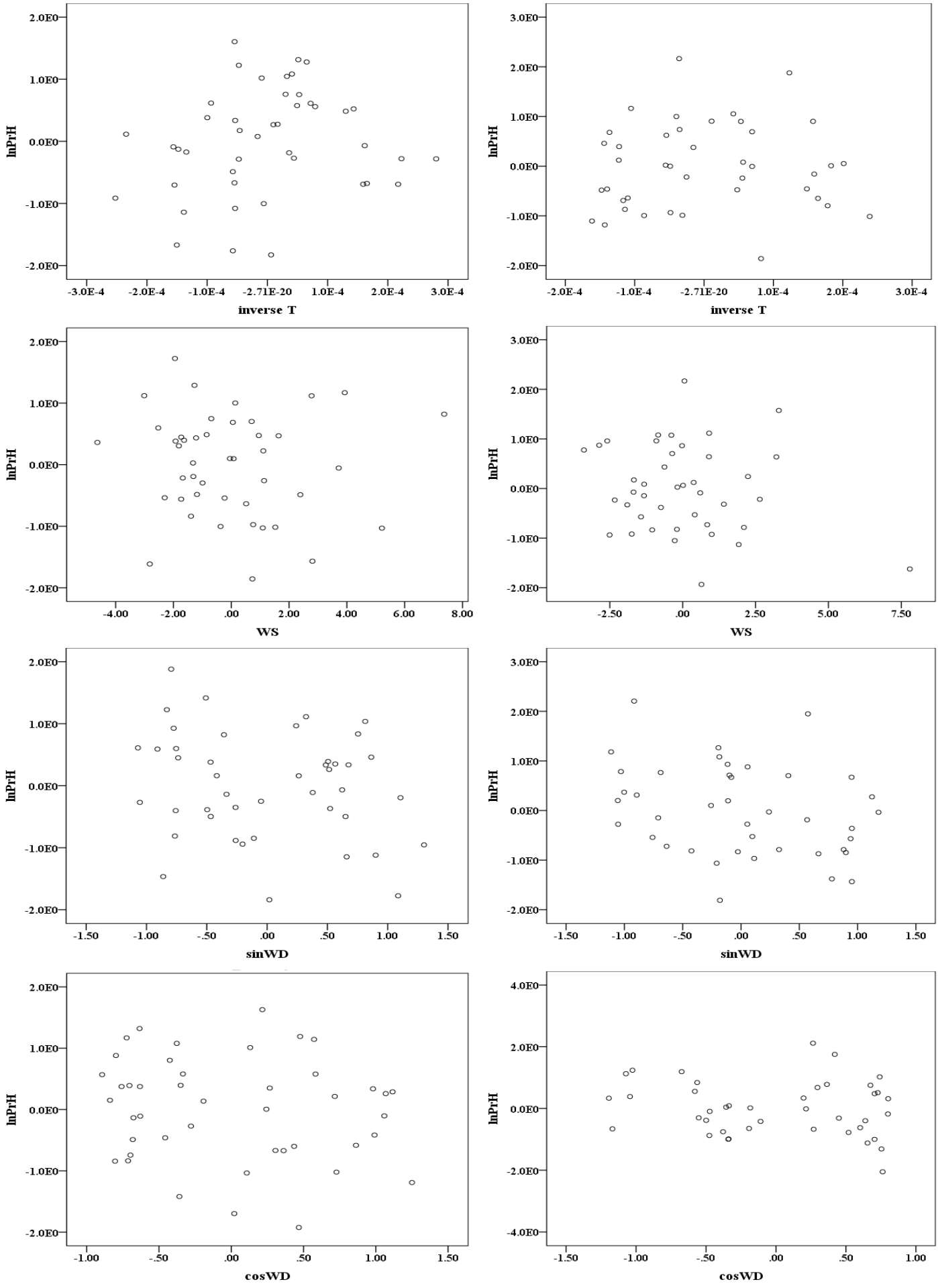

Figure 4. Partial regression of γ-HCH (day on the left and night on the right) in particle-associated phase against meteorological parameters

Highlights

- Diurnal and seasonal variations of HCHs in Dalian air are first time reported
- Gaseous and particle-associated HCHs peaked in autumn and spring respectively
- Multiple regression analysis was used to assess impact of meteorological parameters
- Concentration of particle-associated HCHs tended to remain stable

Supplementary information

Analysis for Sources of Atmospheric α- and γ-HCH in Gas and Particle-associated Phase in Dalian, China by Multiple Regression

QINGBO LI*†, YAN LI††, XIANYU WANG††, RUIQI ZHANG†††, JIANMIN MA††††, MENGXUE SUN†, XIAONING LV†, JIAOLING BAO†

† College of Environmental Science and Engineering, Dalian Maritime University, Dalian 116026, China

††National Research Centre for Environmental Toxicology, The University of Queensland, 39 Kessels Road, Coopers Plains, QLD, 4108, Australia

†††Shaanxi University of Science & Technology, Shaanxi 710021, China

††††Key Laboratory of Western China's Environmental System, Ministry of Education, College of Earth and Environment Sciences, Lanzhou University, Lanzhou 730000, China

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Table S1. Atmospheric concentrations of gaseous α- and γ-HCH (pg m^{-3}) and $C_{\alpha\text{-HCH}}/C_{\gamma\text{-HCH}}$ at the sampling site

		n	α-HCH				γ-HCH				$C_{\alpha\text{-HCH}}/C_{\gamma\text{-HCH}}$			
			Mean	S.D	Min	Max	Mean	S.D	Min	Max	Mean	S.D	Min	Max
Spring (Mar-May)	Day	13	30	14	9.6	50	9.3	12	1.5	45	5.7	3.6	0.95	12
	Night	13	38	23	11	75	10	11	0.8	40	7.4	6.1	1.2	19
Summer (Jun-Aug)	Day	13	36	33	3.8	110	11	7.0	1.6	24	3.7	3.0	0.41	11
	Night	13	42	41	1.1	130	13	9.9	0.66	35	4.4	4.6	0.20	18
Autumn (Sep-Nov)	Day	13	49	31	0.071	97	14	13	0.37	51	5.3	3.5	0.19	11
	Night	11	52	42	0.36	130	14	10	1.8	31	4.3	4.2	NA	14
Winter (Dec-Feb)	Day	11	19	20	2.9	75	6.2	5.3	0.99	15	3.8	2.8	NA	8.3
	Night	12	22	16	2.5	54	7.3	7.0	0.60	22	6.9	11	0.66	42
Annual (Jan-Dec)	Day	50	34	27	0.074	110	10	10	0.37	51	4.7	3.3	NA	12
	Night	49	32	29	0.36	133	11	9.6	0.60	40	5.7	7.0	NA	42

NB: Mean is arithmetic mean value; S.D is standard deviation; Min is minimum value; Max is maximum value; NA means that the value is not available

Table S2. Atmospheric concentrations of particle-associated α- and γ-HCH (pg m^{-3}) and $C_{\alpha\text{-HCH}}/C_{\gamma\text{-HCH}}$ at the sampling site

		n	α-HCH				γ-HCH				$C_{\alpha\text{-HCH}}/C_{\gamma\text{-HCH}}$			
			Mean	S.D	Min	Max	Mean	S.D	Min	Max	Mean	S.D	Min	Max
Spring (Mar-May)	Day	12	3.0	2.3	0.43	8.4	0.51	0.35	0.15	1.2	16	3.6	2.0	19
	Night	9	3.3	2.8	0.22	7.6	1.1	0.89	0.23	2.5	3.4	3.1	0.96	12
Summer (Jun-Aug)	Day	13	0.81	0.75	0.091	2.7	0.42	0.50	0.07 1	2.0	14	2.8	0.091	9.8
	Night	13	1.3	2.2	0.054	8.4	0.51	0.39	0.10	1.4	3.8	4.6	0.12	13
Autumn (Sep-Nov)	Day	9	0.97	1.9	0.043	6.1	0.45	0.38	0.052	1.1	1.2	1.7	0.56	5.6
	Night	9	0.93	1.4	0.032	4.1	0.24	0.16	0.051	0.58	3.2	3.9	0.58	11
Winter (Dec-Feb)	Day	11	1.5	1.4	0.30	4.9	0.48	0.32	0.11	0.95	3.8	4.1	0.50	14
	Night	12	1.3	1.2	0.23	4.8	0.54	0.39	0.13	1.6	2.8	2.2	0.39	6.8
Annual (Jan-Dec)	Day	45	1.6	1.8	0.041	8.4	0.46	0.38	0.051	2.0	9.2	4.2	0.092	19
	Night	43	1.6	2.1	0.032	8.4	0.58	0.56	0.054	2.5	3.3	3.5	0.12	13

Table S3. $C_{gas}/C_{particle}$ for α- and γ-HCH at the sampling site

		n	α-HCH				γ-HCH			
			Mean	S.D	Min	Max	Mean	S.D	Min	Max
Spring (Mar-May)	Day	12	21	25	3.2	84	30	47	2.7	180
	Night	9	46	64	2.9	160	38	41	2.8	130
Summer (Jun-Aug)	Day	13	120	200	NA	740	130	270	NA	1,000
	Night	13	140	150	2.1	420	220	560	2.8	2,100
Autumn (Sep-Nov)	Day	9	290	360	0.21	1,100	170	330	1.1	1,000
	Night	9	120	160	0.64	400	48	41	NA	100
Winter (Dec-Feb)	Day	11	31	69	NA	250	13	12	NA	42
	Night	12	36	54	3.7	200	17	19	0.83	58
Annual (Jan-Dec)	Day	45	100	210	NA	1,100	79	210	NA	1,000
	Night	43	85	120	0.64	420	88	310	NA	2,100

Table S4. Multicollinearity diagnostics of multiple regressions (enter method) (gas phase)

		Dimension	Eigenvalue	Condition index	Variance proportions				VIF
					Inverse T	WS	sinWD	cosWD	
Daytime	α-HCH	1	2.982	1.000	0.00	0.02	0.00	0.01	
		2	1.103	1.644	0.00	0.00	0.53	0.29	1.064
		3	0.791	1.942	0.00	0.00	0.41	0.69	1.017
		4	0.123	4.924	0.00	0.98	0.01	0.00	1.086
		5	**0.001**	**73.379**	**1.00**	0.00	0.05	0.01	1.031
	γ-HCH	1	2.982	1.000	0.00	0.02	0.00	0.01	
		2	1.103	1.644	0.00	0.00	0.53	0.29	1.064
		3	0.791	1.942	0.00	0.00	0.41	0.69	1.017
		4	0.123	4.924	0.00	0.98	0.01	0.00	1.086
		5	**0.001**	**73.379**	**1.00**	0.00	0.05	0.01	1.031
Nighttime	α-HCH	1	2.881	1.000	0.00	0.03	0.00	0.02	
		2	1.120	1.604	0.00	0.01	0.57	0.23	1.073
		3	0.760	1.948	0.00	0.00	0.38	0.74	1.060
		4	0.238	3.477	0.00	0.92	0.04	0.00	1.070
		5	**0.000**	**76.771**	**1.00**	0.04	0.01	0.02	1.053
	γ-HCH	1	2.881	1.000	0.00	0.03	0.00	0.02	
		2	1.120	1.604	0.00	0.01	0.57	0.23	1.073
		3	0.760	1.948	0.00	0.00	0.38	0.74	1.060
		4	0.238	3.477	0.00	0.92	0.04	0.00	1.070
		5	**0.000**	**76.771**	**1.00**	0.04	0.01	0.02	1.053

Table S5. Multicollinearity diagnostics of multiple regressions (enter method) (particle-associated phase)

		Dimension	Eigenvalue	Condition index	Variance proportions				VIF
					Inverse T	WS	sinWD	cosWD	
Daytime	α-HCH	1	2.958	1.000	0.00	0.02	0.00	0.01	
		2	1.075	1.659	0.00	0.00	0.55	0.28	1.104
		3	0.834	1.883	0.00	0.00	0.35	0.70	1.022
		4	0.132	4.732	0.00	0.97	0.02	0.00	1.121
		5	**0.001**	**72.238**	**1.00**	0.00	0.08	0.01	1.022
	γ-HCH	1	2.958	1.000	0.00	0.02	0.00	0.01	
		2	1.075	1.659	0.00	0.00	0.55	0.28	1.104
		3	0.834	1.883	0.00	0.00	0.35	0.70	1.022
		4	0.132	4.732	0.00	0.97	0.02	0.00	1.121
		5	**0.001**	**72.238**	**1.00**	0.00	0.08	0.01	1.022
Nighttime	α-HCH	1	2.906	1.000	0.00	0.03	0.00	0.02	
		2	1.151	1.589	0.00	0.01	0.51	0.21	1.050
		3	0.682	2.064	0.00	0.00	0.44	0.76	1.057
		4	0.260	3.342	0.00	0.91	0.04	0.00	1.096
		5	**0.001**	**74.645**	**1.00**	0.04	0.01	0.00	1.070
	γ-HCH	1	2.906	1.000	0.00	0.03	0.00	0.02	
		2	1.151	1.589	0.00	0.01	0.51	0.21	1.050
		3	0.682	2.064	0.00	0.00	0.44	0.76	1.057
		4	0.260	3.342	0.00	0.91	0.04	0.00	1.096
		5	**0.001**	**74.645**	**1.00**	0.04	0.01	0.00	1.070

Table S6. Parameters of regression model running under stepwise method for gaseous HCHs

	Model (Stepwise)		Dimension	UC		SC	t	Sig.	Eigenvalue	Condition Index	Variance Proportions		VIF
	R^2	p		B	Std.	Beta					1	2	
lnP α-HCH (Day)	0.2384	3.760E-4	(Constant)	-16.816	4.468		-3.763	4.648E-4	1.999	1.000	2.957E-4	2.957E-4	1.000
			inverseT	-4897.983	1278.109	-.488	-3.832	3.760E-4	.001	58.143	1.00	1.00	
lnP γ-HCH (Day)	0.417	5.524E-7	(Constant)	-14.986	3.486		-4.299	8.576E-5	1.999	1.000	2.957E-4	2.957E-4	1.000
			inverseT	-5776.657	997.129	-.645	-5.793	5.524E-7	.001	58.143	1.00	1.00	
lnP α-HCH (Night)	0.324	0.026	(Constant)	-33.804	.162		-208.411	8.106E-69	1.018	1.000	.49	.49	1.000
			sinWD	-.520	.226	-.324	-2.297	.026	.982	1.018	.51	.51	
lnP γ-HCH (Night)	0.202	0.002	(Constant)	-21.001	4.165		-5.043	7.978E-6	1.999	1.000	2.650E-4	2.650E-4	1.000
			inverseT	-3974.350	1178.531	-.449	-3.372	.002	.001	61.414	1.00	1.00	

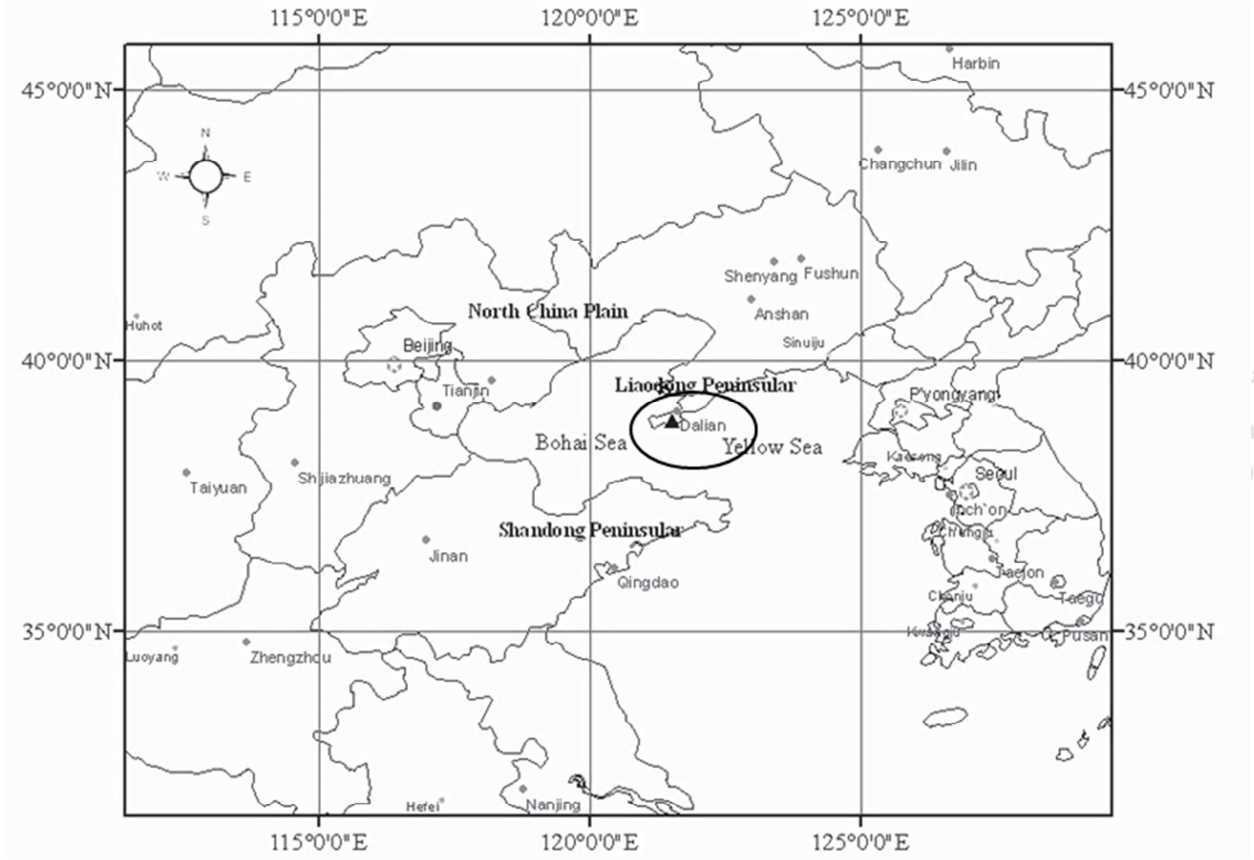

Figure S1. Map of sampling site