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Spatial concentration, congener profiles and inhalation risk assessment of PCDD/Fs and PCBs in the atmosphere of Tianjin, China

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Polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) were monitored in a seasonal passive sampling scheme during June 2008 and January 2009 to investigate the spatial concentration, congener profiles and evaluate the potential inhalation risk in different functional areas in Tianjin, China. The spatial air concentrations and I-TEQs ranged $1.08 \times 10^2 - 8.19 \times 10^3$ fg m⁻³ (average 2.63×10^3 fg m⁻³) and 4.08 - 325 fg I-TEQ m⁻³ (average 91.4 fg I-TEQ m⁻³) respectively for PCDD/Fs. The concentration and I-TEQs of PCBs were $3.08 \times 10^4 - 3.01 \times 10^5$ fg m⁻³ (average 1.39×10^5 fg m⁻³) and 1.72 - 49.6 fg I-TEQ m⁻³ (average 18.5 fg I-TEQ m⁻³). It is obvious that PCB concentrations were several hundred times higher than the PCDD/Fs. However, the ambient air PCDD/Fs contributed a major part to the total toxicity equivalents, varying from 72.7% to 89.0% (average 81.8%). The atmospheric PCDD/F levels were observed to be higher in winter for most of sampling sites in the downtown. Besides, inhalation risk assessment showed that local residents might suffer more risk near the point sources than those in living area, industrial zones and background area in Tianjin City. However, the total daily dioxin intake was approximately several to hundreds of times lower than the WHO criteria, showing relatively low exposure risks from the impact of industry point sources in Tianjin City.

PCDD/Fs, PCBs, ambient air, inhalation risk assessment

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Polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/ Fs) and polychlorinated biphenyls (PCBs) are two groups of semi-volatile organic compounds that could be formed and released into the environment through anthropogenic activities such as combustion and chlorinated chemical industry processes [1,2]. These chemicals have been raised much concern for their persistence in the environment, long-range transport in the atmosphere and bioaccumulative abilities in biological species in the food chain. The Chinese government ratified the National Implementation Plan (NIP) on persistent organic pollutants (POPs) in 2004 to meet the

criteria of the Stockholm Convention for controlling and eliminating four categories of POPs, including pesticides, stockpiles, PCBs and unintentionally produced POPs [3]. The government aims to establish a dioxin and PCB inventory of a variety of emission sources associated with anthropogenic activities by the year of 2015 [4].

With the rapid economic development, a series of environmental issues emerge in some regions of China, which have seriously threatened public health of local citizens in the recent years. Thus, several studies on PCDD/Fs in China have been implemented under the NIP guidance and mainly focused on the atmospheric PCDD/F concentrations in urban areas to evaluate the impact of PCDD/Fs on the local

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residents. For example, the concentrations of PCDD/Fs have been reported in the ambient atmosphere of Guangzhou, Beijing and Shanghai respectively [5-7]. The results showed that the PCDD/Fs levels in the investigated areas were comparable or slightly higher than those of many other urban cities around the world. The observed phenomenon of equal or even higher atmospheric concentration of dioxinlike compounds were thought to be associated with point sources, such as small diffuse combustion points, active E-waste dismantling sites and steel manufacturing plants. However, further investigation should be kept continuing to further support the NIP implementation procedures, especially for some rapid developing areas with much industrial activities. Tianjin is one of the most important traditional industry bases in the Northern China and the manufacturing industry scale expanded with another new National Development Zone established in 2008. It is thought that a large amount of anthropogenic activities would continue to be extremely active in this area since the recent several decades. Thus, the studies concerning the atmospheric PCDD/Fs concentrations in industry bases such as Tianjin City could not only help understanding the spatial ambient concentration and inhalation risk potentials of the dioxin-like compounds in the city, but also benefit from evaluating the participation role of industrial process to the levels of PCDD/Fs in the environment. Furthermore, studies on atmospheric dioxin-like PCBs (DL-PCBs) in rarely reported Chinese cities would also be helpful for providing useful information to the NIP implementation.

During 2008–2009, a project was carried out in Tianjin City to investigate the temporal and spatial distributions of atmospheric POPs in different urban functional areas. In this work, we report the concentrations, profiles and seasonal variation of seventeen PCDD/Fs and twenty-five PCBs (including twelve DL-PCBs) in the ambient air during winter and summer period. Besides urban areas, sampling was also conducted at background area to evaluate the impact of different urban functional areas on the atmospheric PCDD/Fs and PCBs concentrations and distinct inhalation risk potentials to the local residents.

1 Method and materials

1.1 Sample collection

As shown in Figure 1 and Table 1, 22 air samples in both summer and winter period were collected separately using PUF samplers [8] during June 2008 and January 2009 in different industrial zones, living areas and background areas according to the division of functional areas of Tianjin City. The PUF disks (14 cm O.D.; 1.35 cm thickness with surface area of 365 cm²) were housed in stainless steel domed chambers to protect from direct particle deposition, precipitation and UV sunlight. Prior to use, PUF disks were rinsed with water, air-dried, extracted with acetone using an Accelerated Solvent Extractor (Dionex ASE 300, Houston, TX) at 100°C and 1500 psi for 3 extraction cycles and dried



Figure 1 Sampling location for the eleven monitoring sites in Tianjin, Northern China.

Table 1 Basic information of passive air sampling sites in the investigat	ed area
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Site	Abbreviation	GPS (E)	GPS (N)	Sampling area functions
Tianjin 1	TJ1	117°09.066	39°05.823	Living area
Tianjin 2	TJ2	117°09.066	39°05.823	Living area
Benchen Pharmaceutical Company	BC	117°07.310	39°14.201	Point source
Dongli	DL	117°22.090	39°04.124	Living area
Tianjin Chemical Plant	TC	117°48.592	39°13.973	Point source
Han'gu	HG	117°47.500	39°14.843	Industial zone
Tanggu	TG	117°39.400	39°01.261	Industial zone
Xiqing	XQ	117°02.584	39°03.308	Point source
Dagang Oil Field	DG1	117°29.987	38°43.535	Industial zone
Dagang	DG2	117°28.164	38°50.603	Industial zone
Jiushanding	JS	117°31.047	40°13.028	Background area

in a desiccator under vacuum. Sampling procedure continued for 30 d and an average value $3.5 \text{ m}^3 \text{ d}^{-1}$ was employed as the passive sampling rate based on the study of Shoeib and Harner [8]. After sampling, the sample disks were packed in aluminum foil, sealed in polyethylene bags, delivered to the laboratory immediately and kept at -20° C until analysis.

1.2 Sample analysis

All collected samples were extracted with another ASE extraction procedure with hexane and dichloromethane (1:1, v/v) using the same extraction procedures as mentioned above. Retrieved PUF disks were spiked with labeled surrogate standards which included fifteen 13C12-Dioxins (13C-2,3,7,8-TCDF, 13C-1,2,3,7,8-PeCDF, 13C-2,3,4,7,8-PeCDF, 13C-1,2,3,4,7,8-HxCDF, 13C-1,2,3,6,7,8-HxCDF, 13C-2,3,4,6,7, 8-HxCDF, 13C-1,2,3,7,8,9-HxCDF, 13C-1, 2,3,4,6,7,8-HpCDF, 13C-1,2,3,4,7,8, 9-HpCDF, 13C-2,3,7, 8-TCDD, 13C-1,2,3,7,8-PeCDD, 13C-1,2,3,4,7,8-HxCDD, 13C-1,2,3,6,7,8-HxCDD, 13C-1,2,3,4,6,7,8-HpCDD, 13C-OCDD) and twenty-five 13C12-PCB congeners (13C12-PCB-1, 3, 4, 15, 19, 37, 54, 81, 77, 104, 114, 105, 126, 155, 167, 156, 157, 169, 188, 189, 202, 205, 206, 208, 209) before the ASE extraction procedure. For PCDD/Fs and PCBs analysis, isotope dilution-HRGC/HRMS method based on US EPA method 1613B and 1668A were used with minor modification. The ASE extracts were concentrated by a rotary evaporator and then subjected to a multilayer silica gel column (1 g silica, 4 g basic silica (1.2%, w/w), 1 g silica, 8 g acid silica (30 %, w/w), 2 g silica, and 2 g anhydrous sodium sulfate from bottom up) which was conditioned with 80 mL hexane, the target compounds were eluted with 100 mL hexane. The elution was finally concentrated to about 20 µL in nonane solution. Before instrument analysis, the extract was spiked with injection standards (¹³C12-1,2,3,4-TCDD and ¹³C12-1,2,3,7,8,9-HxCDD) for recovery quantification. PCDD/Fs and PCBs were quantified on Agilent 6890 gas chromatography coupled with an Autospec Ultima high resolution mass spectrometer. The HRMS was operated in selective ion monitoring (SIM) mode at resolution ≥10000. Exactly 1 µL of sample solution was injected with a CTC PAL autosampler in splitless mode into a 60m DB-5MS fused silica capillary column. Helium served as the carrier gas with a constant flow of 1.2 mL min⁻¹. The electron emission energy of the electron impact (EI) ion source was set as 35 eV, and the source temperature was 270°C. The oven temperature programs were employed as follows: for PCDD/Fs, the temperature started at 150°C for 3 min, raised to 230°C and held for 18 min, 230 to 235°C at 5°C min⁻¹ and held for 10 min, final temperature of 320°C was raised at 4°C min⁻¹ and held for 3 min. For PCBs, 120°C was held for 1 min, oven temperature raised to 150°C at 30°C min⁻¹, then raised to 300°C at 2.5°C min⁻¹ and held

for 1 min.

1.3 Quality assurance and quality control

All glassware was thoroughly rinsed with dichloromethane before use. For each batch of the seven samples, one method blank sample was incorporated in the analytical procedure to monitor the possible contamination. The results showed that all targeted analyte concentrations were below the detection limits in the blanks. The detection limits (LODs), defined as three times of signal-to-noise (S/N) ratio, were ranged from $0.193 \text{ pg sample}^{-1}$ (hepta-substituted) to 0.505 pg sample⁻¹ (octa-substituted) for PCDD/Fs and $0.296 \text{ pg sample}^{-1}$ (PCB-167) to 2.91 pg sample^{-1} (PCB-15) for PCBs. Sample recovery values were in the range 25.3% -150% (average 79.9%) for PCDD/Fs and 27.8%-146% (average 85.7%) for PCBs, which accorded with the analytical demand. All target analytes in the ambient air samples were not blank-corrected and the concentrations below the LODs were treated as zero.

2 Results and discussion

2.1 Ambient air concentrations of PCDD/Fs and PCBs

The measured concentrations and toxicity equivalents (TEQs) were summarized in Table 2. International Toxic Equivalents scheme (I-TEF&I-TEQ) was used to evaluate PCDD/Fs and twelve DL-PCB in the ambient air samples. The spatial air concentrations and I-TEQs of PCDD/Fs ranged $1.08 \times 10^{2} - 8.19 \times 10^{3}$ fg m⁻³ (average 2.63×10^{3} fg m⁻³) and 4.08–325 fg I-TEQ m⁻³ (average 91.4 fg I-TEQ m⁻³), respectively. Most of the ambient air concentrations in the sampling sites were lower than the standard of 600 fg TEQ m^{-3} for dioxins proposed by Japan [9]. The atmospheric PCDD/Fs levels in Tianjin City fell in the lower level among the criteria category of global urban areas (100-400 fg TEQ m^{-3}) reported in the literature [10]. The atmospheric PCDD/ Fs levels of point source were also lower than the report of point sources (350-1600 fg TEQ m⁻³) by Fielder [11], and the background site of Jiushan exhibited much lower PCDD/F levels and also fell in the concentration range of global rural and remote area category. All the results showed that an obvious characteristic was existed with point sources > industrial zone > living area > background area in the atmospheric PCDD/Fs level and thus it might imply the feature of point source pollution.

Compared with other Chinese cities including Beijing $(18-644\times10^2 \text{ fg I-TEQ m}^{-3})$ [6], Guangzhou $(57-1.28\times10^3 \text{ fg I-TEQ m}^{-3})$ [5] and Shanghai $(2.2-2.08\times10^4 \text{ fg I-TEQ m}^{-3})$ [12], the atmospheric PCDD/Fs in Tianjin City were in the lower level. Nevertheless, the PCDD/F concentrations were comparable to those in Asian cities including Hong Kong, China $(18-430 \text{ fg I-TEQ m}^{-3})$ [13], Taiwan, China $(56-348 \text{ fg I-TEQ m}^{-3})$ [14], Korea $(169-882 \text{ fg I-TEQ m}^{-3})$

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Table 2	Ambient air co	oncentrations	of PCDD/Fs	and DL	-PCBs	(fg m ⁻³	') during	the sampling	g campaign
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2008 summer ^{a)}	TJ1* ^{b)}	TJ2*	BC #	DL *	TC #	HG**	TG **	XQ #	DG1**	DG2**	JS##
$\sum PCDD/Fs$	2.01×10^{3}	1.68×10^{3}	3.02×10^{3}	2.18×10^{3}	3.62×10^{3}	1.13×10^{3}	6.15×10 ³	3.33×10 ³	1.71×10^{3}	2.23×10^{3}	2.39×10^{2}
$\sum PCBs$	1.69×10^{5}	1.80×10^{5}	3.01×10 ⁵	2.26×10^{5}	1.79×10^{5}	1.08×10^{5}	4.95×10 ⁴	2.48×10^{5}	1.10×10^{5}	1.11×10^{5}	3.08×10^4
I-TEQ _{PCDD/F}	96.0	58.3	201	54.4	124	34.7	68.6	221	98.1	58.8	9.20
I-TEQ _{DL-PCB}	18.7	18.3	25.8	13.9	12.6	9.08	5.96	27.8	13.2	10.9	1.72
2009 winter	TJ1*	TJ2*	BC #	DL *	TC #	HG**	TG **	XQ #	DG1**	DG2**	JS##
\sum PCDD/Fs	2.39×10^{3}	1.97×10^{3}	3.27×10^{3}	2.59×10^{3}	6.50×10^3	1.40×10^{3}	8.19×10 ³	4.36×10^{3}	6.54×10^2	1.49×10^{3}	1.08×10^{2}
$\sum PCBs$	1.34×10^{5}	1.22×10^{5}	2.89×10^{5}	1.31×10 ⁵	2.15×10 ⁵	8.20×10^{4}	8.48×10^{4}	2.01×10^{5}	6.84×10^4	1.11×10^{5}	7.83×10^4
I-TEQ _{PCDD/F}	65.4	135	325	71.2	188	79.1	34.3	380	28.3	95.5	4.08
I-TEQ _{DL-PCB}	30.6	29.8	49.6	33.2	25.9	14.3	14.1	53.3	16.1	23.7	3.14

a) Σ PCBs: Sum of concentration for all the investigated PCB congeners. I-TEQ_{DL-PCB}: Sum of toxicity equivalents for the twelve DL-PCBs in the I-TEQ scheme. b) Sampling area functions. *: living area, **: industrial zone, #: point source, ##: background area.

[15] and Japan (83–550 fg I-TEQ m⁻³) [9], but a little higher than those of the European countries and United States such as Greece (40–119 fg I-TEQ m⁻³) [16], Manchester (26– 220 fg I-TEQ m⁻³) [17], London (62–180 fg I-TEQ m⁻³) [18], Spain (10–357 fg I-TEQ m⁻³) [19], Rome (65 fg I-TEQ m⁻³) [20] and Houston (40–55 fg I-TEQ m⁻³) [21]. Considering the characteristic of Chinese metropolis always with lots of factories and enterprises, these point sources around the city like Tianjin (e.g. lower concentrations of 54.4–135 fg I-TEQ m⁻³ in living area and higher concentrations of 124–325 fg I-TEQ m⁻³ near the point sources) might relevant for the higher atmospheric PCDD/Fs level than the cities of other countries.

With regard to PCBs, the air concentrations of \sum PCBs and I-TEQs of DL-PCBs ranged $3.08 \times 10^4 - 3.01 \times 10^5$ fg m⁻³ (average 1.39×10^5 fg m⁻³) and 1.72 - 49.6 fg I-TEQ m⁻³ (average 18.5 fg I-TEQ m⁻³), respectively. The results showed that the ambient PCB concentration fell in a higher level in all the sampling sites, even for the background area $(3.08 \times 10^4$ fg m⁻³).

Comparing the summer and winter PCDD/Fs atmospheric concentrations, no obvious seasonal trend was observed that atmospheric levels were higher in winter than that of summer the DG1, DG2 and JS sites (Figure 2). It is quite different with other reports that PCDD/F concentrations were usually reported to be higher in winter [10,22,23]. This could be reasonably explained by the observations of Li et al. [7] and Correa et al. [21], where PCDD/Fs prefer to presenting exclusively in the particle phase, while passive air sampling usually underestimate the levels of contaminants in the ambient air because the particulate could not be efficiently trapped by passive sampling techniques. However, it could be successfully used to reflect the levels and distribution patterns of contaminants in the ambient air in a large scale. In the other hand, the northwest monsoon in winter benefits the PCDD/ Fs to transfer to Bohai Sea which is lying in the southeast of the city. It is thought that the distribution of industrial point sources around the city, the monsoon and the location could be one possible reason causing lower atmospheric PCDD/Fs levels in winter. For



Figure 2 Seasonal trends of PCDD/Fs (a) and PCBs (b) at different monitoring sites.

the other sites lying relatively close to Tianjin downtown, the results showed the same trend of higher PCDD/Fs levels in winter.

2.2 Congener profiles in the ambient air samples

The congener profiles in the same sampling sites were expressed in the ways of the relative contribution of individual congener to the total concentrations of PCDD/F congeners



Figure 3 Concentration profiles of individual PCDD/Fs in the ambient air samples.

(Figure 3). The results showed that the concentration profiles were not quite similar for most of the sampling sites. Even for the closer sites of TJ1 and TJ2 sites, 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDF existed in TJ1 site (about 6% of the total concentration) but not detected in TJ2. Various factors, such as sources, meteorology, air mass movement and air degradation process, can affect PCDD/Fs profiles in the atmosphere. The PCDD/Fs profile in this study illustrated that the atmospheric PCDD/Fs might result from distinct generating paths or sources for different sampling sites. Nevertheless, similar to the world average profiles from Europe, the United States, Japan and Australia [10], the spatial ambient air PCDD/Fs concentrations could still be characterized by increased concentrations with the increased chlorination levels when each homologue concentrations were summed together. For example, among the PCDD congeners, 1,2,3,4,6,7,8-HpCDD and OCDD could be detected in most sampling sites, while the most toxic 2,3,7,8TCDD was not found in any samples. However, most PCDF congeners were detected in the air samples.

By comparing the congeners in winter and summer, it could be found that the changes in the concentration profile of individual PCDD/Fs were not the same in the different sites. There were little changes in the sites near or in the downtown like TJ1, TJ2, BC, DL, TC, HG, TG and XQ, but the PCDD/F profiles were quite different in DG1, DG2 and JS, which are far away from the downtown. This implied that the point sources and wind direction might contribute more on the atmospheric levels and congener formation. With regard to PCBs, the analyzed congeners were detected in all air samples, with highest and dominated contribution of congeners PCB-28 accounting for 47%-65% to the total PCB concentrations (Figure 4). In addition, PCB-52 and PCB-101 were also high levels components with the individual contribution proportions of 3.5%-10% and 3.6%-9.9%, respectively.



Figure 4 Concentration profiles of individual DL-PCBs in the ambient air samples.

2.3 Inhalation risk potentials for local residents

Human dioxin exposure pathways mainly consisted of food ingestion, inhalation and dermal contact. Food ingestion is the dominant uptake route and accounts for about 95% of the total daily intakes (TDI) of dioxins [9]. Inhalation is the direct exposure to dioxins and this exposure pathway should be dominated in the uptake of atmospheric pollutants. Atmospheric POPs could also partition into soil and vegetation and could be bioaccumulated in the food chain, resulting in numbers of indirect sources of human exposure [24]. In this study, we assessed the inhalation risk of atmospheric PCDD/Fs to the residents of Tianjin using the following equation [5]:

$$Inh = \frac{V_{\rm r} \times C_{\rm air} \times f_{\rm r} \times t_{\rm f}}{BW}, \qquad (1)$$

where Inh is inhalation exposure for adults/children in fg I-TEQ kg⁻¹ d⁻¹; V_r is ventilation rate (20 m³ d⁻¹ for adults, 7.6 m³ d⁻¹ for children); Cair is the air concentration expressed in fg I-TEQ m⁻³; f_r is the alveolar fraction retained in lung (0.75 for adults/children); tf is the time fraction that adults/children spend at the impact site, which is conservatively assumed as 1; body weight BW is set at 70 kg for adults and 15 kg for children.

The PCDD/F and PCB inhalation risk potentials were summarized in Table 3. It was obviously that the residents lived near the point sources $(37.5-73.1 \text{ fg I-TEQ kg}^{-1} \text{ d}^{-1})$

		PCDD/Fs inhalation (fg I-TEQ kg ⁻¹ d ⁻¹)	PCBs inhalation (fg I-TEQ kg ⁻¹ d ⁻¹)	Total inhalation (fg I-TEQ $kg^{-1} d^{-1}$)
TJ1* ^{a)}	Adult	17.3	5.28	22.6
	Children	30.7	9.36	40.1
TJ2*	Adult	20.7	5.16	25.9
	Children	36.6	9.14	45.7
BC#	Adult	56.4	8.09	64.5
	Children	100	14.3	114
DL*	Adult	13.4	5.04	18.4
	Children	23.9	8.94	32.8
TC#	Adult	33.4	4.13	37.5
	Children	59.2	7.32	66.5
HG**	Adult	12.2	2.50	14.7
	Children	21.6	4.43	26.0
TG**	Adult	11.0	2.15	13.2
	Children	19.6	3.81	23.4
XQ#	Adult	64.4	8.69	73.1
	Children	114	15.4	129
DG1**	Adult	13.5	3.14	16.6
	Children	24.0	5.57	29.6
DG2**	Adult	16.5	3.71	20.2
	Children	29.3	6.57	35.9
JS##	Adult	1.42	0.52	1.94
	Children	2.52	0.92	3.44
Average	Adult	23.7	4.40	28.1
	Children	42.0	7.80	49.8

Table 3 Dioxin inhalation assessment relevant toxicity equivalents (TEQs, fg TEQ kg⁻¹ d⁻¹) of the local residents

a) Sampling area functions. *: living area, **: industrial zone, #: point source, ##: background area.

for adults and 66.5–129 fg I-TEQ kg⁻¹ d⁻¹ for children) suffered more risk than those in living area and industrial zones (13.2–25.9 fg I-TEQ kg⁻¹ d⁻¹ for adults and 23.4–45.7 fg I-TEQ kg⁻¹ d⁻¹ for children), and especially compared with the people lived at the background area (1.94 fg I-TEQ $kg^{-1} d^{-1}$ for adults and 3.44 fg I-TEQ $kg^{-1} d^{-1}$ for children). Obviously, the ambient air concentrations of PCDD/Fs contributed to a large part to the total TEQ (TEQ_{PCDD/F} + TEQ_{PCB}), varying from 72.7% to 89.0% (average 81.8%) despite the relatively higher PCB concentrations. The observed TEQ exposure levels in urban and background areas were comparable with that reported in northern China [23], but were relatively lower than those in other Chinese big cities [5,12]. Nevertheless, the TEQ measured in the industry point sources were several times higher than the background site, still showing the impacts of the importance of anthropogenic activities to the PCDD/Fs and DL-PCBs levels. As the inhalation intake doses were reported to contribute a small part in the total daily intake, e.g. 2.6% of TDI for inhalation intake doses reported in Japan [25], the total daily intake of PCDD/Fs and DL-PCBs congeners might not be ignored, while more researches on other important daily exposure pathway of the PCDD/Fs and DL-PCBs congeners in this area are still needed to clarify the total impact of rapid developing industry point sources to the health influences of the local citizens.

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