Risk assessment of BTEX in the groundwater of Songyuan region of Songhua River in China
Zhuo Zhang, Yanguo Teng, Guanlin Guo, Fasheng Li and Chao Zhang

ABSTRACT

BTEX compounds (benzene, toluene, ethylbenzene and xylenes (m-, p-xylene and o-xylene)) in shallow groundwater were investigated in 16 monitoring wells of the Songyuan region (Songhua River basin). Mean concentrations of benzene, toluene, ethylbenzene, and xylenes were found to be 1.53 μg/L, 1.76 μg/L, 2.11 μg/L and 0.30 μg/L, respectively. Ecological and human health risk assessments were conducted to evaluate potential adverse effects of the BTEX compounds in terms of toxicity. Toluene posed a severe ecological risk, xylenes a moderate risk, while benzene and ethylbenzene were both considered to be a low risk. The mean cancer risk for benzene was estimated to be 1.14 × 10⁻⁶/C0, which is higher than the acceptable risk level of 1.0 × 10⁻⁶. The mean non-carcinogenic risks for benzene, toluene, ethylbenzene, and xylenes were lower than the standard threshold level of 1.0, both individually and cumulatively. In terms of a screening assessment, these results indicate that BTEX in groundwater may pose a threat to the ecosystem and residents in the Songyuan region of the Songhua River basin. This information will be useful for the further evaluation and risk management of groundwater in this area.

Key words | BTEX, ecological risk, groundwater, health risk, Songyuan

INTRODUCTION

China has experienced a rapid urbanization process. In urban areas many pollution-producing industries have been gradually replaced by cleaner industries and removed from the city. Nevertheless, industrial pollution still persists and, in many cases, has been transferred to the groundwater and soil environment, directly or indirectly affecting the health of residents. The Songyuan region was an important petroleum base in China, with many small oilfields being present in the area (for example, the Fuyu, Xinmu and Songyuan Oilfields). In the oil-production area, a considerable amount of petroleum may possibly enter the environment, due to leakage from underground storage tanks, accidental spills, or improper waste disposal practices (Sata & Roy 2011). Most toxic components in petroleum products are typically from the aromatic fraction, i.e. BTEX (benzene, toluene, ethylbenzene and xylene) compounds, with concentrations reaching up to 18% in gasoline (Barbaro et al. 1992; Dinerman et al. 2011). BTEX compounds are classified as hazardous pollutants (US EPA 2003) that have high water solubility and readily migrate with water, resulting in groundwater contamination (Kermanshahi et al. 2005; Victoria & Adebayo 2012). The aqueous solubilities of benzene, toluene, ethylbenzene and xylene are 1780 mg/L, 515 mg/L, 161 mg/L, and 204 mg/L, respectively (López et al. 2008). These BTEX compounds are thus often found in the soil and groundwater (Kao et al. 2006) and present a significant environmental hazard, due to their toxic effects (Kermanshahi et al. 2005; Vidal et al. 2012; Junki et al. 2013). In many countries, groundwater is used as a drinking water source, so the adverse impact of BTEX on human health is of great concern (Farhadian et al. 2008; Xin et al. 2013).

Exposure to BTEX can cause a variety of health problems, such as cancer, respiratory irritation, and damage to the central nervous system. Risk assessments are commonly
used to evaluate the impacts of various hazardous substances on the ecosystem and human health, and to determine the level of treatment required to solve specific environmental problems (Zhang et al. 2012). Because the impacts on human health and ecological risks are simultaneous, quantitative assessments are a necessary requirement for reliable observations on the relationships between sources, pathways and receptors (Guo et al. 2013a). Such methods are also appropriate for the development of suitable management strategies relating to hazardous chemicals (Guo et al. 2013b). In the Songhua River basin the annual groundwater recharge volume to rivers accounts for 5 to 8%, which gives an indication of the importance of groundwater quality to the ecosystem of this river (Huan & Wang 2011). Once the contaminated groundwater reaches the river it tends to become a steady source of pollution into the river. Considering that groundwater is also an extremely important source of drinking water in the Songyuan region, it becomes clear that risk assessments of BTEX in groundwater of the Songyuan River basin is an important aspect of risk assessment. Assessment mechanisms of groundwater contamination have not yet been established in China and, consequently, risk assessments (for the purpose of public health and environmental management) of organic pollution in groundwater, have yet not become a routine task. It is a problem and challenge for the development of routine groundwater risk management systems, in terms of organic pollutants (particularly volatile organic pollutants). Some major obstacles to such a development include the following: the lack of a theoretical foundation to such research; a lack of available data, technical support and appropriate control methods; and a lack of appropriate risk management systems. A large number of case studies are urgently needed to provide technical support to groundwater risk management.

The purpose of this study was as follows: (i) to investigate the concentrations and distribution of BTEX in groundwater of the Songyuan region of the Songhua River basin; and (ii) to assess the potential ecological risk using a risk quotient (RQ) and the human health risk. This will provide a theoretical foundation, as well as technical support, to the groundwater risk management.

MATERIALS AND METHODS

Sampling

The study area is located at latitude of 44°48’N–45°30’N and longitude of 124°33’E–125°38’E (Figure 1). In August 2012,
16 groundwater samples from wells were collected from the upper reaches to the lower reaches of the Songyuan region of the Songhua River basin. Sample collection was based on methods outlined in the Technical Specifications for Environmental Monitoring of Groundwater HJ/T164-2004 (MEPPRC 2004). The 16 wells were located near the typical water quality pollution control section of the Songhua River. Sampling depth ranged from 1.80 to 14.25 m. At each sampling site, 20 mL of sampled water was collected in head-space bottles that contained silicone mats. Water samples were acidified with hydrochloric acid (pH < 2). All samples were immediately shipped to the laboratory, where they were stored at 4 °C in the dark. Samples used for BTEX detection were analysed within 7 days.

**Analysis and quality control**

Pre-treatment and analysis of water samples were performed according to methods recommended by the Ministry of the Environmental Protection of the People’s Republic of China (MEPPRC 2002). BTEX in the water samples was measured by gas chromatography/mass spectrometry.

The pre-treatment of BTEX involved the use of the purge and trap (P&T) technique. The P&T conditions were as follows: blowing temperature: 25 °C; blowing time: 11 minutes; desorption temperature: 180 °C; desorption time: 2 minutes; baking temperature: 200 °C; baking time: 10 minutes; injection volume: 5 mL.

All data were subjected to strict quality control procedures, including the analysis of filed blanks, method blanks and parallel samples. The process used to analyse the blank samples was the same as that used for the water samples during the extraction, purification and analysis procedures. The reagent blank consisted of ultrapure water. The detection limit for BTEX tests was 0.03 μg/L. The limit of detection (LOD) of benzene, toluene, ethylbenzene and xylenes was 0.033 μg/L, 0.031 μg/L, 0.027 μg/L and 0.030 μg/L, respectively. The limits of quantitation (LOQs) of BTEX were in range of 0.09–0.11 μg/L. The recoveries at three spiked levels of 0.16, 1.6, 16 μg/L ranged from 90.5 to 106% with relative standard deviations (RSD, n = 6) of 3.9–5.6%. The linear equations were obtained in the concentration range of 0.16–50 μg/L with correlation coefficients greater than 0. 999.

**Ecological risk characterization**

In the Songhua River basin, groundwater recharge can result in the transfer of pollutants into the river (Huan & Wang 2011), which poses an ecological risk to aquatic organisms in the river. The risk quotient (RQ), as a function of environmental exposure and eco-toxicological effects, is commonly used as a method for expressing ecological risk (Guo et al. 2013b). In this study we used the RQ method to assess the ecological risk of BTEX in groundwater. For each ecological receptor, an RQ was calculated by dividing the predicted, or actual, environmental concentration (PEC) by the toxicity end-point, or the predicted no-effect concentration (PNEC). The RQ of individual BTEX was calculated by means of Equation (1), below.

\[
\text{RQ} = \frac{\text{PEC}}{\text{PNEC}}
\]

where PEC is the actual environmental concentration of individual BTEX, and PNEC is the predicted no-effect concentration of BTEX to aquatic organisms. The PNEC is based on the following toxicity criteria: LC50 (median lethal concentration); EC50 (median effect concentration), and NOEC (no-effect concentration) of BTEX from each fraction, divided by an assessment factor (AF) to account for uncertainty in the chronic value (Equation (2)) (Palma et al. 2004; Guo et al. 2013b; Liu et al. 2013).

\[
\text{PNEC} = \frac{\text{LC50 or EC50 or NOEC}}{\text{AF}}
\]

The value of the AF is determined by the availability and the quality of toxicity data for the chemical of concern. The highest AF of 1,000 (the highest conservation and protective factor) is used for the determination of PNEC for compounds when only one acute toxicity value is available (e.g. algae, crustaceans, or fish); an AF of 100 is used when only one long-term NOEC has been determined from crustaceans or fish; an AF of 50 applies to the lowest of two long-term NOEC assessments, covering two trophic levels; the lowest AF of 10 is applied to chemical compounds for which high-quality chronic toxicity data are available, used at all three trophic levels (e.g. algae, crustaceans, and fish) (European Commission 2003). The proposed assessment factors are presented in Table 1.
There is also an important need to calculate the cancer risk, due to benzene, and the non-carcinogenic risks associated with BTEX.

The cancer risk assessment for benzene is calculated based as follows:

\[ \text{Individual cancer risk} = I \times SF \]  

where \( I \) is the daily intake (mg kg\(^{-1}\) day\(^{-1}\)) and SF is the carcinogen potency factor (kg day mg\(^{-1}\)). The SF is the slope of the dose–response curve at very low exposures. Values of SF, including the oral ingestion slope factor (SF\(_o\)) and the inhalation cancer slope factor (SF\(_i\)), are obtained from the toxicity database in the Technical Guidelines for Risk Assessment of Contaminated Sites (Ministry of Environmental Protection of China (HJ 25.3–2014) (MEPPRC 2014) (Table 2).

The assessment of the toxicity of non-carcinogens is based on the concept of the threshold below which no adverse health effects can be observed. Instead of establishing a threshold value, it is common to use the term ‘reference dose’ (RFD) to represent the level of daily intake of a particular substance that should not produce an adverse health effect (Table 2). Non-carcinogenic risk is characterized in terms of a hazard ratio (HR). The HR calculation for BTEX is based on the following equation:

\[ \text{HR} = \frac{I}{Rfd} \]

where \( I \) is the daily intake (mg kg\(^{-1}\) day\(^{-1}\)) and Rfd is the reference dose (mg kg\(^{-1}\) day\(^{-1}\)) of the individual BTEX compound. Values of Rfd, including the oral ingestion reference dose (Rfdo) and the inhalation reference dose (Rfdi), are

### Table 1 | Assessment factor according the available data\(^a\)

<table>
<thead>
<tr>
<th>Information available</th>
<th>Assessment factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lowest short-term L(E)C50 for algae, crustaceans and fish</td>
<td>1000</td>
</tr>
<tr>
<td>One long-term NOEC for crustaceans or fish</td>
<td>100</td>
</tr>
<tr>
<td>Two long-term NOECs (fish or crustaceans or algae)</td>
<td>50</td>
</tr>
<tr>
<td>Three long-term NOECs from three trophic levels (normally algae, crustaceans, and fish)</td>
<td>10</td>
</tr>
</tbody>
</table>

\(^a\)The data were obtained from the Technical Guidance Document for Risk Assessment of the European Commission (Part II), pages 100–101.

When the value of \( RQ \geq 10 \), a very high risk is expected; values of \( 1 \leq RQ < 10 \) indicate a high risk; values of \( 0.1 \leq RQ < 1 \) indicate a medium risk; values of \( 0.01 \leq RQ < 0.1 \) indicate a low risk, and values of \( \leq 0.01 \) indicate that the risk is negligible or unlikely to occur (Sanchez-Bayo et al. 2002; Backhaus et al. 2003).

### Human health risk characterization

According to the US EPA’s weight-of-evidence classification system for carcinogenicity, benzene is designated as the human carcinogen (Group A), with sufficient evidence of carcinogenicity in humans. In contrast, toluene, ethylbenzene, and xylenes have not been classified as human carcinogens (Group D) due to inadequate, or insufficient, evidence (Ertan et al. 2010). Individual BTEX compounds may present significant health hazards, particularly to local residents. There is a need for the characterization of potential adverse health effects on residents living in a particular neighbourhood. The aim of the human health risk characterization is to quantify potential carcinogenic and non-carcinogenic risk levels of BTEX. Health risk has been characterized by the Risk-Based Corrective Action (RBCA) model specifically designed to complete all calculations required for Tiers 1 and 2 of the American Society for Testing and Materials (ASTM) planning process, as defined in the ASTM E-2081-00 Standard (Guo et al. 2013a). BTEX toxicity due to groundwater exposure can occur through two principal pathways: ingestion after drinking groundwater, and inhalation, due to the volatile nature of BTEX, which may present a significant health hazard. There is also an important need to calculate the cancer risk, due to benzene, and the non-carcinogenic risks associated with BTEX.

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>SF for health risk kg day mg(^{-1})</th>
<th>Rfd for health risk mg kg(^{-1}) day(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Benzene</td>
<td>0.055(^a) 0.031(^a)</td>
<td>0.004(^a) 0.008(^a)</td>
</tr>
<tr>
<td>Toluene</td>
<td><em>b</em> <em>b</em></td>
<td>0.080(^a) 1.276(^a)</td>
</tr>
<tr>
<td>Ethylbenzene</td>
<td><em>b</em> <em>b</em></td>
<td>0.100(^a) 0.255(^a)</td>
</tr>
<tr>
<td>Xylenes</td>
<td><em>b</em> <em>b</em></td>
<td>0.200(^a) 0.026(^a)</td>
</tr>
</tbody>
</table>

\(^a\)Technical Guidelines for Risk Assessment of Contaminated Site of Ministry of Environmental Protection of China.

\(^b\)RBCA default.
obtained from the toxicity database in the Technical Guidelines for Risk Assessment of Contaminated Sites (Ministry of Environmental Protection of China (HJ 25.3-2014) (MEPPRC 2014). Two exposure pathways are considered when determining the intake of contaminants, which include terms for frequency, duration of exposure, and body weight of the receptor. The following generic equation is used to calculate the intake:

\[ I = \frac{C \times IR \times EF \times ED}{BW \times AT} \]  

(5)

where C is the concentration (mg L\(^{-1}\)) of the contaminant; IR is the ingestion rate (L day\(^{-1}\)) defined as the amount of contaminated medium consumed through the exposure pathway per unit of time or event; EF is the exposure frequency (days year\(^{-1}\)); ED is the exposure duration (years) defined as the total time period over which contacts occur between receptor and contaminants; BW is the average body weight (kg) of the receptor over the exposure period; AT is the averaging time (years) of the receptor for carcinogens (ATca) and non-carcinogens (ATnc) (Table 3).

### RESULTS AND DISCUSSION

#### BTEX concentration

The mean concentrations of benzene, toluene, ethylbenzene and xylenes in the groundwater samples were 1.53 μg/L, 1.76 μg/L, 2.11 μg/L, and 0.30 μg/L, respectively (Figure 2). These results show that ethylbenzene is the dominant component of BTEX which, on average, accounts for approximately 37%. The toluene, benzene and xylenes accounted for 31%, 27% and 5%, respectively, on average. The mean concentrations of BTEX in groundwater were

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**Table 3 | The exposure parameters used in the RBCA model**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>ATca average time for carcinogens/yr</td>
<td>Child 72</td>
</tr>
<tr>
<td>ATnc average time for non-carcinogens/yr</td>
<td>6</td>
</tr>
<tr>
<td>BW body weight /kg</td>
<td>15.9</td>
</tr>
<tr>
<td>ED exposure duration/yr</td>
<td>6</td>
</tr>
<tr>
<td>EF exposure frequency/(day yr(^{-1}))</td>
<td>350</td>
</tr>
<tr>
<td>IR ingestion rate of water/(L day(^{-1}))</td>
<td>0.7</td>
</tr>
</tbody>
</table>

**Figure 2 | Distributions of benzene, toluene, ethylbenzene and xylenes in groundwater samples of Songyuan region (μg/L).**
higher than the petrochemical industrial area of Tarragona County in Spain, with 0.758 ng/L, 0.13 ng/L, 0.11 ng/L, and 0.08 ng/L for benzene, toluene, ethylbenzene and xylene, respectively (López et al. 2008). The concentration level of individual BTEX in the groundwater was lower than that of the standard values, including drinking water of China (GB 5794-2006) and USA (US EPA 2012). The mean concentrations of benzene, toluene, ethylbenzene and xyles in groundwater were 6 times, 398 times, 142 times, and 1,666 times, respectively, lower than that of the drinking water standards established in China (GB 5794-2006). It should, however, be noted that BTEX does not generally exist in the natural environment, but there are many small oilfields (e.g. Fuyu and Xinmu Oilfields) in Songyuan region, and the most toxic component of petroleum is typically the aromatic fraction, i.e. BTEX, which has a high water solubility and could thus be readily dissolved and migrate with water, causing a risk of groundwater contamination (Dinerman et al. 2011; Victoria & Adebayo 2012). Such a scenario would result in the detection of BTEX in groundwater.

Ecological risk assessment

In this study, the RQ method was used to assess the ecological risk of BTEX in groundwater. Mean values from monitoring studies were used as the PEC. No ecological screening for BTEX has taken place in surface water or groundwater in China, and the toxicity data mentioned in this paper come from citations (OGP 2002). PENC and PEC values are presented in Table 4.

The mean ecological risk quotients of benzene, toluene, ethylbenzene and xyles in the groundwater were 0.09, 3.59, 0.02 and 0.25, respectively (Figure 3). The RQ (toluene) > 1 indicated that the toluene posed a severe risk; the 0.1 < RQ (xyses) < 1 indicated that the xyles was of moderate risk. Risk quotients (of 0.01 < RQ (benzene) < 0.1 and 0.01 < RQ (ethylbenzene) < 0.1) indicated that the benzene and ethylbenzene were considered as low-level ecological risks. The mean RQ of BTEX was 3.95 and the BTEX in groundwater showed a high level of ecological risk. These data indicate that adverse ecological risks have already been detected in the groundwater of the Songyuan region. In the Songhua River basin, recharge of groundwater to river runoff accounts for 5 to 8% each year, so the groundwater containing BTEX as a steady source could flow into river water and pose an ecological risk to aquatic organisms in the river. This indicates that the ecological risk assessment of groundwater is an important aspect of risk management in the Songhua River basin.

<table>
<thead>
<tr>
<th>Compound</th>
<th>Endpoint</th>
<th>Trophic level</th>
<th>Toxic effect levela, μg/L</th>
<th>Assessment factora</th>
<th>PENC, μg/L</th>
<th>PEC, μg/L</th>
<th>RQb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Benzene</td>
<td>NOEC (20d)</td>
<td>Crustacea (male)</td>
<td>170</td>
<td>10</td>
<td>17</td>
<td>1.53</td>
<td>0.09</td>
</tr>
<tr>
<td>Toluene</td>
<td>LC50(96 h)</td>
<td>Crustacea (male)</td>
<td>490</td>
<td>1000</td>
<td>0.49</td>
<td>1.76</td>
<td>3.59</td>
</tr>
<tr>
<td>Ethylbenzene</td>
<td>NOEC (21d)</td>
<td>Crustacea (male)</td>
<td>1000</td>
<td>10</td>
<td>100</td>
<td>2.11</td>
<td>0.02</td>
</tr>
<tr>
<td>Xyles</td>
<td>LC50(96 h)</td>
<td>Fish (female)</td>
<td>1200</td>
<td>1000</td>
<td>1.2</td>
<td>0.3</td>
<td>0.25</td>
</tr>
</tbody>
</table>

The data were obtained from those reported in OGP (2002), pages 13–14.

Risk calculated using mean concentration as PEC value.

Health risk assessment

Forward health risk assessment considers the likely carcinogenic effects of each contaminant for the most sensitive receptors in the residential scenario. In this study, the risks were characterized directly for benzene, toluene, ethylbenzene and xyles. Based upon toxicity data, benzene represents the greatest health threat within the BTEX present, because it is a known carcinogen (NRC 1983; US EPA 1986). The mean benzene-related cancer risk was predicted to be 1.14 × 10⁻⁶. The maximum carcinogenic risk for benzene was 2.01 × 10⁻⁶. For known or suspected carcinogens, 1.0 × 10⁻⁴ and 1.0 × 10⁻⁶ are the generally acceptable lifetime health risk for the cumulative level and the individual level, respectively (Guo et al. 2013a). According to this setting, the individual average cancer risk for benzene in groundwater in Songyuan...
region was acceptable. However, the cancer risk predicted to be less than or equal to $1 \times 10^{-6}$ is usually considered negligible (Guo et al. 2015b). On the other hand, the risk predicted to be less than or equal to $1 \times 10^{-3}$ was defined as a significant risk by the US Supreme Court in 1980 (Rodrics et al. 1987). In our study, we found a benzene-related carcinogen risk at 63% of sample sites exceeded $1 \times 10^{-6}$. Hence, the benzene-related cancer risk in the groundwater is not negligible, but this does not pose a significant risk. Cancer risk is primarily due to direct exposure, with ingestion contributing 70% of exposure, and inhalation being responsible for the remaining 30%.

To evaluate the non-carcinogenic effects of the chemicals, it is common to consider the risk to be negligible if the non-carcinogenic risk is predicted to be less than or equal to 1.0. The individual average non-carcinogenic risks predicted for BTEX in the groundwater were negligible because all of the non-carcinogenic risks are less than 1.0 (Figure 4). The maximum non-carcinogenic risks of benzene, toluene, ethylbenzene and xylenes were predicted to be 0.029, 0.0019, 0.0025 and 0.0002, respectively. The maximum non-carcinogenic risks for BTEX were still much less than 1.0, thereby indicating that they did not pose a risk of adverse health effects.

In the case of exposure to two or more substances that may target different organs in humans, the total cumulative lifetime non-carcinogenic risks should be considered as additive. The cumulative non-carcinogenic risk of BTEX was calculated to be 0.018, so the risk was negligible. Although the concentration of ethylbenzene is higher than that of benzene and toluene, the lifetime non-carcinogenic risk for benzene accounted for about 90% of the total lifetime non-carcinogenic risk in the groundwater. This is mainly due to the low value of RfD for benzene. Non-carcinogenic risk is primarily due to direct exposure, with ingestion contributing 99.98% of exposure and inhalation responsible for the remaining 0.02%. López et al. (2008) reported that direct ingestion of BTEX was the most significant factor (around 85%) for adults and children.

**Uncertainty analysis**

The influence of various factors can result in uncertainties associated with risk assessment. Toxicity data on aquatic organisms used in ecological risk assessment were cited by the US EPA, rather than the sensitive species of China. Certain parameters (for example, the amount of daily consumption of water, body surface area, etc.) of the US EPA are used in the RBCA model. This however needs further confirmation, regarding whether the parameters are suitable for Chinese people. The calculated risks only became available after obtaining results of sampling and analysis, but the pollutant concentration may vary at different periods and the attenuation of BTEX was not considered. These factors may lead to uncertainties for risk assessment.
CONCLUSION

In this study we attempted to determine two potential risks due to BTEX contamination: the ecological risks to aquatic organisms in the Songhua River induced by the contaminated groundwater, and the human health risk to residents who drink groundwater in the Songyuan region. The ecological risk assessment indicated that the mean RQ of BTEX was 3.95 and that the BTEX in groundwater posed a high-level ecological risk. In terms of human health, the mean cancer risk was estimated to be $1.14 \times 10^{-6}$, which is higher than the generally acceptable risk of $1.0 \times 10^{-6}$. Non-carcinogenic health risks of BTEX were lower than the specified level of 1.0 and were thus regarded as acceptable. These results indicate that BTEX in groundwater poses a health threat to residents of the Songyuan region of Songhua River.

The main advantages of the risk assessment approach used in this study, are that they are generally simple, quick, and inexpensive, and that such an assessment can serve as a screening assessment of BTEX in groundwater, in terms of uncertainties. This has an important practical significance for risk management and decision making associated with the treatment of groundwater in polluted areas. Results of this assessment highlighted the necessity to strengthen BTEX monitoring and conduct further research into groundwater evaluation and risk assessment, to ensure the health and safety of residents, and the security of water ecosystems of the Songhua River basin.

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