DRINKING WATER QUALITY IN DISTRIBUTION SYSTEMS OF SURFACE AND GROUND WATERWORKS IN FINLAND

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Abstract. Physico-chemical and microbiological water quality in the drinking water distribution systems (DWDSs) of five waterworks in Finland with different raw water sources and treatment processes was explored. Water quality was monitored during four seasons with on-line equipment and bulk water samples were analysed in laboratory. Seasonal changes in the water quality were more evident in DWDSs of surface waterworks compared to the ground waterworks and artificially recharging ground waterworks (AGR). Between seasons, temperature changed significantly in every system but pH and EC changed only in one AGR system. Seasonal change was seen also in the absorbance values of all systems. The concentration of microbially available phosphorus (MAP, $\mu g PO_4$ -P/l) was the highest in drinking water originating from the waterworks supplying groundwater. Total assimilable organic carbon (AOC, $\mu g AOC$ -C/l) concentrations were significantly different between the DWDSs other than between the two AGR systems. This study reports differences in the water quality between surface and ground waterworks using a wide set of parameters commonly used for monitoring. The results confirm that every distribution system is unique, and the water quality is affected by environmental factors, raw water source, treatment methods and disinfection.

Keywords: drinking water, on-line monitoring, raw water source, seasonal change.

Introduction

Reliable water supply and drinking water distribution are easily taken for granted. However, even in the most advanced societies, drinking water related outbreaks are frequently reported (Zacheus, Miettinen 2011; World..., 2014). As an effort to guarantee the water security in all circumstances, comprehensive risk management is becoming an important part of the everyday operation of waterworks all over the world. Potential threats to the health of water consumers have to be mitigated and therefore more extensive drinking water quality monitoring is also needed (Ikonen *et al.*, 2017).

The drinking water production chain can be divided into three main parts, which all play an important role in the safety of drinking water: the quality of the raw water source, the water purification processes and the drinking water distribution systems (DWDSs). All these parts of the production chain affect the physico-chemical characteristics of the distributed water, such as water temperature and nutrient content (Clark *et al.*, 1982; Geldreich, 1989). Subsequently, these characteristics have substantial impact on water quality, either by supporting or hindering the regrowth of micro-organisms in the bulk water, deposits and biofilms. Many microbes survive in the drinking water distribution systems and when environmental conditions are favourable, they start to multiply and thus deteriorate the water quality (Power, Nagy, 1999).

In developed societies, the concern regarding the water security of DWDSs is rapidly growing with aging pipelines. In the decision making processes, the lack of understanding the importance of a properly working water system may lead to the lack of investments on the infrastructure (Ainsworth, 2004; OECD, 2013). Furthermore, technical properties of DWDSs such as the occurrence of dead-ends in the pipeline, piping materials, diameter of pipelines have influence on the water quality (Inkinen *et al.*, 2014; LeChevallier *et al.*, 1996). Thus, the site-specific DWDS properties have to be considered when planning successful monitoring schemes.

In Nordic regions, clear differences between the four seasons of the year affect the surface water quality and groundwater hydrology (Kløve *et al.*, 2017). For example, snow melts in spring and heavy rains in autumn may deteriorate the raw water quality. The changes are most evident in the temperature of raw water but the season may affect also other quality parameters of the distributed water.

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Large share of Finnish population (90%) are served by centralized DWDSs. The rest of the population have their own private drinking water wells (Keinänen-Toivola *et al.*, 2007). Legislation of the European Union (EU) sets demands for the drinking water quality monitoring and reporting requirements in Finland. Of 154 large EUregulated waterworks in Finland, 41% used groundwater and 19% artificially recharged groundwater in their drinking water production in 2014. The rest of waterworks (40 %) were dependent on surface water sources (Zacheus, 2013). Artificially recharged groundwater is equalized with natural groundwater aquifers in legislation.

The aim of this study was to explore physicochemical and microbiological drinking water quality in a variety of Finnish waterworks supplying either nondisinfected water or using disinfection to ensure the water safety.

The water quality differences were explored between the selected surface waterworks, ground waterworks and artificially recharging ground waterworks. A wide variety of basic laboratory water quality measurements and on-line monitoring was used to detect changes in the drinking water quality. A specific objective was to explore potential seasonal changes in the water quality by performing the monitoring in winter, spring, summer and autumn within one calendar year. Further, the relationships between the monitored water quality parameters were investigated.

Material and Methods

Water sampling and on-line monitoring

In this study, dynamic changes in the quality of distributed drinking water at five DWDSs in Finland were monitored (Systems A-E; Table 1). Water samples were analysed and the on-line monitoring data were collected to capture the potential differences in the water quality between the different water treatment units, the seasons

Table 1. Characteristics of the DWDSs

of the year and the distances from the waterworks (water age). Waterworks were selected for this study according to their raw water source and the disinfection method with the aim to cover the main water treatment processes that are used in the drinking waterworks in Finland.

Sampling points were mainly in public buildings within the same water supply area, except in the system E where points were nearby water mains. Distances between the waterworks and the sampling points within the DWDSs varied from one to 36 kilometers representing the water age from two to 50 hours. The collected water quality data reported herein includes DWDSs data from two waterworks using artificial groundwater, from two surface waterworks and from one groundwater supply (Table 1). The disinfection methods in the studied DWDSs included non-disinfection in two systems and combinations of UV-light, ClO₂, Cl, NH₂Cl and NaOCl in other systems. Water sampling was carried out during winter (January-February), spring (March-May), summer (August-September) and autumn (October-December). Sampling points in each DWDSs included three cold water sampling points, which were located at different distances from the waterworks. In each system, one monitoring period lasted for two weeks and three visits to the sampling points were made during that time. Water samples were taken during the second and third visit. Sterile, plastic bottles were used for microbiological analyses and hydrochloric acid washed glass bottles for the analysis of microbial nutrients and physicochemical analyses. Water quality parameters including residual chlorine (if chlorine disinfection was in use), pH, temperature, and electric conductivity (EC) were determined in situ at the sampling locations at the minimum during the first and the third visit. Water samples were transported to the laboratory in cool-boxes and stored at 3 ± 2 °C and analysed as soon as possible, preferably within 24h from the sampling.

DWDSs	Raw water source	Production volume, m ³ /day	Treatment methods	Disinfection
A	Artificial groundwater	6 300	Aeration, lime stabilization, floc- culation, clarification, addition of sulphuric acid, sand filtration	No disinfection
В	Artificial groundwater	2 300	Aeration, lime stabilization, floc- culation, clarification, sand filtra- tion	No disinfection
C	Surface water	40 000	Ferric sulfate coagulation, flota- tion, sand filtration, activated carbon filtration	UV-light, ClO ₂ , Cl
D	Surface water	126 000	Ferric sulfate coagulation, clarifi- cation, sand filtration, ozonisa- tion, activated carbon filtration	UV-light, NH ₂ Cl
Е	Groundwater	3 000	Aeration, limestone filtration	UV-light, NaOCl

Physico-chemical parameters

Turbidity (NTU) was measured spectrophotometrically at the wavelength of 860 nm with Turb 555IR spectropho-

tometer (WTW GmbH&Co. KG, Weilheim, Germany). Absorbance and UV-absorbance were assayed at wavelengths of 420 nm (Abs 420 nm) and 254 nm (UV-Abs 254 nm), respectively (Shimadzu UV-1601, Shimadzu Co., Kyoto, Japan). pH and electric conductivity (EC) were assayed in field and in laboratory using Multi 3430i meter (WTW GmbH&Co. KG, Weilheim, Germany). For chlorine, Palintest chlorometer (Palintest Ltd., England) was used. Both free and total chlorine were measured from cold water samples. Metal analyses included the measurements of aluminium (Al), copper (Cu), iron (Fe), and manganese (Mn) which were determined by using HACH Lange DR2800 spectrophotometer (Hach Lange GmbH, Düsseldorf, Germany) according to the manufacturer's instructions (methods 8506, 8008, 8012, 8149).

Microbiological parameters

Heterotrophic plate count (HPC) was used to enumerate heterotrophic bacteria, yeasts and moulds by spread-plate technique on Reasoner's 2 Agar (R2A) medium (Difco, Detroit, MI, USA) and incubated at 22 ± 2 °C for 7 days (Reasoner, Geldreich, 1985).

Samples for analyzing total microbial count (TMC) were preserved by adding 37% formaldehyde to sample to have final concentration of 2%. Formaldehyde was filtered through 0.22-µm-pore-size filter before use. DAPI (4.6-diamidino-2-phenylindole dihydrochloride) (Merck, Darmstadt, Germany) staining (Porter, Feig, 1980) and Olympus BX51TF epifluorescence microscope (Olympus BX-51TF, Olympus Co., Japan) was used to analyse the total microbial counts.

Virus like particles (VLP) were analysed as described earlier (Noble, Fuhrman, 1998; Noble, 2001; Rinta-Kanto *et al.*, 2004). Briefly, 1.6 ml of drinking water was filtered on 0.02-µm-pore-size aluminium oxide filter (Anodisc 25, Whatman Ltd., Whatman, Kent, UK). VLPs collected on the filters were stained with SYBR green I nucleic acid stain (Molecular Probes, Life Technologies, California, USA) and enumerated using epifluorescent microscope (Olympus BX-51TF, Olympus Co., Japan, filter cube U-N41001AO).

Microbially available nutrients

Assimilable organic carbon (AOC) was determined by using sodium acetate as a model substrate for pure cultures of *Pseudomonas fluorescens* P17 (strain biotype 7.2 (ATCC 49642) and Aquaspirillum NOX (ATCC 49643) (Van Der Kooij et al., 1982) and by using the method of APHA (1992) to standardize the growth of Pseudomonas fluorescens P17 and sodium oxalate to standardize the growth of Aquaspirillum NOX. Results are shown as organic substance of acetate carbon (µg acetate-C/l) (Miettinen et al., 1997a) and oxalate and acetate carbon in all (total-AOC, µg/l; acetate-C, µg/l) (Miettinen et al., 1999). In brief, the idea is to create growth conditions in which the organic carbon is the only limiting growth factor and the content of AOC is determined based on maximum colony forming units of Pseudomonas fluorescens and Aquaspirillum NOX after incubation of 7, 8 and 9 days.

The assay of microbially available phosphorus (MAP) is based on a method developed by (Lehtola *et al.*, 1999). MAP bioassay is based on inorganic phosphate. In

brief, idea is to create conditions in which phosphorus becomes the only limiting nutrient. The content of MAP is determined by maximum colony forming units of *Pseudomonas fluorescens* P17 strain biotype 7.2 (ATCC 49642). Sensitivity of the method is 80 ng PO₄-P/l (Miettinen *et al.*, 1999). *Pseudomonas fluorescens* P17 growth is standardized by disodium hydrogen-phosphate (Na₂HPO₄) as a substrate. The equivalent amount of phosphorus is calculated based on the known yield coefficients as phosphate to phosphorus (μ g PO₄-P/l) (Lehtola *et al.*, 1999).

Online measurements

In order to follow real-time water quality changes in the DWDS, EC, DO (dissolved oxygen), ORP (oxidationreduction potential), pH, temperature and turbidity were measured on-line. YSI 6920 V2 (# 6920V2-M, YSI Inc., Ohio, USA) instrument was installed to the sampling point 2 and YSI Professional Plus instrument (# 605596, YSI Inc., Ohio, USA) to sampling point 3, which represented the oldest water. For a two weeks monitoring period at each season YSI sensors were connected to a tap and water flow was set at maximum 3 l/min. The collection frequency of the measured data was in every five minutes, except during winter sampling period when the data was collected once per minute for YSI 6920 V2.

Statistical analysis

The statistical analyses were conducted using IBM SPSS Statistics 21 (IBM Corporation, USA). The difference between DWDSs sampling points and seasons was explored using Kruskal-Wallis test. Spearman correlation analysis was used to explore relationships between the measured water quality parameters. The statistical analyses were performed to the whole dataset and separately to each DWDS.

Results

Differences in water quality between DWDSs

In order to study differences between the DWDSs, test was performed to the whole dataset of cold water samples ($N_{total} = 120$). The test showed significant differences between the five DWDSs (p < 0.05) in nearly all water quality parameters.

Average values of total-AOC (μ g/l AOC-C) were higher in water produced from surface water (C and D) than in the water produced from artificial groundwater (A and B) or groundwater (E) (Fig. 1a). Such a difference was not seen in the average acetate equivalent AOCcarbon content (μ g/l) (Fig. 1b).

Concentrations of the analysed metals (Al, Cu, Fe, Mn) were very low and the major part of the results were under the detection limit.

The significant coefficient values of Spearman correlation found between quality parameters are presented in Table 2. It was found that total-AOC and acetate-C had a strong relationship in waters of the waterworks A, B, D and E. Significant relation was found also between Abs 420 nm and iron in the waterworks A, C and D and between free chlorine and total chlorine in the waterworks C and D. In the ground waterworks E, EC exhibited strong correlation with UV-Abs 254 nm, acetate-C and total-AOC. pH and total chlorine, as well as pH and EC correlated in DWDS D (Table 2).



Fig. 1. Microbially available nutrient concentrations (min, average, max) measured as: a) assimilable organic carbon, Total-AOC as μg of AOC-C/l; b) AOC as μg of acetate C/l; c) microbially available phosphorus, MAP as μg of PO₄-P/l in the studied DWDS.

Differences in water quality within each DWDS

Differences in the water quality parameter values between the sampling points 1, 2 and 3 within each system were investigated. In the DWDS A significant differences between the sampling points were found in the Abs 420 nm (p = 0.011) and HPC values (p = 0.016). The lowest absorbance value was always observed at the point 1 and this value (min = 0.006 AU; average = 0.009 AU; max: 0.011 AU) differed from the point 2 (min = 0.009 AU, average = 0.012 AU, max = 0.016 AU; p = 0.036) and point 3 (min = 0.010 AU; average = 0.013 AU; max = 0.019 AU; p = 0.003). In HPC measurement the point 3 of DWDS A was different than the other two points (p =0.008 - 0.028) and had the highest value ($2.0*10^2 1.4*10^4$ CFU/mI) during sampling rounds except once. In the DWDS B, HPC values were significantly different between the points 1 and 2 (p = 0.006) and 1 and 3 (p = 0.003). The HPC in the point 1 of DWDS B ranged between $9.0*10^1 - 2.6*10^2$ CFU/ml and was always lower than the counts from points 2 and 3 ($1.6*10^2 - 7.0*10^2$ CFU/ml).

Table 2. Spearman correlation coefficients (*R*) larger than 0.7 between the water quality parameters in each DWDS (level of significance p < 0.05).

DWDS	Parameter 1	Parameter 2
A, B, D, E	Total-AOC, µg/l	Acetate-C, µg/l
A, C, D	Abs 420 nm	Iron, mg/l**
С	Abs 420 nm	Total chlorine, mg/l
Е	EC field, µS/cm	UV-Abs 254 nm
Е	EC field, µS/cm	Acetate-C
Е	EC field, µS/cm	Total-AOC
C, D	Free chlorine	Total chlorine, mg/l
В	Iron, mg/l*	UV-Abs 254 nm
С	Iron, mg/l**	Total chlorine, mg/l
D	Temperature, °C	VLP
D	pH field	Total chlorine, mg/l
D	pH field	EC field, μ S/cm

Absorbance – Abs 420 nm; Acetate-C – acetate carbon; EC – electric conductivity; Total-AOC – total assimilable organic carbon; UV-Abs 254 nm – UV-absorbance at wavelength 254 nm; VLP – virus like particles.

* Two measurements under the detection limit.

** Five measurements under the detection limit (Total N = 24).

In the DWDS C, the Abs 420 nm value as well as iron concentration were the highest in sampling point 3. Further, also water turbidity was significantly higher at the point 3 (min = 0.01; average = 0.06; max = 0.1; p =(0.001) as compared to the point 1 (min = 0.01; average = 0.02; max= 0.07; p < 0.001). Free and total chlorine concentrations in the DWDS C were significantly lower at the point 3 (min = 0.03; average = 0.09; max = 0.32) compared to the points 1 (min = 0.27; average = 0.39; max = 0.68; p = 0.008) and 2 (min = 0.2; average = 0.33; max = 0.45; p = 0.002). In the DWDS D, no significant differences were detected in the water quality parameter values between the sampling points and in the DWDS E, the chlorine concentration was the only parameter in which significant change between the sampling points was detected. The free chlorine concentration was lower at the point 2 than at the points 1 and 3 (p = 0.021 - 0.021) 0.038). Total chlorine concentration was lower at the point 2 than at the point 3 (p = 0.02).

Seasonal changes in water quality

Season affected significantly to the measured absorbance values within each system (points 1, 2 and 3 at DWDS A-E). Figures 2 and 3 show the average values of UV-Abs 254 nm and Abs 420 nm, respectively, at the studied systems in different seasons.



Fig. 2. Seasonal values of UV-absorbance (254 nm) in DWDSs.



Fig. 3. Seasonal values of absorbance (420 nm) in DWDSs.

In the DWDS A, Abs 420 nm was significantly higher in summer than in spring and winter (p = 0.010-0.048). In winter and spring, the UV-Abs 254 nm values were significantly lower than in summer and autumn (p < 0.05). In the DWDS B, Abs 420 nm was significantly lower in winter than in spring and summer (p < 0.05). In the DWDS C, the UV-Abs 254 nm in winter and spring was significantly different than in summer and autumn (p = 0.001). In the system D, the values of UV-Abs at 254nm were significantly different between winter and spring (p = 0.001) and between winter and summer (p = 0.003). In the system E, the UV- UV-Abs 254 nm values in water at autumn were significantly different than in summer (p = 0.003). In the system E, the UV- UV-Abs 254 nm values in water at autumn were significantly different than in summer (p = 0.003).

Total-AOC concentrations were significantly lower in surface waterworks during autumn than in other seasons (p < 0.05; DWDS C and D). Significant changes in acetate-C concentration between the seasons were observed in systems B-E (p < 0.05) but not in the system A (p = 0.303).

In the DWDSs of surface waterworks, HPC counts and MAP concentrations were highest in the summer and significantly changed between the seasons. HPC in DWDS C was significantly different between winter and spring and winter and autumn (p < 0.05). MAP concentrations were significantly different in spring than in other seasons (Fig. 1c; p < 0.05). HPC in the system D was higher in summer than in winter and autumn (p < 0.05). In the DWDS D, MAP concentrations were higher in summer than in winter (p = 0.001) and spring (p = 0.005). However, the exceptionally high value of MAP (4.8 µg/l PO₄-P) was measured in DWDS E (Fig. 1c).

On-line water quality in DWDSs

On-line monitoring of water quality in the studied DWDSs was conducted during two weeks measurement

periods in four seasons. Water temperatures were higher in summer than in other seasons except in the sampling point 2 of DWDS A, where the highest water temperatures was reached in autumn (Fig. 4). The average water temperature measured on-line was in winter 7.4 $^{\circ}$ C (January-March), spring 6.0 $^{\circ}$ C (March-May), summer 14.0 $^{\circ}$ C (July-September) and autumn 11.2 $^{\circ}$ C (October-December).



Fig. 4. Average temperatures (°C) measured on-line during two weeks monitoring period in four seasons of a year.

The monitored EC values at DWDS A and B were on average higher (177–241 μ S/cm) than in DWDSs of surface waterworks C and D (124–158 μ S/cm) (Fig. 5). DWDS E (ground waterworks with sodium hypochlorite disinfection) had the lowest average EC value (102 μ S/cm). DWDS E also deviated from the other systems with highest average ORP value (674 mV) compared to others systems (244–433 mV). A part of the

ORP measurements were unsuccessful (Fig. 6). Average pH value varied between 8.1–9.0 in all studied DWDSs (Fig. 7). Average DO values varied between 9.1–12.0 mg/l. The on-line measurement of turbidity suffered from excessive baseline variation possibly due to air bubbles in the measurement channel and the turbidity data collection was unsuccessful.



Fig. 5. Average EC (μ S/cm) values in on-line monitoring for two weeks period in different seasons.



Fig. 6. Average ORP (mV) values in on-line monitoring for two weeks period in different seasons.



Fig. 7. Average pH values in on-line monitoring for two weeks period in the four seasons of a year.

Discussion

It is well known that environmental factors affect the drinking water quality (Clark *et al.*, 1982, Geldreich, 1989). The treated drinking water entering the distribution system contains physical, microbial and nutrient loads as particles, cells and inorganic and organic substances (Liu *et al.*, 2013). The two main processes that threaten tap water quality are particle accumulation (Vreeburg, Boxall, 2007) and microbial growth (Van Der Kooij *et al.*, 1982). The seasonal changes can also have effect on the quality of drinking water especially when there are problems in water treatment or in DWDS.

In this study, raw water source and disinfection were the main differences between the systems. Differences in the water quality were found also within the DWDSs. This study indicates that decrease in the disinfectant residual levels in conjunction with growing distance from the waterworks enables regrowth and explains the increase in the HPC in the distribution network. Chlorine concentration mainly decreased significantly when the water flowed further away from the waterworks where either chlorine dioxide and chlorine or sodium hypochlorite was used. However, the change was not found from the sampling points of the system where chloramine was used. It is known that chloramine is more stable in water than sodium hypochlorite (Gould et al., 1984; Yoon, Jensen, 1993). Further, when comparing different types of DWDSs in this study, it was observed that HPC was a parameter that changed significantly between sampling points when the disinfection was not used (DWDS A and B). Our results are in accordance with the findings of Lahti et al., (1993) who found variations in HPC within each DWDS. In the distribution systems of surface waterworks (DWDS C and D) HPC changed significantly between the seasons, which was most likely caused by higher temperature of the distributed water during the summer than in groundwater systems.

Waterworks using groundwater or surface water as raw water source could be differentiated by the water quality parameters such as absorbance, electric conductivity and MAP concentrations.

MAP results were shown to change significantly between seasons in surface waterworks and they were highest in the summer time. In Finland, phosphorus is considered limiting nutrient more than organic carbon (Miettinen et al., 1997b). Significant variations in concentrations of total AOC-carbon was noticed between the DWDSs (average AOC values 72-149 µg/l). However, acetate-C concentrations did not change significantly between the systems (average values 58-83 µg acetate-C/l). It has to be remembered that there are variations in the concentrations of AOC in different distribution systems and seasons due to operational changes in the water treatment (Liu et al. 2002). However, in many waterworks there is lack of information about concentration of AOC, since in legislation, only the measurement of total organic carbon (TOC) is required.

In previous studies it has been shown that chemical coagulation and activated carbon filtration treatment low-

ered the amount of organic matter in drinking water (Lehtola *et al.*, 2002). However, chlorination and ozonation is known for their effect to increase the amount of AOC in water. UV-disinfection is known not to have effects on MAP concentrations but it might decrease AOC concentration. Furthermore, the addition of lime in pH adjustment is known to increase the MAP concentrations in water (Lehtola *et al.*, 2002). Therefore, the treatment methods employed in the studied waterworks might explain the detected variability in the amount of total AOC (μ g AOC-C/I) and high MAP concentrations in the system E.

In DWDS E, temperature was lowest in the all measurement periods compared to other studied DWDSs. This might be due to the location of the sampling points nearby the water mains instead of the true building taps. Growing distance from the water supply did not seem to affect water temperature except in the surface water systems C and D in autumn, where water temperature increased when distance increased. In the studied systems, only few significant changes like Abs 420 nm, free and total chlorine, HPC, iron and turbidity were conducted.

In this study, the chlorinated groundwater (DWDS E) exhibited the lowest EC values and highest ORP and MAP values. EC value was highest in the systems A and B, where water origin was artificially recharged groundwater. DWDS E differentiated of A and B by applying chlorination, but the exact reason behind the difference in the EC values remains unknown. The raw water source seems to play role in this, as the EC values in two surface waterworks were very close to each other.

Recent studies show that variables such as raw water source, temperature, pipeline materials, nutrients availability and disinfection methods affect into the microbial dynamics inside the distribution networks (Scheili *et al.*, 2015; Delafont *et al.*, 2016). Comparison between the systems in present study confirmed that each DWDS is unique. Therefore it is important to monitor and know the water quality and the changes within each system. Based on the series of measurement values it is possible to observe changes in DWDSs.

Conclusions

- 1. Differences in water quality were observed between surface waterworks and ground waterworks.
- 2. The season of the year affected HPC and MAP concentrations in surface waterworks.
- 3. Seasonal changes in water temperature and absorbance were observed.
- 4. Distance from the waterworks (water age) was found to affect the water quality parameters.
- 5. Simple measurements can be used to obtain information about the water quality changes in distribution systems.
- 6. In the future, comprehensive risk management could rely more than before on on-line water quality measurements.

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