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# Overview of the findings from measurements of halogenated compounds at Gosan (Jeju Island, Korea) quantifying emissions in East Asia

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With increased economic growth in East Asia, regional emissions of many anthropogenic halogenated compounds now constitute a substantial fraction of the global totals. Here, we summarize recently reported findings from measurements of a wide range of chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), sulfur hexafluoride ($SF_6$), and other halogenated compounds at Gosan (Jeju Island, Korea) within the advanced global atmospheric gases experiment (AGAGE). General wind patterns at Gosan bring air masses from the surrounding areas, allowing the monitoring of both clean baseline and polluted air masses. We have analyzed our measurements since November 2007 both with an interspecies correlation method and with an inversion method based on the FLEXPART Lagrangian particle dispersion model to estimate these regional emissions. The results show that emissions of halogenated compounds in East Asia account for over 20% of global emissions, both in terms of ozone depletion potential (ODP) and global warming potential (GWP), and emphasize the importance of atmospheric measurements for quantifying emissions of these compounds in this region.

**Keywords:** halogenated compounds; Gosan; East Asia; emissions; greenhouse gases; ozone depleting substances



## 1. Introduction

Anthropogenic halogenated compounds (including chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride ($SF_6$)) are characterized by their long atmospheric lifetimes, during which they act as precursors to the destruction of stratospheric ozone and/or as powerful greenhouse gases (GHGs, see Table 1). The Montreal Protocol (MP; http://ozone.unep.org/new_site/en/index.php) has been instrumental in phasing out the use of halogenated compounds especially dangerous with respect to stratospheric ozone depletion (Prinn et al. 2000; Forster et al. 2007; Velders et al. 2007; Kim et al. 2011), while the emissions of compounds with high global warming potentials (GWPs) not included in the MP, such as the HFCs, PFCs, and $SF_6$, are now regulated under the Kyoto Protocol of the United Nations Framework

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Table 1. Important halogenated compounds (Forster et al. 2007).

| Compound name | Chemical formula | Lifetime (years) | Ozone-depleting potential (ODP) | Global warming potential (100 year GWP) |
|---|---|---|---|---|
| *Chlorofluorocarbons (CFCs)* | | | | |
| CFC-11 | $CCl_3F$ | 45 | 1.0 | 4,750 |
| CFC-12 | $CCl_2F_2$ | 100 | 1.0 | 10,900 |
| CFC-113 | $CCl_2FCClF_2$ | 85 | 0.8 | 6,130 |
| CFC-114 | $CClF_2CClF_2$ | 300 | 1.0 | 10,000 |
| CFC-115 | $CClF_2CF_3$ | 1,700 | 0.6 | 7,370 |
| *Hydrochlorofluorocarbons (HCFCs)* | | | | |
| HCFC-22 | $CHClF_2$ | 12 | 0.055 | 1,810 |
| HCFC-141b | $CH_3CCl_2F$ | 9.3 | 0.11 | 725 |
| HCFC-142b | $CH_3CClF_2$ | 17.9 | 0.065 | 2,310 |
| *Hydrofluorocarbons (HFCs)* | | | | |
| HFC-134a | $CH_2FCF_3$ | 14 | | 1,430 |
| HFC-152a | $CH_3CHF_2$ | 1.4 | | 124 |
| HFC-23 | $CHF_3$ | 270 | | 14,800 |
| HFC-32 | $CH_2F_2$ | 4.9 | | 675 |
| HFC-125 | $CHF_2CF_3$ | 29 | | 3,500 |
| HFC-143a | $CH_3CF_3$ | 52 | | 4,470 |
| *Perfluorinated compounds (PFCs)* | | | | |
| PFC-14 | $CF_4$ | 50,000 | | 7,390 |
| PFC-116 | $C_2F_6$ | 10,000 | | 12,200 |
| PFC-218 | $C_3F_8$ | 2,600 | | 8,830 |
| Sulfur hexafluoride | $SF_6$ | 3,200 | | 22,800 |



Convention on Climate Change (UNFCCC, http://unfccc.int/kyoto_protocol/items/2830.php). Emissions of some of these compounds are being reduced in developing countries through the clean development mechanism of the UNFCCC (http://cdm.unfccc.int).

Understanding the emissions of anthropogenic halogenated compounds in East Asia has been limited, since the developing countries in this region do not report their GHGs emissions to the UNFCCC. Previous studies of these compounds based on measurements at global background sites speculated on the importance of emissions from East Asia (Montzka et al. 2009; Levin et al. 2010; Mühle et al. 2010; Rigby et al. 2010). However, direct measurements to properly quantify the emissions of these compounds in East Asia have been relatively sparse (Palmer et al. 2003; Yokouchi et al. 2006; Guo et al. 2009).

Here, we summarize the findings from the measurements of halogenated compounds performed at Gosan, located on Jeju Island, Republic of Korea (South; here referred to as Korea), which are important for quantifying the emissions of these species in East Asia. A brief description of the analytical methodology used at the station is followed by a discussion of the results derived from our measurements, as reported in recent literature (Kim et al. 2010; Stohl et al. 2010; Li et al. 2011).

## 2. Methods

### *2.1. Measurements of halogenated compounds at Gosan (Jeju Island, Korea)*

Gosan station is located on the south-western tip of Jeju Island, south of the Korean peninsula (126°9′E, 33°17′N, 72 m asl) on a remote hill-top by the coast. As such

effects from local contamination can be assumed to be minimal, which allows for monitoring of long-range transport from the surrounding region. The wind patterns at Gosan are typical of the Asian Monsoon pattern, with strong northern winds in winter, and southern influence during summer (Figure 1(a)). These wind patterns are favorable for monitoring air masses passing through East Asia, especially China and Korea. Air masses passing through Japan and Taiwan are relatively less frequent and constrained to spring and fall. Clean "baseline" conditions are observed when a clean stream of air flows in directly from northern Siberia (in winter, Figure 1(b)) and during transport of southerly oceanic winds (in summer).

The measurement system at Gosan is comprised of an in situ GC-MSD system for measuring ~40 halogenated compounds, based on an advanced cryofocusing technique ("Medusa", see Miller et al. 2008) and operated under the Advanced Global Atmospheric Gases Experiment (AGAGE) (Prinn et al. 2000). The measurements are reported using calibration scales developed by the Scripps Institution of Oceanography (UC San Diego) under AGAGE. Typical precisions of the measurements are better than 1% of background atmospheric concentrations. Operation of the Gosan Medusa system began in November 2007.



### 2.2. *Estimating emissions of halogenated compounds in East Asia*

Emissions of halogenated compounds have been calculated using two methods, namely, a ratio method based on interspecies correlation between measured compounds (Palmer et al. 2003; Yokouchi et al. 2005), and an inverse modeling method (Stohl et al. 2009, 2010) based on a Lagrangian particle dispersion model (FLEXPART; Stohl et al. 2005).

For the ratio method (Yokouchi et al. 2006; Kim et al. 2010; Li et al. 2011), a suitable tracer from which the emission ratios are calculated was chosen from analysis of interspecies correlation matrix tables. Analysis of air mass back-trajectories was performed in conjunction to separate periods of clear influence from China, Korea, Japan, and Taiwan in the Gosan measurements. Analysis showed that HCFC-22 was the most suitable tracer for emissions from China, Korea, and Taiwan, while HFC-134a was found to be better suited for emissions from Japan. The emission rates of the tracer compounds were derived from the FLEXPART inversions, explained below.

Our inversion method is based on footprint emission sensitivities for Gosan calculated using the Lagrangian particle dispersion model FLEXPART (Stohl et al. 2005), the Gosan measurement data and a priori emissions information. The method merges all data sets to improve the emissions compared to the a priori information (see Stohl et al. 2009 for details). Most of the available a priori emissions were estimated from so-called "bottom-up" methods (statistically derived from reported consumption data), and are prone to large uncertainties due to the complexity of properly tallying the emissions from limited information. This is particularly true for East Asia, where information is especially sparse. For developed countries like Japan, the national inventory reports to the UNFCCC (http://unfccc.int/national_reports/annex_i_ghg_inventories/items/2715.php) can be used. For developing countries, we can use emissions reported in the Emissions Database for Global Atmospheric Research (EDGAR; EC-JRC/PBL 2011), as well as national scale emissions reported in the literature (Tsai 2006; Wan et al. 2009). Details of our implementation are available in Stohl et al. (2010).

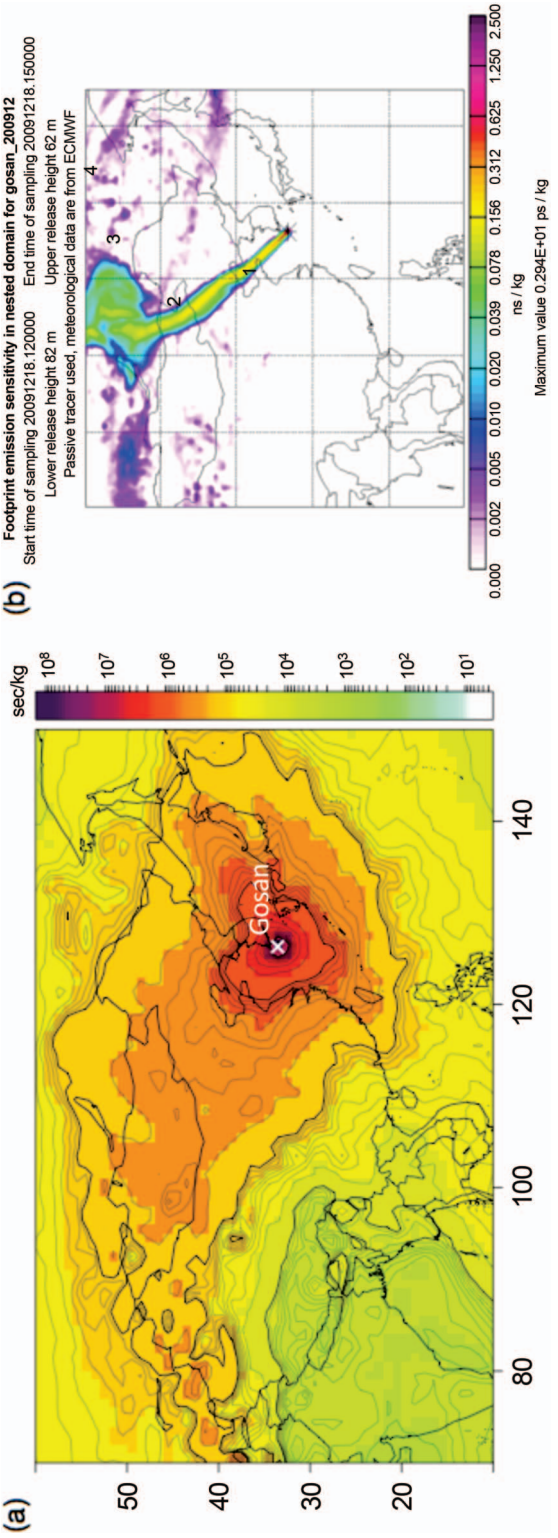


Figure 1. (a) Location of Gosan (Jeju Island, Korea), shown with footprint emission sensitivities calculated using FLEXPART. Larger sensitivities are analogous to greater residence times of air masses arriving at Gosan. (b) Typical airmass back-trajectory during background events.

The ratio method provides a simple yet comprehensive method for estimating the emissions of almost all halogenated compounds measured at Gosan and minimizes uncertainties inherent in more complex modeling schemes. However, it is restricted by core assumptions in the method such as co-located emission sources of the tracer and target compounds and an accurate knowledge of the tracer emissions. The interspecies ratios we observe at Gosan show statistically significant correlation for many compounds at national scales (see Li et al. 2011), suggesting that overall these core assumptions are robust. The inversion method provides a more objective means for estimating emissions including their spatial distribution, which is not available from the ratio method. However, this method requires a relatively large measurement data set in order to provide substantial improvements of the a priori emission information. As such, both methods are complementary in nature, and help provide a balanced view of the emissions in East Asia.



## 3. Results and discussions

### *3.1. Variability of halogenated compounds at Gosan*

The results of the measurements of halogenated compounds at Gosan can be characterized by frequent pollution events superimposed on the Northern Hemispheric baseline concentrations in the colder months (late fall through early spring) and lower concentrations in summer from the Southern Hemispheric influence brought in by the Asian Monsoon. The seasonal patterns are more striking in compounds with a large interhemispheric gradient in concentrations, caused by relatively shorter lifetimes and/or large emissions in the Northern Hemisphere, such as HCFC-22 and HFC-152a. Similar seasonal patterns have been found at nearby stations in Japan (Yokouchi et al. 2006; Saito et al. 2010).

Baseline conditions during the colder months are typically correlated with air masses arriving from North Asia (Siberia) through a narrow pathway in the Yellow Sea (see Figure 1(b)). Concentrations during these baseline conditions agree well with the baseline concentrations at other background sites in the Northern Hemisphere, such as Mace Head (Ireland) and Trinidad Head (California, United States). These baseline events are crucial for defining the magnitude of pollution events in our analysis.

Among the most polluted species measured at Gosan, both in terms of frequency of pollution events as well as magnitude of pollution over baseline conditions, are HCFC-22, $CF_4$, and HFC-23 (Figure 2), which are further discussed below.

HCFC-22 has been the dominant interim-replacement for CFCs in refrigerant applications (Velders et al. 2009; Wan et al. 2009; also see http://www.afeas.org). And while consumption of HCFC-22 has effectively been phased out in most developed countries and replaced with HFC blends, it continues to be widely used in developing countries such as China and Korea (Montzka et al. 2009). As such, large pollution events of HCFC-22 are observed in air masses both from China and Korea, and to a lesser extent from Japan and Taiwan.

$CF_4$ is another compound for which large emissions are observed throughout the region. However, the major sources for its emissions are not uniformly distributed, namely, the primary aluminum smelting industry for China and the electronics industry (semiconductors and flat panel displays) in Korea, Japan, and Taiwan (Mühle et al. 2010; EC-JRC/PBL 2011).

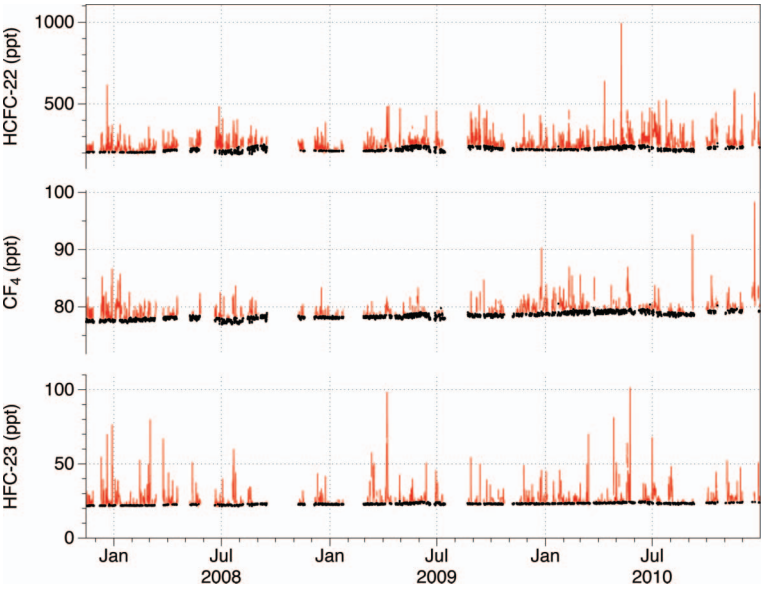


Figure 2. Gosan measurement results for HCFC-22, $CF_4$, and HFC-23. Baseline events (calculated using the AGAGE statistical algorithm) are shown in black, while the pollution events are shown in the red.

On the other hand, pollution events of HFC-23 are seen mostly in air masses from China. HFC-23 is emitted almost exclusively during the production of HCFC-22 (Montzka et al. 2010) and the pollution events seen at Gosan reflect the dominance of China's production in the global totals (Miller et al. 2010).

Further discussions of the measurement results are available in Kim et al. (2010) and Li et al. (2011).

### *3.2. Emissions of halogenated compounds in East Asia*

Our results confirm that emissions of halogenated compounds in East Asia are a significant portion of the global total emissions. Aggregated together, the East Asian emissions reach ~20% in terms of both ozone depletion potential (ODP) and GWP for 2008 (Li et al. 2011). For individual compounds, the large fraction of HCFC emissions found in East Asia (29% for HCFC-22, 32% for HCFC-141b, and 26% for HCFC-142b) are in line with the dominant consumption of these species in this region. The large fraction of HFC-23 emissions (78% of global totals) is also in line with the dominant production of HCFC-22 in China, as noted above. On the other hand, the large emissions of compounds previously thought to be consumed dominantly in developed countries such as HFC-32 (129%, exceeding previous global total estimates) and HFC-125 (20% of global totals), used in refrigerant blends, and HFC-152a (24% of global totals, used in foam blowing) are surprising, and require further research into their actual sources. A complete list of emission rates is available in Li et al. (2011).

The MP is shown to have a substantial impact on emissions in East Asia. The MP mandates the phase out of CFCs for HFCs (with no ozone depleting potential), and

HCFCs (with less ozone depleting potential than CFCs) are allowed as interim substitutes (for details, see http://ozone.unep.org/new_site/en/Treaties/control_measures_summary.php). Relative emissions among MP-related compounds (CFCs, HCFCs, HFCs) from China and Korea (Figure 3(a, b)), both Article 5 (“Developing”) countries under the MP, are dominated by HCFCs, supporting the successful phase-out of CFCs from these countries. Relative emissions in Japan (Figure 3(c)), a non-Article 5 (“Developed”) country under the MP, show somewhat smaller proportions of HCFCs together with larger proportions of HFCs, in agreement with HFCs becoming the dominantly used species in developed countries. The emissions of HCFCs in Japan are likely from older equipment that continue to be used (e.g. old refrigerators and air conditioners), and not from new equipment. Taiwan (Figure 3(d)), while not included in the MP, is shown to have voluntarily phased out CFCs, and emissions from this country are currently heavily based in HCFCs. Our findings regarding the impact of the MP are in good agreement with other measurement-based (Vollmer et al. 2009) and bottom-up based (Wan et al. 2009) emission studies for China (Table 2).

While the total emissions of halogenated compounds are largest in China, per-capita emissions (Figure 4) are significantly larger in Korea than in China, particularly for the HCFCs. This shows that emissions in Korea, while relatively low quantitatively, are still a cause for concern. In addition, if China’s per-capita emissions were to reach Korean levels, as China follows Korea’s path in economic development, the increase in Chinese emissions would have a significant impact on the global budgets of these compounds. As such, continued monitoring of emissions is essential for this region.

The spatial emission patterns derived using inverse methods show great promise in applying this technique to locate emission hot-spots within East Asia. The unique emission characteristics of HFC-23 (emitted almost exclusively in the limited number of HCFC-22 production sites) are ideal for testing these capabilities, and the inversion results were well-correlated with many of the known locations of the HCFC-22 manufacturing plants in China and Japan (Stohl et al. 2010). Further

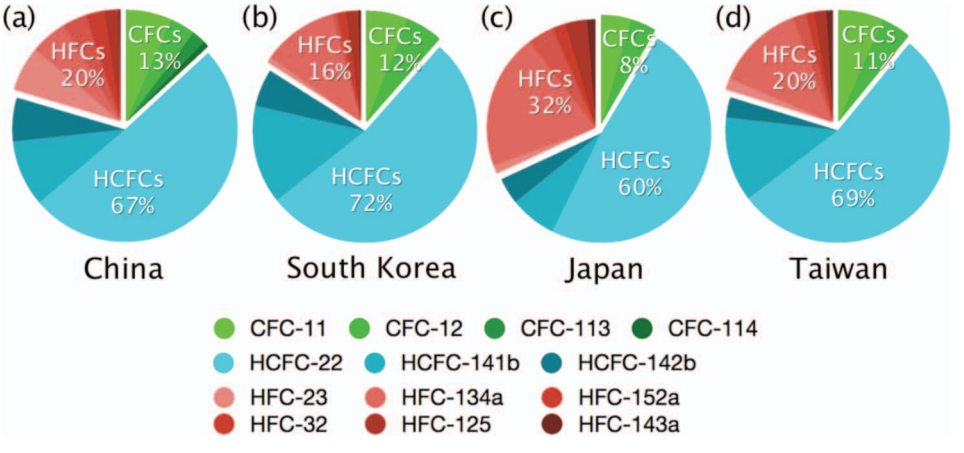


Figure 3. Relative emissions of CFCs, HCFCs, and HFCs in China, South Korea, Japan and Taiwan, for 2008.

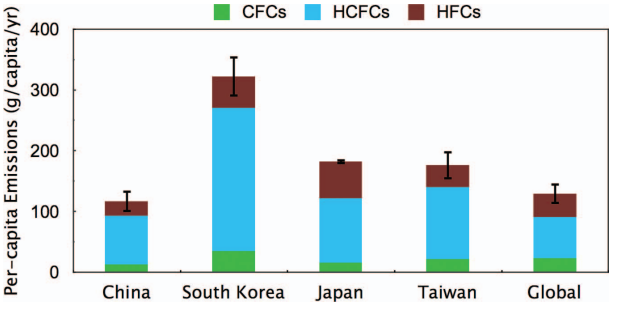


Figure 4. Per-capita emissions of CFCs, HCFCs, and HFCs in China, South Korea, Japan and Taiwan, for 2008.

Table 2. Emissions of CFCs, HCFCs, and HFCs reported for China, 2008.

| | Li et al. (2011) | Vollmer et al. (2009) | Wan et al. (2009) |
|---|---|---|---|
| CFC-11 | 11 (9.7–17) | 33 (26–43) | 14.259 |
| CFC-12 | 6.1 (4.4–8.5) | 14 (9–19) | 3.869 |
| CFC-113 | 3.2 (0.8–5.7) | 0.8 (0.4–1.7) | 0 |
| CFC-114 | 1.3 (0.8–1.7) | | |
| HCFC-22 | 83 (64–109) | 165 (140–213) | 79.268 |
| HCFC-141b | 15 (11–21) | | 12.148 |
| HCFC-142b | 9 (6.9–12.7) | 12 (10–18) | |
| HFC-23 | 10 (8.7–15) | | |
| HFC-134a | 8.3 (6.2–11.4) | | |
| HFC-152a | 5.4 (4.0–7.4) | | |
| HFC-32 | 4 (2.9–5.6) | | |
| HFC-125 | 3.1 (2.3–4.3) | | |
| HFC-143a | 0.6 (0.39–0.79) | | |

work is under way to increase the inversion resolution from the current $1^\circ \times 1^\circ$, and to apply the inversion technique to other gases, many of which are emitted only from specific static sources (e.g. $CF_4$ and $C_2F_6$ from aluminum smelting and semiconductor manufacture).

## 4. Concluding remarks

Overall, the significant emissions found for almost all of the halogenated compounds reaffirm the significance of East Asian emissions to the global budgets of these species. The importance of East Asian emissions will only grow as industrial development continues throughout this region, especially in China. The measurements at Gosan have proved to be useful for defining the current state of emissions in this region, which serves as a baseline for future changes.

Currently, work is under way to further understand the emission source characteristics of halogenated compounds in East Asia. Analysis techniques such as Positive Matrix Factorization are being used to discern emission patterns reflecting the major sources of emissions in this region. For compounds such as the PFCs, emitted both from the aluminum smelting industries and the semiconductor

industries (including the manufacture of microprocessors, memory, flat panel displays, and photovoltaics), reducing the uncertainties in the industry-specific emissions in East Asia could significantly improve the global imbalance between ''top-down'' (measurement derived) and ''bottom-up'' (industry-reported) emissions (Mühle et al. 2010). These efforts will be combined with extending the regional emissions estimates to recent years.

In conclusion, emissions derived ''top-down'' are crucial to validate ''bottom-up'' emissions and to ultimately reach the emission reduction goals set forth by the international community (Nisbet and Weiss 2010). The measurements at Gosan and also in other parts of East Asia, e.g. at Hateruma and Ochi-ishi in Japan (Yokouchi et al. 2006; Saito et al. 2010) and at Shandanzi in China (Vollmer et al. 2009), will provide the data needed to assess the emissions of the halogenated species, both now and in the future.



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