Statistical Comparison of Regional-Scale Tropospheric Aerosol Extinction Coefficient across China Based on CALIPSO Data

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ABSTRACT

Two years of CALIPSO level 2 version 2 data were analyzed to obtain regional distributions of vertical profiles of the aerosol extinction coefficient (AEC) across China. 10 typical geographical regions were selected for comparison, which cover various aerosol pollution levels, climate zones, and underlying surfaces. The whole troposphere was split into two-layers: the Lower- and the Upper-layer—separated by the maximum boundary layer height—which represent the boundary layer and the upper tropospheric layer, respectively. The annual average of the column-average AEC in the 10 regions ranged from 0.066 to 0.243 km⁻¹ in the Lower-layer and from 0.022 to 0.059 km⁻¹ in the Upper-layer. The regional AEC in the Lower-layer was the highest in central and eastern China, followed by that in the Sichuan Basin, the Yunnan-Guizhou Plateau, the capital economic circle, the Pearl River Delta, the desert region, the Tibetan Plateau, northeast China, the northwest semi-arid plateau, and, finally, the East China Sea. The seasonal AEC in the Lower-layer was the highest during spring in the desert and marine regions, during the summer on the Tibetan Plateau, and during autumn or winter in the other regions. The regional and seasonal patterns of AEC in the Lower-layer agreed to a large extent with known regional distributions of surface-layer PM2.5 distributions and dominant seasonal emission sources in their respective regions. Regional and seasonal patterns in the Upper-layer were slightly different from those in the Lower-layer due to different transport pathways of aerosol pollution in different regions. The proportion of occurrence under different pollution conditions and the number of polluted days were also estimated separately for the Lower- and the Upper-layer, based on AEC vertical profiles for each region.

Keywords: Tropospheric aerosol; Aerosol pollution; Aerosol column average; Occurrence frequency.

INTRODUCTION

Knowledge of spatial and temporal distributions of tropospheric aerosols is needed in order to address their impacts on air quality and climate related issues (Menon et al., 2002; Lelieveld et al., 2002; Watson, 2002, Li, et al., 2011; Ma et al., 2014; Sheng et al., 2016). The globally increasing atmospheric aerosol loading has added large uncertainties in regional and global climate models used for addressing aerosol-cloud integration and aerosol climate effects due to the difficulties in quantifying aerosol spatial distributions and their radiative properties (Knippertz et al., 2015; Seinfeld et al., 2016). Aerosol optical properties measured by satellites can fill some of the knowledge gaps (Winker et al., 2007; Martin et al., 2008). For example, data collected by the Moderate Resolution Imaging Spectroradiometer (MODIS) have been used to provide column-integrated aerosol optical properties covering regional to global scales (Kim et al., 2007; Kosmopoulos et al., 2008; Zhang et al., 2010) and estimate ground-level PM2.5 and PM10 in various regions of the world (van Donkelaar et al., 2006; Zhang et al., 2010; He et al., 2012; Zheng et al., 2013).

Compared to MODIS data, the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) provide continuous aerosol profiling covering multiple vertical layers globally, which provides an unprecedented opportunity for advancing the understanding of aerosol distribution characteristics on large-scales (Winker et al., 2007, 2010). Example applications of CALIPSO data include investigating seasonal variations of aerosol vertical distribution globally (Yu et al., 2010; Huang et al., 2013), analyzing annual and seasonal patterns of the aerosol extinction coefficient (AEC) over 13 sub-continental regions representative of industrial, dust, and biomass burning pollution sources (Koffi et al., 2012), identifying an aerosol layer at the tropopause level.
associated with the Asian monsoon (Vernier et al., 2011), and providing vertical profiles of aerosols and AEC in various regions (Campbell et al., 2013; Ma et al., 2014).

China is among the most polluted regions in the world due to its economy booming in the past three decades (Zhang et al., 2012; Han et al., 2014; Sheng et al., 2016). Measurements of aerosol chemical and optical properties were mostly made at limited surface stations (Yu et al., 2013; Sun et al., 2015; Tao et al., 2017). Several studies have used remote sensing data for quantifying aerosol distributions across China (He et al., 2012; Zheng et al., 2013; Sun and Chen, 2017; Xiao et al., 2017; Yu et al., 2017). However, little effort has been made to investigate the vertical distribution characteristics of atmospheric aerosols. The present study aims to characterize regional distribution patterns of atmospheric aerosols at both lower- and upper-layers of the troposphere across China through analyzing aerosol AEC data provided by the CALIPSO Lidar observations. Knowledge gained in this study can be used for improving climate models and for making future emission control policies.

METHODOLOGY

Selected Regions

Based on geographical location and dominant underlying surface type, 10 regions (R1–R10) in China were defined and selected for the comparison of aerosol extinction coefficient profiles and probability distributions. Fig. 1 shows the locations of the 10 regions, and Table SI-1 in the Supplementary Information (SI) document lists their boundary latitude and longitude values. R1 covers most areas of the Taklimakan desert area; R2 is inside the northwest part of the semi-arid plateau covering part of Gobi sand and Loess Plateau areas; R3 is the Beijing-Tianjin-Hebei region (the capital economic circle); R4 covers most of the northeast China, a major agricultural zone with plenty of plains and forests; R5 covers most of the Tibetan Plateau; R6 covers the whole Sichuan Basin and surrounding areas; R7 is in the middle part of central and eastern China, with dense population, and covers the Yangtze River Delta region; R8 mainly covers the Yunnan-Guizhou Plateau; R9 is in the southern coastal area of China and covers the Pearl River Delta economic region as well as Fujian Province; and R10 is inside the East Sea of China. These regions cover different economic belts, climate zones, topography, and pollution levels in China.

Satellite Data

The Cloud-Aerosol Lidar with a Orthogonal Polarization (CALIOP) instrument installed on the CALIPSO satellite measures vertical profiles of elastic backscatter at 532 and 1064 nm near nadir during both day and night phases of the orbit. The backscatter coefficient data were converted to aerosol and cloud properties, including the aerosol extinction coefficient (AEC) and aerosol optical depth (AOD). High spatial resolution continuous measurements of vertical profiles of aerosol and cloud optical properties globally have been available since mid-June of 2006 (Winker et al., 2007, 2009). AEC data at the 532 nm wavelength from June 2006 till September 2008 were analyzed in the present study. Level 2 aerosol profile data at 5 km horizontal resolution covering the 10 regions of China defined above were extracted for all the vertical levels. Table SI-2 in the SI document provides the numbers of samples of the AECs during the four seasons in the 10 regions.

Data Analysis

To explore the contribution of AEC in different value ranges to the average value of AEC in any given region and season, the whole AEC range (0.0–1.25 km⁻¹) was divided into 10 equal intervals, i.e., the first interval is from 0 to 0.125 km⁻¹; the second one, from 0.125 to 0.250 km⁻¹;
and so on; and the tenth one, from 1.125 to 1.25 km$^{-1}$. The first interval of AEC should represent the cleanest condition and the last one, the most heavily polluted condition in terms of aerosol concentration. Based on China’s Air Quality Index (AQI) (HJ633-2012), aerosol pollution can be roughly categorized into three levels: clean, light-polluted, and heavy-polluted, corresponding to AQIs of < 100, 100–200, and > 200, or PM$_{2.5}$ concentrations of < 75, 75–150, and > 150 µg m$^{-3}$, respectively. AEC values corresponding to these PM$_{2.5}$ concentration cutting points will vary with aerosol chemical composition, among other factors. Based on values found in Tao et al. (2015) for different pollution conditions in Beijing, we can assume the first and second AEC intervals as clean conditions, the next four intervals as light-polluted conditions, and the last four intervals as heavy-polluted conditions.

For statistical comparison between the 10 different regions, column-integrated values, rather than data at selected heights, were used for analysis to minimize the uncertainties inherited from the measurement. Considering that aerosol concentration varies greatly with height and so does the AEC value, AEC data were compared separately for two layers: One is from 60 m high to the top of the boundary layer height (BLH) (referred to as the Lower-layer), and the other is from BLH to 8 km (referred to as the Upper-layer). AEC data at a height of 0–60 m (the lowest layer detected by the satellite) were excluded from Lower-layer analysis due to the large uncertainties caused by surface reflectivity, while data from 8 km high were excluded from Upper-layer analysis due to the contributions of cirrus clouds to AEC. Although the AEC data were supposed to have removed the contributions from clouds, the impact of cirrus clouds to AEC could not be excluded completely by the CALIOP instrument due to the very thin layers of such clouds and their weak contributions to AEC (Omar et al., 2013). Note that the Lower-layer represents the whole boundary layer, while the Upper-layer represents the upper troposphere.

BLH changes with time and location, and average BLH can be determined from the vertical profiles of AEC, i.e., at the height where the maximum variance of the AEC appears (Stull and Eloranta, 1984; Menut et al., 1999; Lammert et al., 2006; Emeis et al., 2008; Luo et al., 2014a). CALIPSO provides lidar data for retrieving BLH globally (Jordan et al., 2010; McGrath-Spangler and Denning, 2013; Zhang et al., 2016). The present study, however, developed a method for extracting the maximum BLH for any given region (R1–R10) and during any given season (spring [March–May], summer [June–August], autumn [September–November], and winter [December–February]) using the approach described below. In the first step, the probability of each AEC interval appearing at every height was calculated as the ratio of the number of AECs in the selected AEC interval and height to the total number of AECs for all AEC intervals within the whole column on a seasonal basis. In the second step, vertical profiles of AEC probability were plotted for all the AEC intervals, and BLH for each AEC interval was identified based on the maximum variance of the AEC probability, as mentioned above (Fig. SI-1). A linear regression between BLH and AEC interval was then plotted, and the interception of BLH (with AEC = 0) was defined as the maximum BLH (see a linear regression for R1 and R2 in spring in Fig. SI-2(a) as an example). The seasonal maximum BLH was used to separate Lower-layer and Upper-layer defined above for AEC analysis. The seasonal maximum BLH was found to be as low as 0.5 km in region R10 and as high as 3 km in R5 and R8, which was consistent with their respective underlying surfaces or terrains (e.g., ocean in R10 and plateau in R5 and R8).

**RESULTS**

**Overview of AEC Vertical Profiles**

The annual average AEC decreased from the surface with increasing height and reached the minimum values at an altitude around 8–9 km. AEC at the surface ranged from 0.05 to 0.32 km$^{-1}$, depending on the region, and ranged from 0.012 to 0.022 km$^{-1}$ at an altitude of 8–9 km (Fig. 2). The vertical variabilities of the annual average AEC within the Lower-layer were in the range of 0.127–0.060, 0.082–0.051, 0.269–0.084, 0.095–0.062, 0.356–0.025, 0.288–0.173, 0.286–0.170, 0.323–0.094, 0.194–0.072, and 0.071–0.045 km$^{-1}$ for regions R1 to R10, respectively. The vertical variabilities of the annual average AEC within the Upper-layer were in the range of 0.104–0.013, 0.069–0.012, 0.081–0.017, 0.070–0.015, 0.039–0.010, 0.165–0.013, 0.163–0.021, 0.091–0.021, 0.130–0.023, and 0.070–0.020 km$^{-1}$ for regions R1 to R10, respectively. Regional differences in the probability profiles associated with different pollution levels (clean, light-polluted, and heavily-polluted) are very large (Fig. 3), with the maximum probability appearing at different heights in different regions, likely dominated by different aerosol sources and BLH values. Seasonal differences in the average AEC profiles were significant in some regions (e.g., R1, R3, R4, R8), especially in the Lower-layer (Fig. SI-3). Considering the large differences in AEC between the Lower-layer and Upper-layer, the column-averaged AEC and probability are discussed separately below on an annual and seasonal basis. All values mentioned in the following sections refer to regional- as well as column-averaged values unless specified differently.

**Regional AEC in the Lower-layer**

**Annual AEC**

The annual average AEC for the Lower-layer showed regional differences up to a factor of 3.7 (ranging from 0.066 to 0.243 km$^{-1}$) and in the sequence of R7, R6, R8, R3, R9, R1, R5, R4, R2, and R10 from the largest to the smallest. The lowest AEC in R10, among all the regions, was apparently due to the minimum aerosol loading over marine surfaces. The highest AECs were mostly found in industrial areas with high population densities, such as the Yangtze River delta (R7), Sichuan Basin (R6), capital economic circle (R3), and Pearl River Delta (R9). Surface-level PM$_{2.5}$ concentrations in these regions were consistently higher than in most of the other regions. For example, Tao et al. (2017) showed the annual PM$_{2.5}$ from available measurements to be 115 µg m$^{-3}$ in the capital economic circle,
Fig. 2. Vertical profiles of annual average extinction coefficient (AEC) in the 10 regions based on the CALIPSO level 2 lidar data.

Fig. 3. Annual average vertical profiles of probability of occurrence for clean, light-polluted, and heavy-polluted conditions in the 10 regions.

100 $\mu$g m$^{-3}$ in the Sichuan Basin, 148.9 $\mu$g m$^{-3}$ in central and eastern China, and 76.8 $\mu$g m$^{-3}$ in the Pearl River Delta economic region. It is noted that the Yunnan-Guizhou Plateau (R8) was also among the regions with the highest AEC; however, no PM$_{2.5}$ measurements were available in this region for a direct comparison with the other regions. PM$_{2.5}$ levels were suspected to be at high levels due to the establishment of a large number of coal-mining factories since the 1990s (Zheng et al., 2010). Decreasing trends in sunshine duration from the 1990s till 2005 have been observed in this as well as other regions of China, likely due to the increased aerosol loading (Kaiser et al., 2002; Zheng et al., 2008). It is further noted that BLH in R8 was higher than in the other regions (Fig. SI-2), and the maximum AEC probability for polluted conditions reached 2 km in this region while being below 1.5 km in most of the other regions (Fig. SI-1). This is likely another reason for the high AEC in R8 in the Lower-layer. The regional distribution of AEC presented above was mostly consistent with that of aerosol optical depth based on the 10 years of MODIS remote sensing data (Luo et al., 2014b).

The proportion of occurrence (POC) in the 10 regions was in the range of 48.1–92.9%, 5.5–42.1% and 1.3–9.8% for clean, light-polluted and heavy-polluted conditions, respectively, on an annual basis (Table 2). POC was calculated for the whole Lower-layer (or Upper-layer) by...
Table 2. The probability of occurrence (POC) for clean, light polluted and heavy polluted conditions in the Lower-layer and Upper-layer over the 10 regions. POC was calculated for the whole Lower-layer (or Upper-layer) by adding all the cases together covering various altitudes on seasonal and annual basis.

<table>
<thead>
<tr>
<th>Region</th>
<th>Lower-layer</th>
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<th>Annual</th>
<th>Spring</th>
<th>Summer</th>
<th>Autumn</th>
<th>Winter</th>
<th>Light polluted</th>
<th>Annual</th>
<th>Spring</th>
<th>Summer</th>
<th>Autumn</th>
<th>Winter</th>
<th>Heavy polluted</th>
<th>Annual</th>
<th>Spring</th>
<th>Summer</th>
<th>Autumn</th>
<th>Winter</th>
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<td>R4</td>
<td>87.6</td>
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<td>62.9</td>
<td>44.9</td>
<td>60.8</td>
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<td>87.1</td>
<td>79.5</td>
<td>40.4</td>
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<td>72.6</td>
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<td></td>
<td>R7</td>
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<td>12.1</td>
<td>5.5</td>
<td>34.5</td>
<td>42.1</td>
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<td>11.2</td>
<td>16.3</td>
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<td>1.8</td>
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<td>30.6</td>
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adding the cases together, covering various altitudes on a seasonal and annual basis. R7, R6, and R9 were the top three regions for high POC for both heavy-polluted (6.3–9.8%) and light-polluted (33–42.1%) conditions, followed by R3 and R8. R5 and R10 were the only two regions with POC values lower than 2% for heavy polluted and lower than 10% for light-polluted conditions. While in general, regions having high PM$_{2.5}$ concentrations and/or high AEC also had high POC for heavy- and light-polluted conditions, there were some inconsistencies between the regions in terms of their orders of the highest to lowest values (e.g., R3 and R8), which were likely caused by other factors such as different aerosol chemical compositions and meteorological conditions in different regions (Tao et al., 2017).

The numbers of days with light- and heavy-polluted conditions were also estimated based on the vertical profiles of AEC POC (Table 3). Note that if at any height within the Lower-layer AEC followed into the 3rd–6th (7th–10th) intervals, it was treated as light-polluted (heavy-polluted). On an annual basis, R7, R8, and R9 were the top three regions with the most light-polluted (90–113) and heavy polluted (15–22) days. R3 (the capital economic circle) only had 62 light-polluted days and 13 heavy-polluted days. R5 (the Tibetan Plateau) and R10 (the East China Sea) had the lowest number of light- (20 and 43, respectively) and heavy-polluted (9 and 5, respectively) days. Again, the regional pattern of the number of polluted days was somewhat different from that of surface-layer PM$_{2.5}$ concentration. For example, R3 and R4 (in northern China) were expected to have as many polluted days as or even more than R7, R8, and R9, based on ground measurements of PM$_{2.5}$ concentrations. This implies that using AEC profile data may only allow us to semi-quantitatively identify PM$_{2.5}$ concentration levels in different regions due to the large variations in PM$_{2.5}$ chemical composition.

Seasonal AEC

The 10 regions studied here cover different climate zones, with very different seasonal meteorological conditions, which can influence pollutant sources, transport, chemical transformation, and atmospheric dry and wet removal processes. Besides, there are different dominant seasonal
Table 3. The estimated number of days under light- and heavy-polluted conditions in the Lower-layer and Upper-layer over the 10 regions on annual and seasonal basis. This is done by multiplying the 365 days with the maximum probability of EC vertical profile of the 10 individual EC intervals (the 3rd–6th intervals were summed as light-polluted and the 7th–10th intervals were summed as heavy polluted).

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<th>R1</th>
<th>R2</th>
<th>R3</th>
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<td>41</td>
<td>62</td>
<td>47</td>
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<td>53</td>
<td>96</td>
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<td>Upper-layer Light polluted Annual</td>
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Emission sources in different regions, as summarized in Tao et al. (2017). To explore how the seasonal emission sources were reflected in AEC patterns, seasonally averaged AEC, POC, and the estimated number of polluted days were generated (Tables 2 and 3).

Seasonal variations in AEC were mostly in the range of 39%–73% in all the regions, except for reaching a factor of 3.4 in R5. The very high seasonal variation in R5 was mostly due to the high AEC value in summer. The highest seasonal average for AEC appeared in spring in R1 and R10, summer in R5, autumn in R3 and R8, and winter in R2, R4, R6, R7, and R9. The highest seasonal AEC in spring in R1 (desert region) and R10 (marine) was likely due to strong winds in this season lifting surface-level soil dust or sea-salt aerosols to upper levels. In contrast, the lowest seasonal average for AEC was found in spring in R3 (the capital economic region), likely due to the frequent strong winds from the western and northern mountains to the southern plains of the region, which were conducive to pollutant diffusion. Surface PM$_{2.5}$ concentrations observed in several major cities of this region (Beijing, Tianjin, and Shijiazhuang) averaged 138, 121, 155, and 186 µg m$^{-3}$ in spring, summer, autumn, and winter, respectively (Tao et al., 2017), which were mostly consistent with the seasonal AEC patterns. The highest seasonal AEC in autumn in R8 reflected the influence of extensive biomass burning activities in this region (Cao et al., 2005). The highest seasonal AEC values in the other four regions in winter should be due to the residential fossil fuel and coal burning, as discussed in Tao et al. (2017).

Seasonal patterns of the number of light- and heavy-polluted days were also different from region to region. For example, there were more (light plus heavy) polluted days in spring and summer than in fall and winter (19 versus 12–14) in R1, least polluted days in spring (4–15) than in the other seasons (5–47) in R2, R3, R4, R5, R6, R7 and R9, most polluted days in summer (12–28) than in the other seasons (4–15) in R4, R5 and R6. Every season had a substantial number of polluted days (20–33) in R8.

**Regional AEC in the Upper-layer**

**Annual AEC**

The annual average for AEC for the Upper-layer showed regional differences up to a factor of 2.7 (ranging from 0.022 to 0.059 km$^{-1}$), which were smaller than those in the Lower-layer. The sequence from the largest to smallest (R9, R7, R1, R6, R8, R4, R3, R2, R10, and R5) was also very different from that in the Lower-layer, suggesting different vertical transport patterns of aerosol pollution in different regions. For example, AEC in R1 ranked the third largest among all the regions in the Upper-layer but ranked sixth in the Lower-layer, which can be partially explained by dust transport patterns shown by Huang et al. (2008). The vertical distribution characteristics of dust aerosols in the Taklimakan Desert showed a two-layer structure with the highest dust frequencies appearing at 9–11 km and at 3 km. In contrast, AEC in R3 ranked only seventh in the Upper-layer but fourth in the Lower-layer, indicating inefficient vertical transportation as a result of efficient horizontal diffusion, as mentioned above.
Despite high altitudes, POC on an annual basis still reached 3.3%–14.2% and 0.7%–1.8% for light- and heavy-polluted conditions, respectively. The totals for light- and heavy-polluted days were estimated to be 33 to 109 days, depending on the region. POC for polluted conditions and the estimated number of polluted days were larger in the eastern and southern regions (R9, R7, and R8) than in the western and central regions (R1 and R6), northern regions (R4 and R3), East China Sea (R10), northwest region (R2), and Tibetan Plateau (R5). Such a regional pattern was similar to yet slightly different from that for AEC, as discussed above.

Conclusions

Regional and seasonal AECs in the boundary layer and upper troposphere were quantified for 10 typical geographical regions in China. Regional variations in the annual average AEC were up by a factor of 3.7 in the Lower-layer and a factor of 2.7 in the Upper-layer. Seasonal variations in AEC within each region were mostly on the order of a few percent. The proportion of occurrence (POC) under clean, light-polluted and heavy-polluted conditions was also calculated based on the vertical profiles of AEC distributions. On an annual basis, POC in the Lower-layer ranged from 5.5% to 42.1% and from 1.3% to 9.8%, depending on the region, for light- and heavy-polluted conditions, respectively. POC in the Upper-layer still reached above 10% in some regions. Depending on the region, on an annual basis, the number of light-polluted days was estimated to be in the range of 20–113 and the number of heavy-polluted days, in the range of 5–22 days in the Lower-layer, with corresponding numbers of 24–96 and 5–13 in the Upper-layer.

This study provides an AEC database that can be used to evaluate climate models and guide future measurement needs in aerosol pollution. For example, the Yunnan-Guizhou Plateau was found to have AEC values similar to those in the most polluted regions of central and northern China; these results need to be verified by field measurements of chemically resolved PM$_{2.5}$ data. There are also some discrepancies between the AEC values, the estimated number of polluted days and the limited surface-level PM$_{2.5}$ data in terms of regional distributions, which may be explained by air-quality model simulations. The boundary-layer height generated in this study also needs to be verified in future studies, which may in turn improve the method developed here.

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Supplementary Material

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

Reference


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