Variability in the backscattering efficiency of particles in the Bohai and Yellow Seas and related effects on optical properties

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Abstract: The backscattering efficiency of particles is a crucial factor that relates light backscattering with biogeochemical properties. In this study, based on in situ measurements of the backscattering coefficient \( b_{\text{bp}}(\lambda) \), particle biogeochemical variables and remote sensing reflectance \( R_{\text{rs}}(\lambda) \) in two typical shallow and semi-enclosed seas, namely the Bohai Sea (BS) and Yellow Sea (YS) during the late spring, late summer and late autumn, we examined particulate pseudo-backscattering efficiency variability at 640 nm \( P_{Q_{\text{bsb}}}(640) \) and related optical effects. The results show that the \( P_{Q_{\text{bsb}}}(640) \) levels varied by nearly two orders for all of the samples examined. This high degree of \( P_{Q_{\text{bsb}}}(640) \) variability significantly affected \( b_{\text{bp}}(640) \) and the mass-specific backscattering coefficient \( b_{\text{bp}*}(640) \), showing that approximately 63.7% and 20.8% of the variability in the \( b_{\text{bp}*(640)} \) and \( b_{\text{bp}}(640) \) was attributed to the \( P_{Q_{\text{bsb}}}(640) \), respectively. More importantly, consistent with the observations of Wang et al. [J. Geophys. Res.: Oceans 121, 3955 (2016)], the \( P_{Q_{\text{bsb}}}(640) \) results clearly showed two clusters and this clustering changed the relationships between \( b_{\text{bp}*(640)} \), \( b_{\text{bp}}(640) \) and \( R_{\text{rs}}(640) \) with the biogeochemical variables. However, we confirm that \( P_{Q_{\text{bsb}}}(640) \) clustering generally remained intact across seasons. Therefore, a simple scheme based on a threshold of the \( P_{Q_{\text{bsb}}}(640) \) data is proposed for the classification of particle types. With this classification, impacts of \( P_{Q_{\text{bsb}}}(640) \) on \( b_{\text{bp}*(640)} \) and \( b_{\text{bp}}(640) \) were clearly reduced, and co-variation trends of \( b_{\text{bp}*(640)} \), \( b_{\text{bp}}(640) \) and \( R_{\text{rs}}(640) \) with biogeochemical variables can be in turn more accurately described. Overall, this study provides general information on \( P_{Q_{\text{bsb}}}(640) \) variability in the BS and the YS and consequent effects on optical properties. The scheme for particle type classification may also provide a useful basis for better modeling marine biogeochemical processes related to particulate backscattering and for the development of ocean color algorithms.

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References and links
1. Introduction

Particles suspended in seawater such as phytoplankton, detritus and mineral particles play an important role in the determination of optical properties of water [1,2]. Sunlight that enters the sea surface is either absorbed or scattered by suspended particles [3]. Light scattered in a backwards direction is of fundamental importance for many marine biogeochemical processes that involve optics [4,5]. In particular, it is essential for interpreting satellite ocean color data [6,7]. A detailed understanding of how particle assemblages affect backscattering properties is thus of importance for studies on water radiative transfer, remote sensing of ocean color and biogeochemical processes coupling optics based on in situ and satellite measurements [8].

According to Mie theory, assuming a spherical shape and material homogeneity for a given particle, the backscattering coefficient of particles \( b_{bp}(\lambda) \) at a given wavelength can be expressed as the sum of the product of the backscattering efficiency and the cross-sectional area of all particles [9]. Using the mean backscattering efficiency of all particles weighted by area \( Q_{bse}(\lambda) \) and the total cross-sectional area concentration (AC), the calculation of Mie theory can be simplified as [10,11],

\[
b_{bp}(\lambda) = Q_{bse}(\lambda)AC
\]

Based on experimental and field observations, backscattering properties of particles have been studied in various water bodies [12–21]. It is generally known that \( b_{bp}(\lambda) \) is positively correlated with mass concentrations of total suspended matter (TSM) [6,22], and various remote sensing algorithms for deriving TSM from satellite measurements have been proposed [23–26]. Such observations are based on the correlation between TSM and AC [27,28], which is expressed as,

\[
TSM = \frac{2AC}{3\rho_aD_\lambda}
\]

where \( \rho_a \) and \( D_\lambda \) denote the mean apparent density and mean diameter of particles, respectively. Clearly, \( b_{bp}(\lambda) \) and the remote sensing of TSM are affected by combined effects of particle density, size and backscattering efficiency. In past decades, some studies have examined the influence of particle size, density and composition on backscattering properties, although the findings of these studies have not always been consistent [7,11,21,29–34]. However, the effects of backscattering efficiency on backscattering properties and thereby on ocean color remote sensing are still not well documented.

The results of several theoretical, experimental and field studies have shown that backscattering efficiency can significantly vary with changes in particle compositions and size distributions [7,8,11,19,21,35–37]. Based on theoretical principles, Stramski et al. [8] investigated the effects of particle compositions on backscattering efficiency and showed that when soft organic particles transform into mineral particles, backscattering efficiency levels increase by a factor of 30. Vaillancourt et al. [7] calculated backscattering efficiency levels at 620 nm for cultured 28 phytoplankton species of 11 marine classes and showed that backscattering efficiency levels vary from 0.001 to 0.068. In reference to the Bedford Basin, based on in situ observations made before and after a spring phytoplankton bloom, Flory et al. [36] found that the efficiency of backscattering at 589 nm decreased by a factor of nearly 4 when organic particles gradually became dominant and detrital particles began to aggregate with the starting and the progression of the blooming. More recently, along the west coast of Great Britain, Bowers et al. [19] observed that the backscattering efficiency level at 665 nm changes by an order of magnitude when the proportion of inorganic matter to TSM changes from 35% to 90%. Meanwhile, they found a strong positive relationship \( (R^2 = 0.62) \) between backscattering efficiency and the ratio of mineral particles to TSM, implying that particulate backscattering efficiency may be a good indicator for detecting particle compositions.
It is well known that particle assemblages often undergo dynamic responses to changes in hydrographic environments of water columns [38]. In the open ocean, suspended particles are mainly composed of phytoplankton cells and of their detrital products; in coastal regions, with the exception of phytoplankton and detritus, mineral particles introduced through river discharge, upwelling, mixing, etc., are also abundant [19]. Complex and dynamic particle compositions in coastal waters may consequently result in considerable variability in backscattering efficiency levels and thereby in $b_{bp}(\lambda)$ and $b_{bp}^*(\lambda)$. Recently, based on field measurements collected during the late summer in two typical shallow and semi-enclosed seas, the Bohai Sea (BS) and Yellow Sea (YS), we found that the backscattering efficiency of suspended particles at 640 nm significantly vary in magnitude [21]. Meanwhile, backscattering efficiency levels were clearly clustered into two types. Type 1 samples containing relatively high proportions of organic or large particles were found to show low levels of backscattering efficiency and the backscattering efficiency of type 2 samples, which are mainly composed of relatively small mineral particles, was found to be high. Overall, as stated above, while some studies have examined the variability of particle backscattering properties with regards to biogeochemical variables, effects of backscattering efficiency on $b_{bp}(\lambda)$, $b_{bp}^*(\lambda)$ and thus ocean color remote sensing and the extent to which such effects can be compared with respect to particle concentrations, density levels and size distributions still need to be completely examined and quantified.

In this paper, based on field observations made in the BS and YS over three seasons, i.e., late spring, late summer and late autumn, we first investigate backscattering efficiency variability levels and examine whether the clustering of backscattering efficiency as observed in the late summer in Wang et al. [21] is supported in other seasons. Subsequently, a simple scheme for particle classification based on backscattering efficiency is proposed. Finally, effects of backscattering efficiency on $b_{bp}(\lambda)$ and $b_{bp}^*(\lambda)$ are quantified, and potential effects of particle types on ocean color remote sensing are discussed.

2. Materials and methods

2.1 Study area and sample collection

In this study, we report on field data collected from the BS and YS. The BS is a shallow sea with an average depth of 18 m [39]. The Yellow River, as the second largest sediment-load river in the world, flows into the BS with a total runoff of $890 \times 10^8$ m$^3$ each year [40]. The YS is also a relatively shallow sea with an average water depth of 44 m [39]. It is located between Mainland China and the Korean Peninsula and is connected to the BS to the north through the Bohai Strait. The topography of the YS in the central region is rather smooth but steep on both the western and eastern sides. These two relatively shallow seas are heavily influenced by monsoons in the winter and by freshwater discharge in summer. Interactions among winds, bottom topography, freshwater discharge, and tidal forcing cause significant regional and seasonal variability in water properties in the BS and YS [41]. Meanwhile, surrounding areas of the BS and YS are the most economically developed regions in northern China. The rapid proliferation of industries, agriculture, aquaculture and domestic sewage from these areas has seriously polluted the BS and YS over the past several decades [42].

Three cruises were conducted in the BS and YS during the late spring (May 2014), late summer (August 2015) and late autumn (November 2014). The locations of observation stations employed by the three cruises were generally similar [Fig. 1]. At each station, a profiling package with a HOBI Labs Hydroscat-6, Sequoia Scientific LISST-100X (type C) and Seabird SBE911P conductivity-temperature-depth (CTD) profiler was used to synchronously measure light backscattering, particle size distribution (PSD) and water column temperature and salinity values, respectively. The package was deployed in a surface layer (roughly 5 m) for several minutes to allow the equilibration of the sensor temperature and seawater temperature. The package was then lifted to the surface and slowly (approximately
0.2 m s\(^{-1}\)) lowered to the depth 2–3 m just above the bottom to measure a vertical profile of the water column. To avoid package perturbations to the water column, only downward looking measurements were used for our data analysis. These are denoted ‘downcast’ measurements in this study. Simultaneously, water samples were collected from three layers (the surface, the middle layer just below the mixing depth and the bottom layer) to measure TSM values. In addition, to determine remote sensing reflectance \((R_{rs}(\lambda))\) values, a Satlantic Hyper-Profiler II radiometer was performed whenever the observing condition was appropriate.

2.2 Particle mass concentration, backscattering and size distribution measurements

TSM concentrations were determined using a gravimetric technique. Seawater (0.5–2 L) collected using Niskin bottles was filtered onto pre-weighed 47-mm Whatman GF/F glass fiber filters under low vacuum pressure levels (< 0.01 MPa). To remove salt, the filters were rinsed three times using 50 ml MilliQ water after filtration and then were kept frozen at –20 °C until being dried at 105° for 4 h in a laboratory. These filters were then reweighed to determine TSM concentrations.

Total backscattering coefficients \((b_b(\lambda))\) at 410, 442, 488, 532, 550, and 640 nm were measured using a HOBI Labs Hydroscat-6, which was calibrated before the cruise to confirm its performance within factory specifications. This instrument records the total volume-scattering function at a fixed angle (approximately 140°) in a backwards direction [43]. To improve the accuracy of the backscattering measurements, path length attenuation effects were corrected using the sigma correction method. The absorption and scattering data used in sigma correction for the data in August 2015 were obtained from the simultaneous measurements of a WET labs AC-S, while those used in sigma correction for the data in May and November 2014 were estimated using the empirical models provided in Hydrosoft. The backscattering coefficient of particles \(b_{bp}(\lambda)\) was then calculated by subtracting the backscattering coefficient of pure water from the \(b_b(\lambda)\). Further information on the processing procedure can be found in the HOBI Labs User’s Manual.

An LISST-100X Type-C particle size analyzer (Sequoia Scientific Inc.) was used to determine the particle size distribution (PSD). The background scattering of LISST instrument was acquired before the cruise. In brief, this instrument measures the diffraction pattern.
produced by suspended particles in a volume of water. Based on Mie theory calculations, the diffraction pattern is used to determine the particle volume concentrations with a mean diameter of 32 sized bins that are logarithmically placed across a continuous size spectrum from 2.5 to 500 μm [44,45]. Volume concentrations and mean sizes of particles were processed using the LISST-SOP software program provided by the manufacturer (LISST-100X Particle Size Analyzer, 2013). Meanwhile, as noted in previous studies [11,46], LISST instrument measurements are not typically stable at the smallest and largest size ranges, likely due to the presence of particles that are smaller or larger than the measured size range. The instability of the smallest size ranges may also be associated with stray light [47]. Therefore, data from the smallest and largest size ranges were excluded in our analysis.

2.3 Particle backscattering efficiency determination

To determine the backscattering efficiency of particles, we first calculated the cross-sectional area concentration $AC_i$ from LISST measurements of the volume concentration for a mean diameter. By assuming particles are spherical, the cross-sectional area concentration of particles of the $i$th size bin ($AC_i$) is calculated as,

$$AC_i = \frac{3}{2D_i} VC_i$$

where $VC_i$ and $D_i$ denote the volume concentration and mean diameter of particles in size bin $i$, respectively. The total cross-sectional area concentration $AC$ was then obtained as,

$$AC = \sum_{i=2}^{31} AC_i$$

where $i$ ranges from 2 to 31 with $3.2 \mu m \leq D_i \leq 390 \mu m$ (the first and last size bins were excluded). The mean backscattering efficiency of particles $Q_{obs}(\lambda)$ was then calculated as the ratio of $b_{bp}(\lambda)$ to $AC$.

Meanwhile, the mean apparent density $\rho_a$ and mean diameter $D_a$ of particles was calculated [10,11], to compare their effects on backscattering properties with those of $Q_{obs}(\lambda)$, as follows:

$$\rho_a = \frac{TSM}{VC}$$

$$D_a = \frac{\sum_{i=2}^{31} AC_i D_i}{AC}$$

where VC denotes the total volume concentration obtained by summing $VC_i$ values of the size bins from 2 to 31. Therefore, the particulate backscattering coefficient $b_{bp}(\lambda)$ and mass-specific backscattering coefficient $b_{bp}^*(\lambda)$ can be expressed as follows [10,11]:

$$b_{bp}(\lambda) = \frac{3}{2} \frac{Q_{obs}(\lambda) TSM}{\rho_a D_a}$$

$$b_{bp}^*(\lambda) = \frac{3}{2} \frac{Q_{obs}(\lambda)}{\rho_a D_a}$$

At this stage, it should be noted that there is a potential mismatch in the particle size ranges of the measurements of TSM, AC, and $b_0(\lambda)$. The TSM represents the mass concentration of particles retained on the GF/F filter (nominal pore size of 0.7 μm), which have a size larger than approximately 0.4 μm [11]. The AC determined in this study accounts for the particles between 3.2 and 390 μm, due to the limited capability of the LISST measurement.
there is no size limitation for $b_{bp}(\lambda)$ measured by the Hydroscat-6. Such particle size discrepancies of various measurements may bring uncertainties in analysis of bio-optical properties of this study, such as potential overestimation of $b_{bp\ast}(\lambda)$ and $Q_{bbe}(\lambda)$. Particularly, due to the mismatch in particle size ranges between $b_{bp}(\lambda)$ and AC measurements, the key parameter $Q_{bbe}(\lambda)$ obtained in this study may be not strictly the true particulate backscattering efficiency. Thus, it was referred to pseudo-backscattering efficiency ($P_{Q_{bbe}}(\lambda)$) hereafter. However, note that if the particles smaller than 3.2 $\mu$m and larger than 390 $\mu$m have consistent contribution proportion to AC for all of the samples, the $P_{Q_{bbe}}(\lambda)$ may be proportional to the true backscattering efficiency. In this case, the clustering characteristics of $P_{Q_{bbe}}(\lambda)$ obtained in this study should represent those of the true backscattering efficiency, though the clustering boundary may not be consistent with the natural condition. Despite these limitations, the LISST-derived particle size and AC (used to calculate $P_{Q_{bbe}}(\lambda)$ in this study) are useful parameters, and have been commonly used to understand particulate backscattering, scattering and attenuation properties guided by the optical theory [10,11,28–34]. In addition, in the following analysis, we focuses on the $P_{Q_{bbe}}(\lambda)$ variability at 640 nm ($P_{Q_{bbe}}(640)$) and consequent effects on optical properties to be consistent with the results of the previous study [21].

2.4 Remote sensing reflectance measurements

Remote sensing reflectance $R_{rs}(\lambda)$ at daytime stations was derived from Satlantic Hyper-Profiler II measurements. This instrument includes a deck radiometer that measures above-water downwelling irradiance ($E_d(\lambda, 0^\circ)$) values and two underwater radiometers that measure vertical profiles of downwelling irradiance ($E_d(\lambda, z)$) and upwelling radiance ($L_u(\lambda, z)$). The three radiometers were inter-calibrated by the manufacturer before the cruise. The measured spectra ranged from 349 to 804 nm with a mean bandwidth of approximately 3.3 nm.

While making our observations, a deck sensor was mounted on the deck to prevent shadows from the ship’s structure from having an effect. Underwater sensors were stabilized in the surface waters for several minutes using a hand-controlled cable. This allowed the instrument to drift further away from the ship to avoid contact with the ship’s shadow and to also equilibrate the sensor and seawater temperatures. Subsequently, the underwater sensors cluster was lowered from the surface in a vertically free-falling mode until they reached the euphotic layer (at a depth of 1% surface photosynthetically active radiation) [48]. We only used downcast radiometric measurements to perform calibration, data filtering, binning and interpolation based on the Prosoft 7.7.16 software program supplied by the manufacturer [49]. Data with tilt angles of $> 5^\circ$ or/and a downward velocity of $> 0.5$ m s$^{-1}$ were excluded during processing. The $R_{rs}(\lambda)$ spectra were then calculated as

$$R_{rs}(\lambda) = \frac{L_u(\lambda)}{E_d(\lambda, 0^\circ)} \quad (9)$$

where $L_u(\lambda)$ denotes the water-leaving radiance derived from profile measurements of $L_u(\lambda, z)$ of upper layer waters (see further information in Rudorff et al. [49]).

3. Results

3.1 Backscattering efficiency variability

Large variability in the pseudo-backscattering efficiency $P_{Q_{bbe}}(640)$ was observed within and across different seasons in the BS and YS [Table 1]. During the late spring, for the samples combined in the surface, middle and bottom layers, the $P_{Q_{bbe}}(640)$ results varied from 0.0022 to 0.1049 with a mean value of 0.0277 and a standard deviation of 0.0183 (coefficient of variation of 65.9%). During the late summer, the $P_{Q_{bbe}}(640)$ data showed more significant variability, covering a range of 0.0022 to 0.1884 (mean ± standard deviation of 0.0319 ± 0.0268). The coefficient of variation correspondingly increased to 84.0%. In contrast,
P\textsubscript{Qbbe}(640) variability in the late autumn decreased compared with that in the late spring and late summer. The coefficient of variation for the late autumn was the lowest at 41.7%, and the mean value of the P\textsubscript{Qbbe}(640) data was the highest at 0.0338 (standard deviation of 0.0141).

To investigate P\textsubscript{Qbbe}(640) variation patterns, we analyzed the frequency distributions of the P\textsubscript{Qbbe}(640) data in log-space. The logarithmic normality tests of these distributions were examined using Kolmogorov–Smirnov test with the null hypothesis of log-normal distribution, and the \(p\)-values were provided for each test. As is shown in Fig. 2, clear differences were observed in distributions of the P\textsubscript{Qbbe}(640) results across different seasons. Similar to the P\textsubscript{Qbbe}(640) distribution for the late summer reported by Wang et al. [21], the P\textsubscript{Qbbe}(640) data of the late spring showed two clear clusters with low and high mean values, and each was approximately log-normally distributed. However, in the late autumn, with exception of low P\textsubscript{Qbbe}(640) values found for some samples, the P\textsubscript{Qbbe}(640) values of most samples were high. More importantly, we found that unlike two normal distribution patterns for the late spring and late summer, the P\textsubscript{Qbbe}(640) for the late autumn generally included one cluster, and the distribution was similar to that of the cluster with high P\textsubscript{Qbbe}(640) values observed for late spring and late summer.

### Table 1. Statistics on the P\textsubscript{Qbbe}(640) from different water layers in terms of maximum (Max), minimum (Min), standard deviation (SD) and coefficient of variation (CV).

<table>
<thead>
<tr>
<th>Data set</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>CV (%)</th>
<th>N</th>
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<tbody>
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<td></td>
<td></td>
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<tr>
<td>Late spring</td>
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<td>0.0277</td>
<td>0.0183</td>
<td>65.9</td>
<td>331</td>
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<tr>
<td>Late summer</td>
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<td>0.0319</td>
<td>0.0268</td>
<td>84.0</td>
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<td>0.0776</td>
<td>0.0338</td>
<td>0.0141</td>
<td>41.7</td>
<td>291</td>
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<td></td>
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<tr>
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<td>0.0211</td>
<td>0.0175</td>
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<tr>
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<td>0.0139</td>
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<td>Late autumn</td>
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<td>0.0283</td>
<td>0.0140</td>
<td>49.4</td>
<td>106</td>
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<tr>
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<td></td>
<td></td>
<td></td>
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<tr>
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<tr>
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<td>0.0333</td>
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<td>Bottom Layer</td>
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<tr>
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<td>0.0630</td>
<td>0.0353</td>
<td>0.0109</td>
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<td>0.0566</td>
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</table>
Fig. 2. Frequency distribution of the pseudo-backscattering efficiency at 640 nm (P\text{\textsubscript{Q\text{bbe}}}(640)) for samples collected during the late spring (a), late summer (b) and late autumn (c). Red dashes denote the bound for P\text{\textsubscript{Q\text{bbe}}}(640) classification. Blue lines denote the log-normal fitting curves. The p-values indicate the significance level of the logarithmic normality tests.

To further identify differences in P\text{\textsubscript{Q\text{bbe}}}(640) across different seasons, the P\text{\textsubscript{Q\text{bbe}}}(640) values of each sampling layer for the late spring, late summer and late autumn were compared [Table 1]. In surface waters, the mean P\text{\textsubscript{Q\text{bbe}}}(640) value was the lowest in the late summer at 0.0139 but was the highest in the late autumn at 0.0283. The lowest level of P\text{\textsubscript{Q\text{bbe}}}(640) variability was observed in the late autumn with a coefficient of variation of 41.8%. These observations were also true for samples drawn from middle layers. In bottom layers, the P\text{\textsubscript{Q\text{bbe}}}(640) values for the late autumn also showed the lowest level of variability (coefficient of variation of 24.9%). The mean P\text{\textsubscript{Q\text{bbe}}}(640) for the late summer was the highest at 0.0566 while it was the lowest in the late spring (a mean value of 0.0353).

Comparisons between P\text{\textsubscript{Q\text{bbe}}}(640) frequency distributions for the surface, middle and bottom layers of the late spring, late summer and late autumn are shown in Fig. 3. Clearly, for the bottom layer, although absolute values of the P\text{\textsubscript{Q\text{bbe}}}(640) showed some differences across the three periods, distribution patterns were generally similar, showing a single cluster with high P\text{\textsubscript{Q\text{bbe}}}(640) values. However, for the surface and middle layers, the P\text{\textsubscript{Q\text{bbe}}}(640) distribution for the late autumn differed from that of the late spring and late summer. Generally speaking, the P\text{\textsubscript{Q\text{bbe}}}(640) values in the late spring and late summer displayed approximately two clusters, while values for the late autumn generated roughly a single cluster. Overall, comparisons made between Figs. 2 and 3 show that P\text{\textsubscript{Q\text{bbe}}}(640) variability for the late autumn clearly differs from that of the late spring and late summer, and these differences are mainly attributable to the surface and middle water layers. Such differences in P\text{\textsubscript{Q\text{bbe}}}(640) distributions are likely attributable to particle types related to various water conditions as discussed in a later section.
3.2 Particle classification scheme based on backscattering efficiency

The analysis presented in section 3.1 confirms that although the number of clusters in $P_{bb}(640)$ showed some seasonal differences, the clustering of particles is generally stable across seasons. Backscattering efficiency is a key variable related to backscattering properties shown in Eqs. (7) and (8). Therefore, in this paper, we propose a simple scheme to classify particles based on distribution patterns of the $P_{bb}(640)$ results, in order to improve our understanding of particle backscattering properties and in turn the remote sensing of ocean color.

As is shown in Fig. 4(a), when we combined all of the samples collected from three water layers of in the late spring, late summer and late autumn, two clusters of samples were clearly found. Based on the frequency distribution of the $P_{bb}(640)$ data, a threshold of the $P_{bb}(640)$ data with a value of 0.0134 was empirically determined for the classification. It should be stated that determination of the empirical threshold depends on the bin size of the histogram, while it was found that changes of bin size caused negligible influences on the particle classification for our data set. The $P_{bb}(640)$ values of type 1 samples showed a mean value of 0.0063 with a standard deviation of 0.0027 while the mean $P_{bb}(640)$ of the type 2 samples was much higher than that of the type 1 samples, generating a value of 0.0397 ± 0.0200. Upon applying this classification scheme to the data for different seasons and to seasonal surface data, the samples were reasonably classified [Figs. 2, 3 and 4(b)].
Fig. 4. Classification of particle types based on the frequency distribution of the pseudo-backscattering efficiency of particles at 640 nm ($P_{Q_{bb}}(640)$) for all of the samples (a) and surface samples (b). Red dashes denote a $P_{Q_{bb}}(640)$ threshold of 0.0134 for the classification. Blue lines denote the log-normal fitting curves. The $p$-values indicate the significance level of the logarithmic normality tests.

Using the proposed classification method, 47.4%, 36.6% and 1% of the samples respectively drawn from the surface, middle and bottom layers during the late spring were classified as type 1; and corresponding 52.6%, 63.4% and 99% of the samples drawn from the surface, middle and bottom layers were respectively classified as type 2. During the late summer, 65.4% of the surface samples, 34.7% of the middle layer samples and 100% of the bottom layer samples were classified as type 1 particles, and the remainder were classified as type 2 particles. Type 1 particles collected from the surface and middle layers during the late spring and late summer mainly derived from coastal regions (data not shown). During the late autumn, most samples were classified as type 2 particles, accounting for 80%, 88.4% and 100% of samples collected from the surface, middle and bottom layers, respectively.

Meanwhile, for the potential application of the $P_{Q_{bb}}(640)$-based classification scheme to satellite observations, we propose a threshold for $Q_{bb}(550)$ to classify the particles, as the band at 550 nm is often used for most current ocean color sensors. We found that $P_{Q_{bb}}(550)$ was strongly related to $P_{Q_{bb}}(640)$ with an $R^2$ value of 0.991 in log-log space (data not shown). Consequently, a threshold of 0.0157 for $P_{Q_{bb}}(550)$ is designated for classifying particles from satellite measurements in the future.

3.3 Backscattering efficiency effects on backscattering properties

In theory, the particulate backscattering coefficient $b_{bp}(\lambda)$ is regulated by the backscattering efficiency value together with the cross-sectional area or by combined effects of backscattering efficiency, TSM and product of apparent particle density and mean diameter ($\rho a D A$)$^{-1}$ [Eqs. (1) and (7)]. Similarly, the second-order variability of $b_{bp}(\lambda)$ represented by the mass-specific backscattering coefficient $b_{bp}^*(\lambda)$ is controlled by backscattering efficiency, TSM and ($\rho a D A$)$^{-1}$ values [Eq. (8)]. Here we quantify the extent to which the $P_{Q_{bb}}(640)$ can affect the relationship between backscattering and biogeochemical properties by applying a linear regression in log-log space. We analyze these relationships in log-log space with respect to their log-normal distributions.

For $b_{bp}^*(640)$, it was found that $P_{Q_{bb}}(640)$ clearly separated relationships between $b_{bp}^*(640)$ and ($\rho a D A$)$^{-1}$ into two patterns [Fig. 5]. When we combined all of the samples, a
The coefficient of determination $R^2$ with a value of 0.363 was found between $b_{bp}^*(640)$ and $(\rho_a D_A)^{-1}$, and an $R^2$ value of 0.673 was obtained between $b_{bp}^*(640)$ and $P_{Q_{bbe}}(640)$ [Fig. 5 and Table 2]. Note that $P_{Q_{bbe}}(640)$ calculations are dependent on $b_{bp}(640)$ while $(\rho_a D_A)^{-1}$ is independent of $b_{bp}(640)$. These observations imply that for all of the samples, only 36.3% of the variability in $b_{bp}^*(640)$ can be explained by $(\rho_a D_A)^{-1}$, and the remaining 63.7% can likely be attributed to $P_{Q_{bbe}}(640)$, which generally agrees with the $R^2$ value of 0.673 between $b_{bp}^*(640)$ and $P_{Q_{bbe}}(640)$. After classification, the effects of $P_{Q_{bbe}}(640)$ on $b_{bp}^*(640)$ significantly decreased, showing that $b_{bp}^*(640)$ variability for type 1 and 2 samples is mainly attributed to $(\rho_a D_A)^{-1}$ with contributions of 60.9% and 71.5%, respectively. For the type 2 samples, this observation was generally consistent with the relationship between $b_{bp}^*(640)$ and $P_{Q_{bbe}}(640)$, showing an $R^2$ value of 0.320. However, we found that for the type 1 samples, the $R^2$ value for the relationship between $b_{bp}^*(640)$ and $P_{Q_{bbe}}(640)$ was only 0.021. This observation implies that $b_{bp}^*(640)$ variability in type 1 samples cannot be fully explained by $(\rho_a D_A)^{-1}$ and $P_{Q_{bbe}}(640)$. Other factors may also modulate $b_{bp}^*(640)$ variability as we discuss below.

Vol. 24, No. 26 | 26 Dec 2016 | OPTICS EXPRESS 29372

The relationships of $b_{bp}(640)$ with AC, $P_{Q_{bbe}}(640)$, TSM and $(\rho_a D_A)^{-1}$ were analyzed [Fig. 6 and Table 2]. It was noted that $P_{Q_{bbe}}(640)$ calculation depends on $b_{bp}(640)$, while AC, TSM and $(\rho_a D_A)^{-1}$ are independent of $b_{bp}(640)$. In regards to $b_{bp}$ (640) variability in all of the samples, the effects of $P_{Q_{bbe}}(640)$ decreased to 20.8%, and most $b_{bp}$ (640) variability was attributed to AC, which showed high $R^2$ values of 0.792. The classification of particles shows that 92.0% of the variability in $b_{bp}$ (640) of type 2 samples was explained by AC, and corresponding effects of $P_{Q_{bbe}}(640)$ may decrease to 8%. For type 1 samples, AC was responsible for 61.2% of $b_{bp}$ (640) variability and still more than 30% of the variability remained unexplained, echoing results of the $b_{bp}^*(640)$ analysis presented above. Regarding...
factors concerning TSM, \((\rho_a D_A)^{-1}\) and \(P_{Qbbe}(640)\), we found that for all of the samples, the majority (65.7%) of the variability in \(b_{bp}(640)\) was attributed to TSM; and contributions of \((\rho_a D_A)^{-1}\) was 14.1%. In this sense, \(P_{Qbbe}(640)\) effects likely accounted for approximately 20.2%. After classification, 71.0% and 26.8% of the variability in \(b_{bp}(640)\) of type 2 samples was attributable to TSM and \((\rho_a D_A)^{-1}\), respectively, and \(P_{Qbbe}(640)\) may thus only explain 2.2% of the variability. For the type 1 samples, TSM and \((\rho_a D_A)^{-1}\) were respectively responsible for 38.9% and 1.1% of \(b_{bp}(640)\) variability, and the remaining percentage of variability remained unexplained. Overall, from above regression analysis of the bio-optical variables [Figs. 5 and 6, Table 2], it was generally found that \(P_{Qbbe}(640)\) effects on \(b_{bp}(640)\) decreased compared to effects on \(b_{bp}^*(640)\) for all of the samples. However, we note that the clustering of particles based on \(P_{Qbbe}(640)\) generated clearly different patterns in the resulting relationships between \(b_{bp}(640)\) and various biogeochemical properties (i.e., AC, TSM and \((\rho_a D_A)^{-1}\)).

3.4 Effects of backscattering efficiency on remote sensing reflectance

TSM is commonly used to refer to the amount of particle, and remote sensing algorithms have been developed to derive TSM from remote sensing reflectance \(R_{s}(\lambda)\) values. Therefore, we further examined effects of the clustering of particles generated by \(P_{Qbbe}(640)\) on \(R_{s}(\lambda)\) values based on 86 samples with concurrent measurements of \(R_{s}(\lambda)\) and bio-optical properties. Figure 7 presents scatter plots for \(R_{s}(640)\) and \(b_{bp}(640)\) and those for \(R_{s}(640)\) and TSM. We found that the \(R_{s}(640)\) of all of the samples strongly depended on \(b_{bp}(640)\) showed an \(R^2\) value of 0.927 between them. Meanwhile, relationships between \(R_{s}(640)\) and \(b_{bp}(640)\) for type 1 and type 2 samples were similar. However, as was expected, clear differences were observed in relationships between \(R_{s}(640)\) and TSM in terms of \(R^2\) values and in terms of covariation trends between type 1 and type 2 samples. Comparisons made between Figs. 6(c), 7(a) and 7(b)
show that disparate covariation $R_{r}(640)$ trends with TSM for both types of samples were mainly related to different relationships between $b_{bp}(640)$ and TSM caused by the clustering of $P_{Q_{bb}}(640)$.

All data: $y = 0.0002x^{1.2657}$ $R^2 = 0.5886$

Type 1: $y = 0.0003x^{0.4882}$ $R^2 = 0.407$

Type 2: $y = 0.0005x^{1.1448}$ $R^2 = 0.6245$

Correlations between $R_{rs}(\lambda)$ and TSM were also further analyzed for type 1 and type 2 samples at between 400 – 700 nm to investigate the potential effects of particle clustering on the remote sensing of ocean color. As is shown in Fig. 8, clear differences in the spectra of these correlations were found between type 1 and type 2 samples. For the type 2 samples, $R_{rs}(\lambda)$ showed similar high correlations with TSM from 400 to 700 nm in agreement with the optical properties of typical mineral-dominated particles [6]. However, for type 1 samples, correlations between $R_{rs}(\lambda)$ and TSM were obvious spectral dependence. In particular, in the range of 400 – 500 nm, correlations showed negative values; within a range of 650 – 700 nm, correlations clearly decreased. These negative correlations between 400 and 500 nm and low correlations between 650 and 700 nm may be resulted from strong phytoplankton absorption processes, as the type 1 samples were likely dominated by organic matter as was shown in our previous work [21]. These results suggest that the clustering of particles based on backscattering efficiency levels would generate different variation patterns in the relationships between biogeochemical variables and optical properties for different types of particles, and discretion should be used in particle clustering in the development of algorithms for estimating TSM and potentially other biogeochemical variables from $R_{rs}(\lambda)$.
4. Discussion

4.1 Backscattering efficiency variability in relation to hydrological environments

Backscattering efficiency of suspended particles is a key factor that links their biogeochemical properties with backscattering properties. As was observed in this study, the pseudo-backscattering efficiency at 640 nm $P_{\text{Qbbe}}(640)$ varies considerably and shows clear seasonal differences [Table 1 and Fig. 2]. For all samples collected in the late spring, late summer, and late autumn, the $P_{\text{Qbbe}}(640)$ varied from 0.0022 to 0.1884 by a factor of roughly 85. This degree of variability is greater than those reported in previous studies based on experimental data and in situ observations [7,11,19,35–37], while 90% of the $P_{\text{Qbbe}}(640)$ values found in this study range from 0.0040 to 0.0620, which were generally consistent with reported value ranges. For instance, the values of backscattering efficiency collected at 620 nm from 28 phytoplankton cultures varies between 0.0014 and 0.0681 as reported by Vaillancourt et al. [7]; and the backscattering efficiency values at 650 nm for mineral particles falls within a range of 0.0460 to 0.0620 [37].

Although the $P_{\text{Qbbe}}(640)$ values of all of the samples are highly varied, we found that $P_{\text{Qbbe}}(640)$ values were generally clustered into two types; within each type, $P_{\text{Qbbe}}(640)$ variability levels significantly decreased compared to that of the all of samples [Fig. 4]. This observation complements the findings of our previous study conducted in the late summer [21]. More importantly, we found that such $P_{\text{Qbbe}}(640)$ clustering holds well in different seasons [Fig. 2], although the clustering number showed seasonal differences. Based on measurements of particle compositions and size distributions for the late summer and based on comparisons made with the backscattering efficiency values dominated by organic and mineral particles presented in other studies, Wang et al. [21] inferred that type 1 samples with a mean value of 0.0054 ± 0.0018 are mainly composed of relatively high proportions of organic or large particles and that type 2 samples with a mean value of 0.045 ± 0.0305 mainly contain relatively small mineral particles. Consistent with this, although the present study considers more data for the late spring and late autumn, the mean $P_{\text{Qbbe}}(640)$ values of the type 1 and type 2 samples were generally similar at 0.0063 ± 0.0027 and 0.0397 ± 0.0200, respectively. These findings indicate that $P_{\text{Qbbe}}(640)$ clustering is a natural phenomenon in the BS and YS.

As noted above, although $P_{\text{Qbbe}}(640)$ clustering generally holds well in different seasons, the occurrence of type 1 and type 2 samples for all data showed some seasonal differences [Figs. 2 and 3]. For bottom water layers, $Q_{\text{bbe}}(640)$ distributions were generally similar across seasons, showing that almost all of the samples can be classified as type 2 that are likely dominated by mineral particles [Fig. 2]. However, in the surface and middle layers, clear differences were found in $P_{\text{Qbbe}}(640)$ distributions between late autumn, late spring and late summer. During the late spring and late autumn, type 1 samples that were likely dominated by organic or large particles contributed comparable proportions while during the late autumn, samples collected from the surface and middle layers were still mainly composed of type 2 samples. These seasonal differences in $P_{\text{Qbbe}}(640)$ distributions in surface and middle layers may be related to the respective dynamic hydrographic environments found during different seasons.

During the late spring and summer, offshore waters in the BS and YS are heavily stratified, favoring the dominance of phytoplankton. Furthermore, in coastal regions, strong tides in shallow regions often induce vertical mixing. In the late autumn, driven by the strong NE monsoon, most of the water columns in the BS and YS are thoroughly mixed [41]. Heavy water column mixing may draw mineral particles from the bottom to middle and surface layers [50,51], and this may change particle compositions and consequently $P_{\text{Qbbe}}(640)$ variability levels. To confirm this inference, we analyzed the spatial distributions of $P_{\text{Qbbe}}(640)$ and the boundary of the clustering in surface water layers in the three seasons [Fig. 9], and examined the profiles of temperature and salinity at observation stations with type 1 and 2 samples [Fig. 10]. In generally, the type 2 samples were mainly from the coastal region and the type 1
samples were more located offshore. Meanwhile, the water columns with type 1 samples at the surface layers showed typical temperature and salinity profiles of stratified water columns while those with type 2 samples at surface layers were thoroughly mixed. In addition, with the exception of vertical mixing, river discharge, and especially the Yellow River which carries large loads of suspended sediment to the BS, may also affect the composition of particles in the estuary. Overall, the seasonal dynamics of hydrographic environments of these waters may change particle assemblages, which may further induce significantly different variability and distributions in P\textsubscript{Qbbe}(640) in different seasons; and caution should be used in regard to the associated optical properties.

Fig. 9. Distributions of pseudo-backscattering efficiency P\textsubscript{Qbbe}(640) in surface layers in late spring (a), late summer (b) and late autumn (c). Thick black lines denote the isograms with the value of 0.0134 which is defined as a threshold for classifying type 1 (< 0.0134) and type 2 samples (> 0.0134).

Fig. 10. Profiles of temperature (a) and salinity (b) for the observation stations showing type 1 samples in the surface layer. Profiles of temperature (c) and salinity (d) for observation stations showing type 2 samples in the surface layer. Green, blue and red lines denote samples collected during the late spring, late summer and late autumn, respectively.
4.2 Implications for remote sensing

Considerable $P_{bbe(640)}$ variability in the BS and YS consequently induced significant effects on backscattering properties [Figs. 5 and 6 and Table 2]. For all of the samples, the $P_{bbe(640)}$ value caused roughly 63.7% of $b_{bp}(640)$ variability. For $b_{bp}(640)$, although variability was mainly attributed to the AC or the TSM, the $P_{bbe(640)}$ data was still able to explain roughly 20.8% of the variability. More importantly, $P_{bbe(640)}$ clustering clearly differentiated the covariation patterns both for $b_{bp}(640)$ with ($\rho_\mu D_\lambda$)$^{-1}$ and for $b_{bp}(640)$ with AC and TSM. After classifying samples using the $P_{bbe(640)}$-based scheme, the effects of $P_{bbe(640)}$ on $b_{bp}(640)$ and $b_{bp}(640)$ was reduced. Moreover, especially the trends in the co-variation of $b_{bp}(640)$ and $b_{bp}(640)$ with biogeochemical properties could be more accurately described.

For type 2 samples, we found that the effects of $P_{bbe(640)}$ on the $b_{bp}(640)$ and $b_{bp}(640)$ decreased to roughly 28.5% and 8.0%, respectively. For Type 1 samples, although 60.9% and 61.2% of the variability in $b_{bp}(640)$ and $b_{bp}(640)$ was attributed to ($\rho_\mu D_\lambda$)$^{-1}$ and AC, respectively, due to weak correlations between $P_{bbe(640)}$ and $b_{bp}(640)$ and $b_{bp}(640)$, it is difficult to ascertain the effects of $P_{bbe(640)}$; and large proportions of $b_{bp}(640)$ and $b_{bp}(640)$ variability still remained unexplained. These observations may be related to the presence of non-spherical shaped particles. According to Mie theory, particles are assumed to be spherical in shape and to have material homogeneity. However, this assumption may not hold true for some particles and especially for some algal species that are complex in shape (e.g., chains, irregular structures, etc.) [10,11]. Meanwhile, as noted in section 2.3, there is a mismatch in some particles and especially for some algal species that are complex in shape (e.g., chains, irregular structures, etc.) [10,11]. Meanwhile, as noted in section 2.3, there is a mismatch in

As was expected, the remote sensing reflectance $R_{\rho}(640)$, which is strongly dependent on $b_{bp}(640)$, was also affected by clustering in $P_{bbe(640)}$, showing two obvious patterns in relationships between $R_{\rho}(640)$ and TSM [Fig. 7]. After we classified the samples using the $P_{bbe(640)}$-based scheme, co-variation trends of $R_{\rho}(640)$ with TSM were more accurately identifiable for the type 1 and type 2 samples, respectively, although considerable variability was still found in these relationships. Meanwhile, as is shown in Fig. 8, the TSM values of the two sample types showed clearly different correlations with $R_{\rho}(\lambda)$ between 400 and 700 nm. These observations imply that classification of particles or water types must be carefully considered when using or developing remote sensing algorithms for deriving TSM in the BS and YS. To more accurately understand water properties from satellite measurements, studies focusing on water classifications have been conducted in other regions [52,53]. Most classification methods are empirically based on the typical spectral characteristics of $R_{\rho}(\lambda)$ for different water types. By contrast, the classification scheme used in this study is proposed based on the different backscattering properties of particles guided by optical theory. Currently, $b_{bp}(\lambda)$ can be derived from satellite measurements using an inherent optical algorithm [54,55]. Meanwhile, our recent work has shown that the cross-sectional area AC can be estimated from $R_{\rho}(\lambda)$ [56], and both evaluations based on in situ and satellite data indicate promising performances of the algorithm. Once satellite $b_{bp}(\lambda)$ and AC are available, the classifications of particles based on the $P_{bbe}(\lambda)$ becomes feasible. Here, we need to point out that there are also some studies, such as Twardowski et al. [57] and Boss et al. [14], and the study of Chen et al. [20] in BS and YS, to suggest that particulate backscattering ratio (ratio of $b_{bp}(\lambda)$ to scattering coefficient) can be used to study particle composition. However, it must be admitted that the classification of particle based on particulate backscattering ratio may be not applicable to current ocean color satellite measurements, since retrieval of particulate
scattering coefficient is difficult from these measurements in theory. Although Roesler and Boss [58] proposed an algorithm for deriving attenuation from current satellite ocean color observations and Ibrahim et al. [59] also presented an inversion algorithm for attenuation from polarized radiometry measurements; the algorithm of Roesler and Boss [58] is actually based on the information of backscattering and absorption, and that of Ibrahim et al. [59] may only be applied in future ocean color missions.

We must note that the \( R_{s}(\lambda) \) values is not only regulated by particulate backscattering properties but also by the absorption of particles and colored dissolved organic matter (CDOM) [60]. This study mainly examined the effects of \( P_{Q_{bb}}(\lambda) \) at 640 nm on backscattering properties and thereby on \( R_{s}(\lambda) \), at which the absorption of particles and CDOM is negligible. However, absorption at blue bands and about 675 nm may be strong due to the strong absorption of phytoplankton. Therefore, the effects of \( P_{Q_{bb}}(\lambda) \) on \( R_{s}(\lambda) \) in visible regions with respect to the absorption properties must be further clarified in future works to thoroughly understand the potential effects of particle types on \( R_{s}(\lambda) \). Meanwhile, future studies will be conducted to investigate satellite applications of the \( P_{Q_{bb}}(\lambda) \)-based clustering scheme and the potential influences of \( P_{Q_{bb}}(\lambda) \) on ocean color remote sensing algorithms, and thereby to improve the algorithm for each particle type to achieve better estimations of water constitutes, such as TSM.

5. Conclusions

High levels of variability and clearly evident clustering were observed in pseudo-backscattering efficiency \( P_{Q_{bb}}(640) \) in the Bohai Sea and Yellow Sea. After analyzing \( P_{Q_{bb}}(640) \) variability levels during different seasons, we can confirm that the clustering of \( P_{Q_{bb}}(640) \) is generally seasonally stable. Consequently, a simple scheme is proposed for the classification of particle types. Our analysis of optical effects of \( P_{Q_{bb}}(640) \) shows that \( P_{Q_{bb}}(640) \) can significantly affect the backscattering coefficient \( b_{bp}(640) \), mass-specific backscattering coefficient \( b_{bp}^{*}(640) \) and remote sensing reflectance \( R_{r}(640) \). In particular, as a result of \( P_{Q_{bb}}(640) \) clustering, relationships between \( b_{bp}^{*}(640) \), \( b_{bp}(640) \) and \( R_{r}(640) \) and biogeochemical variables differ for different particle types, and discretion should be exercised when applying general bio-optical models that couple these relationships. After classifying using the scheme proposed in this study, the effects of \( P_{Q_{bb}}(640) \) on \( b_{bp}^{*}(640) \), \( b_{bp}(640) \) and \( R_{r}(640) \) were reduced, showing that co-variation trends of \( b_{bp}^{*}(640) \), \( b_{bp}(640) \) and \( R_{r}(640) \) with biogeochemical variables can be more accurately demonstrated. The findings of this study may help clarify the biogeochemical processes that couple particulate backscattering properties and the remote sensing of ocean color in the Bohai Sea and Yellow Sea. Moreover, future studies will explore the satellite applications of the \( P_{Q_{bb}}(640) \)-clustering scheme, and examine the effects of particle types on the whole spectral region of \( R_{s}(\lambda) \) thereby improve remote sensing algorithms.

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