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Characterizations of the light-scattering attributes of mineral particles in Lake Ontario and the effects of whiting

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ABSTRACT

Light-scattering attributes of minerogenic particles from the upper waters of Lake Ontario, collected lakewide from pelagic waters in late 2007 summer and early 2008 spring cruises and over the summer interval at a near-shore site in 2008, were characterized by scanning electron microscopy interfaced with automated image and X-ray analyses (SAX). SAX results were used to estimate minerogenic scattering and backscattering coefficients (b_m and $b_\mathrm{b,m}$) through Mie theory. Two minerogenic particle regimes with respect to light scattering were resolved: (1) clay mineral dominance and (2) dominance by 'whiting' ($CaCO_3$ precipitate) in late summer in portions of the lake. Clay minerals made noteworthy and important contributions to overall particulate scattering and backscattering coefficients (b_p and b_bp, respectively) in spring and early summer. Dramatic increases in values of b_p, and particularly b_bp, as well as decreases in Secchi disk depth (SD), were observed during whiting from the associated large increases in b_m and $b_\mathrm{b,m}$. Features of these events were the primary drivers of the spatial patterns in late summer and temporal differences observed for scattering and SD. Particles in the size range of 1–10 μm were responsible for minerogenic scattering during stratification, but those with sizes >10 μm made noteworthy contributions at certain sites during spring turnover. The credibility of the SAX-Mie estimates of b_m and $b_\mathrm{b,m}$ was supported by the extent of optical closure obtained with paired bulk measurements of overall b_p and b_bp (2007 summer cruise), and independent estimates of organic particulate scattering and backscattering through empirical bio-optical models.

Introduction

The process of light scattering is important in regulating apparent optical properties (AOPs, which depend on the geometry of the light field as well as the water mass characteristics) that are of fundamental concern for water quality and related monitoring issues, including water clarity (Davies-Colley et al., 2003; Effler et al., 2008) and remote sensing signals (Binding et al., 2007; Woźniak and Stramski, 2004). The total scattering and backscattering coefficients (b and b_b; symbols, along with acronyms, listed in Table 1) quantify central features of the scattering regime that represent integrations of the volume scattering function over angular ranges of 0–π and π/2–π, respectively (Kirk, 1994). These coefficients are described as inherent optical properties (IOPs), as they do not depend on the geometry of the light field. In contrast to the open oceans (case 1 waters; Morel and Prieur, 1977), inorganic (minerogenic) particles are generally important components of light scattering in case 2 waters, which include lacustrine systems (Kirk, 1985; Peng et al., 2009; Swift et al., 2006). Minerogenic particles have three general origins: terrigenous inputs (Kirk, 1985; Peng and Effler, 2007), resuspension (Peng and Effler, 2010), and autochthonous production (e.g., calcite; Vanderploeg et al., 1987). In case 2 waters, minerogenic particle scattering does not covary with that associated with phytoplankton (Mobley et al., 2004), and scattering is essentially entirely attributable to particles (i.e., contributions from water negligible; $b \approx b_\mathrm{p}$ and $b_\mathrm{b} \approx b_\mathrm{bp}$, subscript 'p' refers to particulate component).

To understand the drivers responsible for widely observed major variations in optical properties within individual systems and differences among systems and to identify the origins of responsible scattering particles, it is necessary to consider the contributions of a potentially diverse array of particle types (Stramski et al., 2001, 2007). These needs and goals cannot be met solely through use of common bulk measurements such as concentrations of chlorophyll *a* ([Chl]) and suspended particulate matter, which simply cannot provide adequate constituent resolution. Stramski et al. (2001, 2004, 2007) have advocated and advanced much greater partitioning in ocean studies through what they have described as the reductionist approach. Stramski et al. (2001) implemented the approach in a modeling analysis of IOPs in the ocean that focused primarily on the partitioning of multiple planktonic particle types, while also including the effects of minerogenic particles (albeit with limited partitioning). Greater emphasis on increased partitioning of minerogenic particles is warranted for lacustrine systems because of their greater relative importance in these case 2 waters and potentially different origins (Peng and Effler, 2010; Peng et al., 2009).

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Table 1
Acronyms, abbreviations, and symbols.

Acronyms and abbreviations	
AOPs	apparent optical properties
ASP	aspect ratio
[Chl]	chlorophyll *a* concentration (mg m^{-3})
DIPA	differential individual particle analysis
IOPs	inherent optical properties
IPA	individual particle analysis
PA	projected area of a particle (m^2)
PAV_m	projected area concentration of minerogenic particles (m^{-1})
PSD	particle size distribution
SAX	scanning electron microscopy interfaced with automated image and X-ray analyses
SD	Secchi disk transparency depth (m)
Symbols	
$a(\lambda)$	spectral absorption coefficient (m^{-1}); wavelength (λ) sometimes omitted
$b(\lambda)$	spectral scattering coefficient (m^{-1})
b_p (b_m or b_o)	particulate (mineral or organic component) scattering coefficient (m^{-1})
$b_b(\lambda)$	spectral backscattering coefficient (m^{-1})
b_{bp} ($b_{b,m}$ or $b_{b,o}$)	particulate (mineral or organic component) backscattering coefficient (m^{-1})
$c(\lambda)$	spectral attenuation coefficient (m^{-1})
d	size of a particle (μm)
$d_{50,bm}$ and $d_{50,bbm}$	the 50th percentile (or median) sizes of mineral scattering and backscattering coefficients (μm)
m	relative (to water) complex refractive index of a particle, $n-n'i$,
n	real part of m
n'	imaginary part of m
N_m	number of mineral particles per unit volume of water (m^{-3})
Q_{bm} and Q_{bbm}	scattering and backscattering efficiency factors of a mineral particle
V	water sample volume for SAX (m^{-3})
λ	light wavelength in vacuum (nm)

Application of an individual particle analysis (IPA) technique, scanning electron microscopy interfaced with automated image and X-ray analysis (SAX), has recently supported direct estimates of contributions by minerogenic particles to b_p and b_{bp} (Peng and Effler, 2007, 2010; Peng et al., 2007, 2009). SAX directly measures the light-scattering features of individual minerogenic particles and overall populations, including number concentration (N), the particle size distribution (PSD), composition, and particle shape. SAX results have been used to successfully make direct, or so-called "forward" (Sullivan et al., 2005), estimates of minerogenic components of b_p and b_{bp} (i.e., b_m and $b_{b,m}$) by providing inputs for Mie theory calculations of the scattering and backscattering efficiency factors (Q_{bm} and Q_{bbm}, dimensionless) for individual particles, as described for b_m below

$$b_m(\lambda) = \frac{1}{V}\sum_{i=1}^{N_m} Q_{bm,i}(m_i,\lambda,d_i)PA_{m,i} \tag{1}$$

where λ is the wavelength of light, N_m is the number of minerogenic particles per unit volume of water, $PA_{m,i}$ is the projected area of minerogenic particle i, and V is the sample volume. The efficiency factors of particles depend on particle size (d), the complex refractive index (m, function of composition), and wavelength. The SAX-Mie approach can support partitioning of b_m and $b_{b,m}$ into contributions from multiple minerogenic particle types, such as clay minerals, quartz, and calcite, from the chemical characterization (X-ray) capabilities of SAX (Peng et al., 2007), consistent with the reductionist approach.

The credibility of the SAX-Mie-based forward estimates of b_m and $b_{b,m}$ has been obtained through successful pursuits of optical modeling closure with bulk measurements of scattering. These initiatives have been made for two types of systems: (1) those where minerogenic particles dominate (i.e., phytoplankton contributions can be ignored; Peng and Effler, 2007, 2010) and (2) those where phytoplankton makes noteworthy contributions to scattering (Peng et al., 2007, 2009). The first of these two types represents a more direct test of the SAX-Mie estimates. The second type has required paired independent estimates of phytoplankton contributions through adoption of empirical bio-optical models that are based on [Chl] observations (Huot et al., 2008; Loisel and Morel, 1998; Morel and Maritorena, 2001). Despite limitations in these bio-optical models, their applications in combination with the SAX-Mie-based estimates of b_m and $b_{b,m}$ have supported good overall closure with bulk measurements as well as representative partition of b_p and b_{bp}, for example,

$$b_p = b_o + b_m \tag{2}$$

where b_o is the magnitude of scattering associated with organic (primarily phytoplankton) particles, as estimated from a bio-optical model.

These efforts to implement the reductionist approach and demonstrate a degree of optical closure have included the widely different conditions of Lake Superior (Peng et al., 2009) and the western basin of Lake Erie (Peng and Effler, 2010), and are supporting an initiative to develop more robust remote sensing algorithms for the Laurentian Great Lakes (O'Donnell et al., 2010). Temporal coverage was limited (single cruises) and clay minerals were the dominant minerogenic component in both of these cases. Increased temporal coverage, cases of more diverse chemical classes of minerogenic particles contributing significantly, and inclusion of the other Great Lakes, have been recommended in extending this approach and to support the improved remote sensing initiative (Peng and Effler, 2010). Representation of the effects of "whiting", the precipitation of calcite ($CaCO_3$), that is common to many hard water lakes (Homa and Chapra, 2011), would be a particularly valuable test of the SAX-Mie and overall closure approaches, with specific relevance to the optical regimes of Lake Michigan, Lake Erie, and Lake Ontario (Binding et al., 2007; Strong and Eadie, 1978).

The goals of this paper are to (1) document the light-scattering attributes of minerogenic particle populations in the near-surface waters of Lake Ontario based on SAX characterizations, (2) report forward method estimates of b_m and $b_{b,m}$ through the SAX-Mie approach, (3) pursue optical closure with bulk measurements, based on these estimates of b_m and $b_{b,m}$, in combination with the application of bio-optical models to represent organic particulate components, and (4) advance the reductionist approach in lacustrine systems through resolution of the important effects of multiple types of minerogenic particles. Spatial differences are described, and temporal coverage of the SAX characterizations is expanded compared to earlier efforts with these approaches. The dramatic effects of the whiting phenomenon on scattering encountered in this study are documented. Insights into whiting events based on the SAX characterizations are presented and implications for AOPs are considered.

Methods

Study area, surveys, field measurements, and sampling

This study included field measurements and sampling at pelagic sites in Lake Ontario as well as a single near-shore location, 7 km offshore of Oswego Harbor (Fig. 1). The pelagic sites surveyed in this work, representing a subset of those included in US EPA's routine monitoring (R/V *Lake Guardian*), extended over much of the open waters of the lake. Site designations from the EPA program were retained here. Sampling and measurements reported here for the lake's pelagic near-surface waters were made as part of two cruises, over 10–12 August, 2007 and 24–25 April, 2008. The near-shore measurements were made from a smaller vessel on six occasions over