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Characterization of Florida red tide aerosol and the temporal profile of aerosol concentration

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Abstract

Red tide aerosols containing aerosolized brevetoxins are produced during the red tide bloom and transported by wind to coastal areas of Florida. This study reports the characterization of Florida red tide aerosols in human volunteer studies, in which an asthma cohort spent 1 h on Siesta Beach (Sarasota, Florida) during aerosolized red tide events and non-exposure periods. Aerosol concentrations, brevetoxin levels, and particle size distribution were measured. Hourly filter samples were taken and analyzed for brevetoxin and NaCl concentrations. In addition, the aerosol mass concentration was monitored in real time. The results indicated that during a non-exposure period in October 2004, no brevetoxin was detected in the water, resulting in non-detectable levels of brevetoxin in the aerosol. In March 2005, the time-averaged concentrations of brevetoxins in water samples were moderate, in the range of 5–10 µg/L, and the corresponding brevetoxin level of Florida red tide aerosol ranged between 21 and 39 ng/m³. The temporal profiles of red tide aerosol concentration in terms of mass, NaCl, and brevetoxin were in good agreement, indicating that NaCl and brevetoxins are components of the red tide aerosol. By continuously monitoring the marine aerosol and wind direction at Siesta Beach, we observed that the marine aerosol concentration varied as the wind direction changed. The temporal profile of the Florida red tide aerosol during a sampling period could be explained generally with the variation of wind direction.

Keywords

red tide; aerosol; brevetoxin

1. Introduction

Karenia brevis (*K. brevis*) is a marine dinoflagellate responsible for the Florida red tide that forms almost annually in the Gulf of Mexico. *K. brevis* produces polyether brevetoxins that cause neurotoxic shellfish poisoning. Red tide aerosols are airborne marine particles containing brevetoxins. Environmental exposure of inhaled red tide aerosol may cause respiratory symptoms. During two red tide events in Florida in 1999, brevetoxin levels in air and seawater

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were measured, and personal interviews and pulmonary function tests were conducted on people before and after they visited Florida beaches (Backer et al., 2003). During moderate and high exposure periods, 36 ng/m³ or 80 ng/m³ of brevetoxin, respectively, were detected in the air. Lower respiratory tract symptoms (e.g., tightness of chest, wheezing, and shortness of breath) were reported by 8% of the people who had no/low exposure, 11% who had moderate exposure, and 28% who had high exposure. Upper respiratory symptoms (e.g., eye and throat irritation, nasal congestion, and cough) were also increased in the moderate- and high-exposure groups. We also found in the fall of 2000 during a red tide event near Corpus Christi, Texas, that low air concentrations of brevetoxins in the range of 3–4 ng/m³ initiated symptoms in the upper respiratory tract, including cough, irritation in the throat, and itchy eyes (Cheng et al., 2005a).

Lifeguards and other people working on the beaches may be exposed to brevetoxin during red tide events. A study of healthy lifeguards working on southwest Florida beaches during a 2001 red tide event (brevetoxin concentrations of 1–49 ng/m³) and in 2002 when no red tide was reported, revealed increased upper respiratory symptoms during the exposure periods (Backer et al., 2005). However, aerosolized brevetoxins did not affect the lifeguards' measured pulmonary functions whether or not they exercised. In an experimental sheep model of asthma, inhalation challenge with aerosolized brevetoxins (as well as pure brevetoxins 2 and 3) caused a significant and rapid increase in airway resistance (Abraham et al., 2005). They also showed that the response of the animal exposed to the whole brevetoxins was similar to those of pure brevetoxin 2 and 3.

Florida red tide aerosols are marine aerosols containing airborne brevetoxins (Pierce et al., 1989). Sea-salt aerosols, the major component of marine aerosols, are produced at the sea-air interface by mechanisms powered by wind energy, including breakup of bubbles from whitecap waves (Blanchard, 1983; Fitzgerald, 1991). Analysis of aerosol size distribution for brevetoxin and NaCl content indicated that the Florida red tide aerosol consisted of coarse particles with a mass median aerodynamic diameter (MMAD) in the range of 5–12 µm (Cheng et al., 2005b). The consistency of particle size distributions analyzed for sea-salt and brevetoxins indicated that brevetoxin appears to be a component of marine aerosol particles associated with sea salts (Cheng et al., 2005b).

Attempts to assess brevetoxin exposure to people on the beach have been stymied by unexpected environmental concentrations of brevetoxins (e.g. low levels of brevetoxins in the aerosol when there clearly were red tides). A previous study indicated that onshore wind propelled the red tide aerosol inland and that the wind direction changed during the sampling day (Pierce et al. 2003). The purpose of this study was to investigate the influence of wind direction on the temporal profile of the Florida red tide aerosol measured at Siesta Beach. Detailed characterization of Florida red tide aerosols included the concentrations, brevetoxin levels, and particle size distribution. Hourly filter samples were taken and analyzed for brevetoxin and NaCl concentrations. In addition, real-time monitoring of the aerosol mass concentration was also reported and used to construct temporal profiles of aerosol concentration. Wind direction was monitored in real-time fashion and was used to correlate with the aerosol concentration. The relative aerosol concentration as a function of wind direction was used to establish a wind direction factor, providing a important parameter for exposure assessment of the aerosolized brevetoxins.

2. Materials and Methods

2.1 Field sampling study

The exposure studies of red tide aerosol were conducted at Siesta Beach (Sarasota, Florida). Aerosol measurements were performed between March 11 and March 14, 2005, during a

Florida red tide episode as well as during periods between October 16 and October 18, 2004, when brevetoxin levels in the water and air were below the limit of detection (LOD). The detailed study design for the epidemiological study has been described (Fleming et al., 2005, 2007).

Air samplers were set up along Siesta Beach to collect marine aerosols. The entire environmental sampling methods and analysis have been reported elsewhere (Cheng et al., 2005b, 2005c; Pierce et al., 2005). High-volume air samplers (Model G2000H, Andersen Instruments, Smyrna, GA) were placed near a lifeguard tower about 50 m from the water to collect large quantities of material for analysis of brevetoxins. The samplers were located in an area where the study participants spent the time on the beach. One sampler was used to collect hourly airborne particles in a filter substrate for total aerosol concentration, whereas other samplers housed a five-stage, high-volume cascade impactor (Model SA235, Andersen Instruments) for total concentration as well as particle size distribution during approximately 7–8 h of exposure time. Cellulose filters (15 cm x 14 cm) (Filter Paper 41, Whatman International Ltd., Florham Park, NJ) were used as the filter and impactor collection substrates. The sampling time usually started between 9:00 a.m. and 5:00 p.m. when the study volunteers reported to Siesta Beach. The collected samples represented the time-integrated ambient concentration and particle size in the sampling area.

Seawater samples were collected in 1-L glass bottles three times each day (8:30 a.m., 12:00 p.m., and 4:00 p.m.) from the surf zone adjacent to each air sampler location to indicate changes in seawater cell counts and toxin composition throughout each day. A 20-mL sub-sample was collected from each bottle and fixed with Utermohls solution for microscopic identification and enumeration of *K. brevis* cells. The remaining water sample was processed for brevetoxin analysis by enzyme-linked immunosorbent assay (ELISA) and for verification by liquid chromatography-mass spectrometry (LC/MS) (Pierce et al., 2005).

2.2 Real-time monitoring of marine aerosols

The total mass concentration of the marine aerosol was monitored continuously using a DataRAM real-time aerosol monitor (Monitoring Instruments for the Environment, Inc., Bedford, MA). This optical instrument measures particles in the concentration range of 0.1 $\mu\text{g}/\text{m}^3$ –400 mg/m^3 . The instrument was placed near a high-volume sampler and weather station.

2.3 Monitoring of weather conditions

A portable, solar-powered weather station was deployed at Siesta Beach near the high-volume air sampler to determine wind speed and direction, temperature, and relative humidity (Complete Weather Station, Davis Instruments, Hayward, CA). It continuously monitored the weather conditions during the sampling period, and the data were downloaded daily. The wind direction measured at Siesta Beach included 16 quadrants (E, W, N, S, NE, SE, SW, NW, NNE, ENE, ESE, SSE, SSW, WSW, WNW, and NNW) as shown in Fig. 1.

2.4 Analysis of air samples

After collection, the high-volume impactor substrates were stored at -20°C . Filters were removed from the cold storage and equilibrated to room temperature prior to extraction. First, substrates were cut into several sections. Approximately 3.0 cm^2 were used for ELISA analysis for total concentration of brevetoxins and related compounds. Approximately 90- cm^2 of samples were extracted and analyzed by a LC/MS, and ~90- cm^2 sections (cut with laboratory scissors) of cellulose filter substrates were used for NaCl analysis.

The extraction and analyses of filter samples for LC/MS analysis of brevetoxins have been described in detail (Cheng et al., 2005c). A portion of the cellulose substrate was extracted with 25 mL of ionic strength adjustor solution, and then its sodium content was determined by Sodium Ion-Selective Electrodes (Fisher Scientific, Denver, CO). The standard curve of this method was established by spiking known amounts of NaCl in the cellulose filter, and then extracting it with the same procedure. The standard curve was linear in the range of 1–1000 $\mu\text{g/mL}$ of NaCl. Cellulose filters have low sodium background, so therefore the detection limit of this method was 1 μg .

3.0 Results and Discussion

3.1 Environmental conditions

Table 1 lists the environmental conditions recorded at Siesta Beach during the 2004 and 2005 field sampling studies. The data show that the temperature and relative humidity were stable during each sampling day, except for the relative humidity on October 17, 2004. The averaged daily wind speed had a coefficient of variation between 15 and 61%. On October 16 and 17, 2004, the wind was mostly offshore, but on October 18, the wind direction was onshore most of the time. In the March 11–14, 2005, sampling period, the wind direction was onshore most of the time.

3.2 Time-averaged aerosol concentration and size distributions

Table 2 lists the time-averaged concentrations of brevetoxin in water and in aerosols obtained for each sampling day. In the October 2005 sampling period, the brevetoxin concentrations in water were below the limit of detection ($\text{LOD} = 0.03 \mu\text{g/L}$), resulting in a non-detectable brevetoxin concentration for the red tide aerosol of $< 0.2 \text{ ng/m}^3$. In March 2005, the time-averaged brevetoxin concentrations in water samples were moderate, in the range of 5–10 $\mu\text{g/L}$ (LC/MS analysis), and the corresponding brevetoxin concentrations of Florida red tide aerosol were between 21 and 39 ng/m^3 (LC/MS analysis).

Fig. 2 shows an example of particle size distribution of brevetoxins in Florida red tide aerosol measured with the high-volume impactor on March 11, 2005. A single-mode lognormal size distribution was used to fit size data. The MMAD and the geometric standard deviation (GSD) calculated with the nonlinear curve fitting procedure of SigmaPlot (SSPL, Chicago, IL) were 6.70 μm and 1.90, respectively. The MMAD and GSD of the brevetoxin size distribution for each sampling date during the March 2005 study are also listed in Table 2. The size distributions were similar to that reported previously (Cheng et al., 2005a).

3.3 NaCl concentration of marine aerosols

Figs. 3 and 4 show the temporal variation in NaCl and brevetoxin concentrations taken hourly using the high-volume sampler for the sampling periods of October 16–18, 2004, and March 11–14, 2005, respectively. As discussed in section 3.2, the October 2004 aerosol samples were collected when there was no red tide. Also presented in the figures are the total aerosol concentration taken with a real-time aerosol monitor (DataRAM) at the same sampling location during the same time period.

Fig. 3 shows that during the October 16–17 sampling period, the aerosol concentrations were less than 15 $\mu\text{g/m}^3$ most of time. During these sampling periods the wind directions were mostly offshore (in the direction of NE to NW for October 16 and ESE to N on October 17). On October 18 the aerosol concentration was quite low in the morning and started to increase between 10:30 and 11:00 a.m. Then the aerosol concentration continued to increase and reached levels of $> 40 \mu\text{g/m}^3$. The changes in aerosol concentration coincided with changes in wind direction from offshore (ESE) to onshore (SW) around 11:00 a.m., indicating the importance of wind

direction on the marine aerosol concentration at the beach. No brevetoxin was detected during the October 16–18, 2004, sampling period because there was no red tide bloom, and no brevetoxin was detected in the water samples.

Fig. 4 shows the hourly concentration of brevetoxin and NaCl concentrations obtained in the high-volume filter sample collected at Siesta Beach as a function of time for the period of March 11–14, 2005, during a Florida red tide episode. The total aerosol concentration from the DataRAM is also shown in the figure. These data showed that NaCl and the total aerosol concentrations are in good agreement. The temporal profile of the brevetoxin concentration also tracked well with the total aerosol concentration, although the brevetoxin concentration was on the order of 0.01% of the total aerosol concentration. Also included in the figures are the wind direction and speed information. On March 11, the wind was onshore in the direction of SW to W, resulting in a relatively high aerosol concentration (Figure 4A). On March 13 and 14, the wind direction was generally onshore in one direction of SSE to S, resulting in a more uniform aerosol concentration profile (Figure 4C and D). On March 12 between 9:00 and 11:00 a.m., the off shore wind was blowing from N to NE, and the NaCl and total aerosol concentrations were at a background level of $<20 \mu\text{g}/\text{m}^3$. The brevetoxin concentration was non-detectable. Between 11:00 a.m. and 12:00 p.m., the wind direction shifted from NE to NW, and all the aerosol concentrations started to increase slightly. Between 12:00 noon and 5:30 p.m., the wind direction was onshore between W and SSW, and the aerosol concentration remained high (Figure 4B).

Our data showed that the aerosol concentrations of NaCl measured with the high volume sampler and the total aerosol concentration measured with the DataRAM were in good agreement. The figures also showed that the NaCl aerosol concentrations were, in general, slightly lower than the aerosol mass concentrations taken with the DataRAM.

As a further comparison of these two sets of measurements, the mean value of the filter sampler and DataRAM aerosol concentrations within the same hourly sampling period of the high-volume sample was calculated. Fig. 5 shows the plot of aerosol concentrations taken from the same sampling period. A linear regression analysis of the data shows the following best-fit relationship:

$$C_{\text{NaCl}} (\mu\text{g}/\text{m}^3) = -5.5 + 0.92 C_{\text{DataRAM}} (\mu\text{g}/\text{m}^3) \quad (1)$$

Where C_{NaCl} and C_{DataRAM} are NaCl and mass concentrations measured by the DataRAM, respectively. The coefficient of determination is $R^2 = 0.845$, indicating that equation (1) can relate the NaCl concentration with the mass concentration reported by the DataRAM instrument.

The DataRAM reading was based on an optical technique, and its calibration depended on the optical properties of the aerosol (Cheng et al., 1995). A good correlation in equation (1) indicated that the DataRAM was a useful monitor for marine aerosol.

3.4 Aerosol concentration and wind direction

The data shown in the previous section demonstrated that the Florida red tide aerosol concentration measured at Siesta Beach was influenced by the wind direction. The aerosol concentration was high when the onshore wind was blowing toward the beach from SSE, S, W, to WNW, whereas the aerosol concentration decreased with the offshore wind from the NE, N, to NW (Fig. 1). To further quantify the wind direction factor on the aerosol concentration measured at Siesta Beach, we plotted the aerosol concentration measured with the DataRAM in micrograms per cubic meter as a function of wind direction for the October

16–18, 2004, sampling period (Fig. 6). The aerosol concentration in each direction was the mean value ($\pm$ SD) for the October 2004 sampling period. Fig. 7 presents this relationship on a polar chart showing the Siesta Beach coastline. This figure shows that on the average, the highest mean aerosol concentration was recorded at approximately $35 \mu\text{g}/\text{m}^3$ with a SW onshore wind and then at approximately $30 \mu\text{g}/\text{m}^3$ in the SSW direction. The reason for the maximum aerosol concentration when the wind was from the SW seems to be that the angle was at 90° to the Siesta Beach coastline. The aerosol concentration was lower when the wind was from the S, W, and SSE. For the offshore winds from the NW, NWW, N, NNE, NE, ENE, E, and ESE, the aerosol concentrations were approximately at or below $10 \mu\text{g}/\text{m}^3$. A similar graph was obtained for the sampling time of March 11–14, 2005.

The relative aerosol concentration in each wind direction of the October 16–18, 2004 was calculated as the mean concentration in each wind direction divided by the maximum mean aerosol concentration at the SW direction. Similar relative concentrations for the March 11–14, 2005, sampling period were also obtained. Fig. 8 shows the relative concentration, which is the average of the two values, obtained for the October 2004 and March 2005 sampling periods, as a function of wind direction. Based on the relative concentration, we defined the wind direction index (WDI) as 1 for the SW, indicating 100% onshore, 0.8 for SSW and WSW, 0.6 for S and W, 0.4 for SSE and WNW, and 0.2 for SE and NW. For the offshore winds of N, E, NNE, ENE, ESE, and NNW, the WDI was set at 0 (Figure 8). Using this WDI, the mean values of the wind direction for each sampling day was calculated (Table 1). This WDI was based on the aerosol concentration measured at the beach containing marine aerosol and the related wind direction.

4. Conclusions

We have shown the temporal variation of aerosol mass, NaCl and brevetoxin concentrations during the two sampling periods. The concentration profiles for the aerosol mass, NaCl, and brevetoxin were in good agreement, indicating that NaCl and brevetoxin are components of the red tide aerosol. By monitoring the marine aerosol and wind direction continuously at Siesta Beach, we observed that the marine aerosol concentration varied as the wind direction changed. Wind direction is a key parameter which is important in the transport of aerosolized brevetoxins from the red tide bloom to the beach. The temporal profile of the Florida red tide aerosol during a sampling period could be explained generally with variation of wind direction. From these observations, we showed that the marine aerosol concentration increased to the maximum when the wind was onshore at a 90° angle to the coastline. Based on the relative marine aerosol concentration measured at Siesta Beach, we defined a WDI with 1 indicating a 100% onshore direction, 0 indicating an offshore wind, and a value of between 1 and 0 indicating a partial onshore wind. The average and geometric standard deviation of the daily WDI can be used to document the fraction of time the wind direction is onshore as well as the variation of the wind direction. The WDI is a useful parameter for human exposure to aerosolized brevetoxins and can be used potentially to establish predictive models for exposure concentrations at Siesta Beach.

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Fig. 1.
A map of Siesta Beach showing the wind direction.

Fig. 2.

Particle size distributions of individual brevetoxins and the total amount of brevetoxins (LC/MS analysis). The curve is a lognormal distribution fitted to the total brevetoxins.

The MMAD and the geometric standard deviation (GSD) of the lognormal distribution were 6.70 μm and 1.90, respectively

DataRAM and NaCl Concentrations

Fig. 3.

Comparison of NaCl concentration in air samples obtained using a high-volume sampler and the aerosol mass concentration measured by the DataRAM during the October 16–18, 2004, sampling period. The bar graph is the NaCl concentration, and the curve is the aerosol mass concentration measured by the DataRAM. The brevetoxin concentrations were below the LOD.

Figure 4 (A)

Figure 4(B)

Figure 4(C)

Figure 4 (D)

Fig. 4.

Comparison of NaCl and brevetoxin concentration in air samples obtained using a high-volume sampler and aerosol mass concentration measured by the DataRAM during the March 11–14, 2005 (A,B,C,D) sampling period. The solid bar graph is the NaCl concentration, and the dashed bar graph is the brevetoxin concentration. The real time aerosol mass concentration measured with the DataRAM is also plotted in solid curve. The upper chart is the temporal profile of wind direction and wind speed.

Fig. 5. Correlation of NaCl concentration in air samples obtained using a high-volume sampler and aerosol mass concentration measured by the DataRAM. The empty circles are data obtained during the October 2004 sampling period. The filled circles are data obtained during the March 2005 sampling period.

Fig. 6. Aerosol concentration (mean and SD) as a function of wind direction measured at Siesta Beach using a DataRAM real-time aerosol monitor during the October 16–18, 2004, sampling dates.

Fig. 7. Aerosol concentration as a function of wind direction measured at Siesta Beach and plotted in a polar coordinate during the October 16–18, 2004, sampling period.

Fig. 8.

Relative aerosol concentration (mean of two sampling periods) as a function of wind direction measured at Siesta Beach during the October 16–18, 2004, and March 11–14, 2005, sampling periods. Based on the relative concentration, a wind direction index is shown also in the figure

Table 1

Daily Environmental Conditions during Field Studies at Siesta Beach

Date	Temperature (C)	Humidity (%)	Averaged Wind Speed (Km/h)	Wind Direction ^a
16-Oct-04	22.3 ± 2.7	50.4 ± 7.4	8.8 ± 4.5	0.07 ± 0.10
17-Oct-04	24.3 ± 3.4	53.6 ± 15.5	5.8 ± 3.5	0.05 ± 0.18
18-Oct-04	25.7 ± 1.9	76.6 ± 5.2	9.8 ± 1.8	0.57 ± 0.44
11-Mar-05	18.8 ± 0.4	72.0 ± 2.8	9.8 ± 5.3	0.76 ± 0.18
12-Mar-05	18.7 ± 0.8	34.0 ± 7.3	9.8 ± 2.9	0.50 ± 0.40
13-Mar-05	20.0 ± 0.4	88.9 ± 1.6	15.3 ± 2.3	0.51 ± 0.10
14-Mar-05	20.6 ± 0.2	94.9 ± 1.2	12.2 ± 2.4	0.40 ± 0.09

^a 1 for SW, indicating 100% onshore, 0.8 for SSW and WSW, 0.6 for S and W, 0.4 for SSE and WNW, and 0.2 for SE and NW

Table 2

Daily Brevetoxin Concentration and Size Distribution of the Time-averaged Samples

Date	Brevetoxin Water Concentration (µg/L)	Brevetoxin Air Concentration (ng/m ³)	MMAD (µm)	GSD
16-Oct-04	<LOD ^a	<LOD ^b	NA	NA
17-Oct-04	<LOD	<LOD	NA	NA
18-Oct-04	<LOD	<LOD	NA	NA
11-Mar-05	5.01±1.3	38.64±15.3	6.56±0.20	1.86±0.06
12-Mar-05	7.38±2.0	26.71±10.7	4.84±0.23	2.02±0.03
13-Mar-05	10.15±1.2	20.60±6.3	6.41±0.12	1.80±0.03
14-Mar-05	8.07±1.8	21.73±12.0	7.04±0.46	1.76±0.01

^aLOD = 0.03 µg/L (LC/MS analysis)^bLOD = 0.2 ng/m³ (LC/MS analysis)