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The Influence of Air Mass Origin on the Wet Deposition of Nitrogen to Tampa Bay, Florida

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THE INFLUENCE OF AIR MASS ORIGIN ON THE WET DEPOSITION OF
NITROGEN TO TAMPA BAY, FLORIDA

by

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A thesis submitted in partial fulfillment
of the requirements for the degree of
Master of Science
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THE INFLUENCE OF AIR MASS ORIGIN ON THE WET DEPOSITION OF
NITROGEN TO TAMPA BAY, FLORIDA

Ronald David Smith Jr.

ABSTRACT

Atmospheric deposition of nitrogen has been implicated in the destruction of seagrass beds and in the decline of water quality of Tampa Bay, Florida. The objective of this research was to determine the tendency for air masses of different origins to wet-deposit nitrate and ammonium species to the bay.

Precipitation chemistry data was obtained via the NADP AIRMoN Gandy Bridge monitoring site for the period of 1 August 1996 through 31 December 2000. Rainfall events were classified by using the NOAA HYSPLIT trajectory model, precipitation chemistry data, and tropical storm history data. Average nitrate and ammonium concentrations and nitrogen fluxes were calculated based upon the chosen categories.

The average annual nitrogen flux for nitrate and ammonium were 2.1 kg/ha/yr and 1.4 kg/ha/yr, respectively. For trajectory-classified data, the lowest nitrate and ammonium nitrogen fluxes were observed with air masses from the west and south, over the Gulf of Mexico. The highest ammonium nitrogen flux was seen from trajectories from the east, while local trajectories demonstrated the highest average nitrate nitrogen flux. For chemically-classified

data, the highest nitrate and ammonium fluxes were associated with the local combustion classification. Rainfall from tropical weather systems deposited lower average nitrate nitrogen fluxes than non-tropical events, but ammonium nitrogen fluxes were the same between tropical and non-tropical precipitation.

Even the events representing the cleanest air masses contributing precipitation to Tampa Bay had nitrate and ammonium concentrations more than two times the background concentrations associated with the northern hemisphere.

INTRODUCTION

An estuary is a semi-enclosed coastal area where fresh water outflow meets seawater. Many species of fish, crustaceans, birds, and other coastal species rely on the special properties of estuaries for some critical stage of their lives. Estuaries are often used as a safe zone for spawning and as a nursery for young organisms. The unique attributes of these zones of transition include the dilution of salinity from seawater, the buffering of tidal and wave action, and the exclusion of large predatory species, which makes the area more hospitable for the species that utilize this ecosystem.

Tampa Bay Estuary is located along the western coast of the state of Florida. It is bordered by Hillsborough County on the east, Pinellas County on the west, and Manatee County on the south. It is the largest open-water estuary in the State of Florida spanning nearly 101,000 hectares (ha) at high tide. The Bay is bordered by more than 2 million people and has an extreme diversity of land use within its watershed. Tampa Bay Estuary watershed land use consists of about 40% undeveloped, 35% agricultural, 16% residential, and 9% commercial and mining operations (Greening, 2001). This adds up to a great amount of pressure placed on this important ecosystem.

Like all estuaries, Tampa Bay is an important link in the life cycles of many commercially important fish, shellfish, and crustaceans. This is also essential

habitat for both native species and migratory birds using this area on a seasonal basis. As many as 400,000 pairs of nesting sea birds roost along the shores of this estuary. Other species that utilize the waters of the Tampa Bay Estuary include species of sea turtles, manatees, and dolphins (Tampa Bay Estuary Program, 1996).

Along with the animals that frequent the Tampa Bay Estuary, the bay is also an essential habitat for several species of seagrasses that attract and protect many animal species throughout their lives. Turtle grass (*Thalassia testudinum*), shoal grass (*Halodule wrightii*), and manatee grass (*Syringodium filiforme*) can all be found in various regions of Tampa Bay and serve important functions pertaining to the overall health of the bay. Besides providing food and shelter for fish, crustaceans, and mollusks, they also clarify the water column by settling sediments and attenuating wave action. The health of these seagrass beds has been cited as an indicator for the overall health of the entire bay (Water Resources Atlas of Florida, 1998).

In the 1930's, seagrasses are estimated to have covered 31,000 hectares of Tampa Bay. However, by 1982 only 8,800 hectares of seagrasses remained (Clement et al., 2001). Figure 1 illustrates the decline of seagrasses in Tampa Bay since the 1950s. Many acres of seagrasses have been lost due to direct physical loss of habitat from dredge and fill projects as well as other projects undertaken for the maintenance of navigable waters in the bay. However, by far the largest culprit of seagrass decline during this time period has been the introduction of nutrients into Tampa Bay.

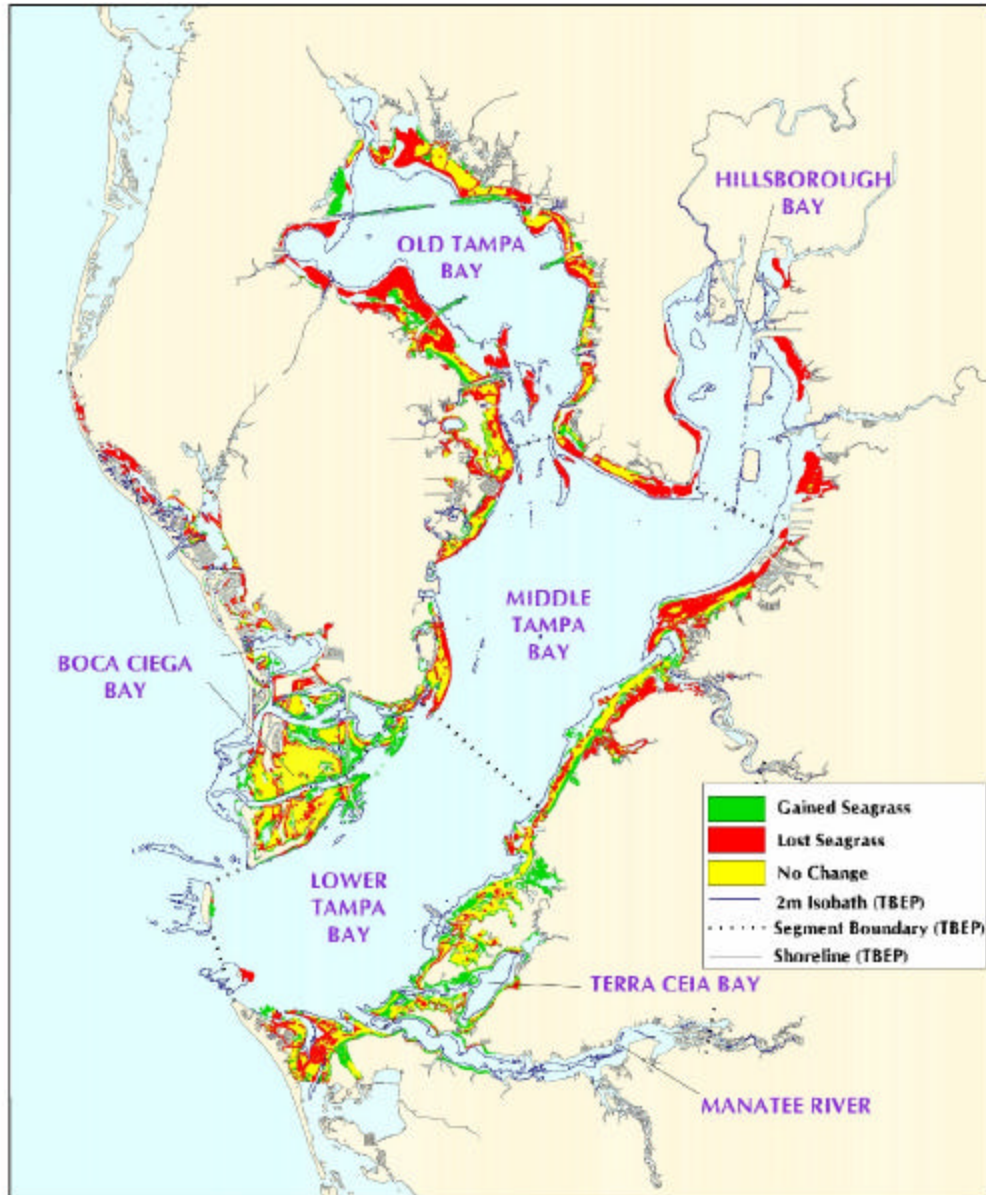


Figure 1. Seagrass coverage changes in Tampa Bay from 1950 to 1990. Reprinted from Tampa Bay Estuary Program (2000).

Since primary production in Tampa Bay is, with respect to nutrients, nitrogen-limited, the main chemical species of interest are those which contribute excess biologically-available nitrogen to this ecosystem (Howarth, 1988).

Therefore, it is important to understand and effectively regulate the introduction of biologically-available nitrogen to the Tampa Bay Estuary ecosystem.

Inputs of nitrogen have been implicated in the degradation of waterways for several decades. After the addition of nitrogen to marine environments was discovered as a causative factor for the decline of seagrasses in estuaries around the United States, point sources of nitrogen and other nutrients were identified and stricter regulations about their discharge were instituted. These early regulations focused on sources such as under-treated discharge from sewage treatment facilities. Now the focus has shifted to many non-point sources that are often more difficult to locate and are therefore more difficult to regulate.

Zarbock et al. (1996) identified five categories of potential major sources of total nitrogen loading to Tampa Bay. These categories encompassed non-point sources including stormwater runoff, point sources, atmospheric deposition, groundwater and springs, and natural nitrogen losses. This study estimated that atmospheric deposition accounted for approximately 29% of the nitrogen contributions to Tampa Bay for the period from 1992-1994, and that sections of the Bay with larger surface areas, like Old Tampa Bay west of the city of Tampa, received a proportionately higher contribution of nitrogen from atmospheric deposition.

Though the watershed for Tampa Bay is approximately 5700 km², the airshed, defined the region from which 75% of the deposition to the watershed is thought to originate (East Coast Atmospheric Resource Alliance, 1995), may

encompass a region that is three times the area of the state of Florida. Because of the multitude of nitrogen sources present in the Tampa Bay airshed, and the potential for any emissions within this zone to affect primary production in Tampa Bay, it is important to know the relationship between emission source locations and the amount of nitrogen physically entering the bay in order to facilitate the recovery of the ecosystem of the bay.

The Atmospheric Integrated Research Monitoring Network (AIRMoN) was instituted by the National Atmospheric Deposition Program (NADP) in the 1990s to collect precipitation samples each day that precipitation occurs. Daily rainfall samples are analyzed for ionic components, and results are made available to educational, scientific, and commercial communities. The goal of the AIRMoN system is to identify pollutant source-receptor relationships and recognize how emissions changes affect the chemistry of precipitation (Lamb and Bowersox, 2000).

This study will utilize data from rainfall collected at the AIRMoN wet deposition collection site located adjacent to Old Tampa Bay. By coupling the data from the precipitation chemical analyses with information about the trajectory along which the precipitating event traveled, this study will expand the knowledge of the relationship between source locations and types, and nitrogen wet deposition to Tampa Bay.

LITERATURE REVIEW

The Global Nitrogen Cycle

Natural Sources

Nitrogen is an essential element for the growth and normal biological functioning of all organisms. Although nitrogen is an element that is abundant throughout the earth, less than two percent is available for use by the biota. The majority of nitrogen resides in the atmosphere and is tied up in the form of an extremely stable, triple-bonded N₂. Therefore, in order to make enough biologically available nitrogen to complete their life processes, some organisms must expend energy to convert N₂ to a usable form. Enzymes of certain bacteria commonly reduce atmospheric N₂ to NH₃ or NH₄⁺. This process is known as biological nitrogen fixation. The ammonia or ammonium may then be either incorporated into living organisms, or easily oxidized to NO₂⁻ or NO₃⁻ by other bacteria in a process known as nitrification. This process is represented by the following enzymatically-mediated chemical reactions:



Lightning also provides the energy necessary to naturally break the N₂ triple bond, thereby making the nitrogen available for use by organisms (Galloway, 1998 and Vitousek et al., 1997).

Lightning has been estimated to contribute 3-5 Tg N yr⁻¹, while biological nitrogen fixation by terrestrial organisms contributes 90-130 Tg N yr⁻¹ (Galloway et al., 1995). The contribution of marine microorganisms to nitrogen fixation is more difficult to quantify and is said to range from <30 Tg N yr⁻¹ to >300 Tg N yr⁻¹ (Carpenter and Capone, 1983; Carpenter and Romas, 1991).

Many organisms utilize the nitrogen made biologically available by processes in this cycle, but eventually, nearly all of this biologically available nitrogen returns to its inactive state once again by the bacterial process of denitrification. The natural state of this system generally keeps the available nitrogen balanced with the needs of the system.

Anthropogenic Sources

The human impact on the global nitrogen cycle has been profound, particularly in the last century. The need for increased food production has been a major contributor to anthropogenic nitrogen fixation since the 1940's. As the human population experienced a dramatic increase in growth following World War II, and new arable land was limited, the importance of increasing the production of existing land was recognized. The invention of the Haber-Bosch process of fixing N₂ to NH₃ for use in fertilizer greatly increased the productivity of existing arable land while altering the natural state of the global nitrogen cycle. Total industrial N-fixation for use in fertilizer has now reached approximately 80 Tg N yr⁻¹ (Galloway, 1998)

Enhanced nitrogen fixation by legume and rice cultivation also plays a significant role in increasing the global fixed-nitrogen load. Estimates of the average contribution of cultivated crops of legumes and rice to the nitrogen cycle are now 40 Tg N yr⁻¹ (Galloway, 1998 and Vitousek et al., 1997).

Combustion of fossil fuels for energy production introduces available nitrogen in the form of NO_x. NO_x emissions include both NO and NO₂, though the NO species is much more common as a byproduct of fossil fuel combustion. Reactive nitrogen is liberated by two different means during the combustion of fossil fuels. First, NO_x is released directly from long-term geologic stores by the actual physical breakdown of the fuels. Second, the high temperature achieved in the combustion of these fuels breaks the N₂ triple bond of atmospheric nitrogen, thereby oxidizing it to NO_x. According to Vitousek (1997) fossil fuel combustion contributes >20 Tg of biologically available nitrogen per year to the atmosphere, while Galloway (1998) estimates the contribution of this combustion by-product to be on the order of 30 Tg N yr⁻¹.

Figure 2 from Brasseur (1999) depicts schematically, the complete global nitrogen cycle including both natural and anthropogenic sources.

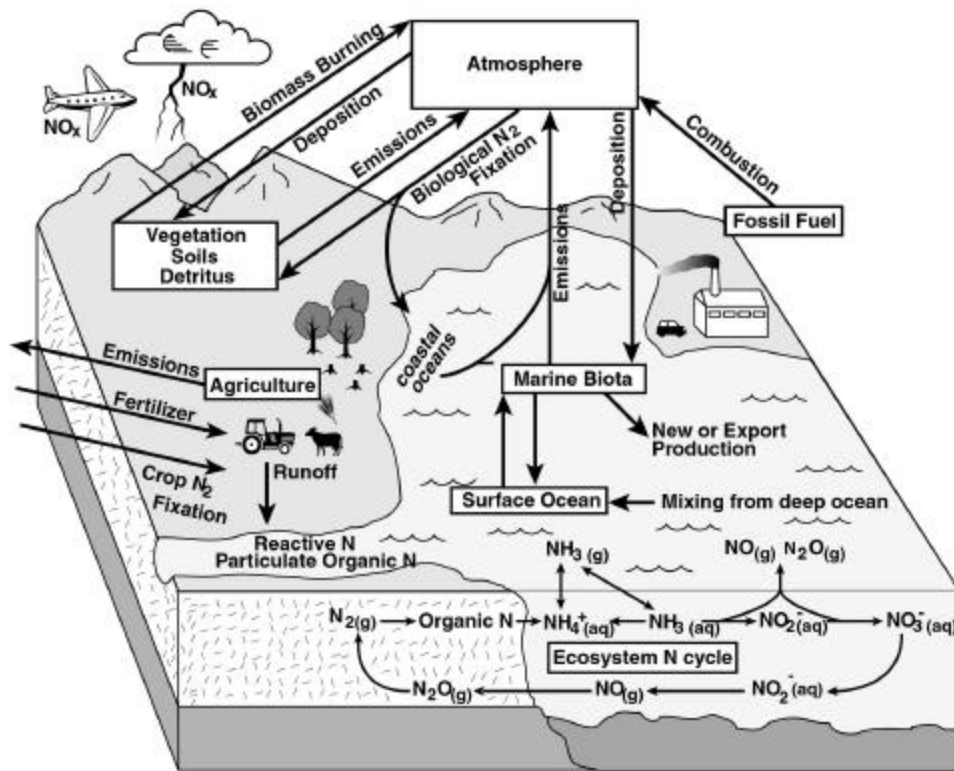


Figure 2. Schematic of global nitrogen cycle from Brasseur (1999).

Estimates of the total rate of anthropogenically-fixed nitrogen now ranges from 140 to 150 Tg N yr⁻¹ based on the estimates presented by both Galloway (1998) and Vitousek (1997).

Global Atmospheric Nitrogen Budgets

Ammonia

Table 1 gives estimates of the atmospheric nitrogen budget for major ammonia production sources including both natural and anthropogenic sources. Domestic animals and fertilizer loss contribute the largest portion of the ammonia

budget at 21.3 and 9 Tg N yr⁻¹, respectively. The largest natural contribution to the ammonia budget originates in the ocean and has a magnitude of 8.2 Tg N yr⁻¹.

Table 1. Major sources of global NH₃ emissions. (Adapted from Brasseur et al., 1998).

Sources	Tg N yr ⁻¹
Domestic animals	21.3
Fertilizer loss	9
Ocean sources	8.2
Soil emissions	6
Biomass burning	5.7

NO_x

Table 2 likewise shows the major emissions sources of NO_x globally. The highest contributions of NO_x are from soil release and fossil fuel combustion, contributing 20.2 and 19.9 Tg N yr⁻¹, respectively.

Table 2. Major sources of global NO_x emissions . (Adapted from Brasseur et al., 1999).

Sources	Tg N yr ⁻¹
Soil release	20.2
Fossil fuel combustion	19.9
Biomass burning	12
Lightning discharge	8
Oxidation of NH ₃	3

Environmental Effects of Nitrogen

Gaseous Nitrogen Compounds

While the production of fixed nitrogen is associated mainly with food production, production of electricity, and transportation, which are all necessary for human life in its present state, the introduction of these excess amounts of nitrogen into the environment can have far-reaching and devastating effects on the ecology of a region. In the atmosphere, nitrous oxide (N₂O) gas, which is produced by industrial activities, can be an important greenhouse gas, while NO_x associated with hydrocarbons or VOCs in the atmosphere, in the presence of sunlight, can undergo reactions to produce ozone or photochemical smog. Tropospheric ozone, or photochemical smog, can have negative health effects on humans such as triggering breathing difficulties in sensitive populations, especially the elderly and those with respiratory ailments. Smog has also been implicated in reducing visibility in scenic areas such as national parks (Heinsohn and Kabel, 1999).

Deposited Nitrogen

Eutrophication

Deposition of reactive forms of nitrogen is of greatest concern in sensitive or impaired ecosystems, like Tampa Bay. Just as the addition of available nitrogen aids farmers in increasing the yield of crops on land, when these compounds reach waterways and wetlands, either by direct deposition or by runoff, the excess nitrogen can greatly increase the primary production of the water body. Studies by the Tampa Bay Estuary Program (TBEP) have indicated that there is approximately a one-to-one ratio of nitrogen loading to chlorophyll-a production in Tampa Bay (Janicki and Wade, 2001).

This increase in nitrogen or other nutrients such as phosphorus, to a water body leads to a condition known as eutrophication. Eutrophication has now become a serious problem in virtually all coastal water bodies around the world (Richardson and Jorgensen, 1996). Through nutrient introduction, the number of phytoplankton or microalgae may increase to levels above those naturally supported by the ecosystem. This algal bloom can have numerous deleterious effects on the balance of the ecosystem and may have serious negative health effects for humans as well. First, the algal bloom may itself be toxic to other species. Some algae produce neurotoxins that may harm or kill other marine life within its immediate vicinity. Toxic algal blooms are often the cause of massive fish kills along shorelines, and have been known to drive fish and mobile crustaceans from the area creating a virtual dead zone. Direct effects on humans can also be observed as algae concentrations become high. Large

areas may be closed to the commercial harvest of seafood resulting in serious economic losses, and certain toxic blooms can cause respiratory distress in humans downwind of the affected water body (Clement et al., 2001).

The presence of excess microalgae in the water column also restricts the availability of sunlight to shallower-than-natural depths. This increase in turbidity and subsequent loss of available sunlight decreases the amount of area suitable for the growth of submerged aquatic vegetation such as seagrasses. With losses of seagrass meadows, more sediment is exposed to tidal action and turbulent waters, and the resuspension of sediments can lead to even greater turbidity (Meyer-Reil and Köster, 2000).

As algae blooms die off at increasing rates because of their unnaturally high populations, dissolved oxygen may be depleted due to their decay. If dissolved oxygen depletion is extensive, hypoxic or anoxic conditions may occur (EPA Office of Water, 2002). Low oxygen conditions often force mobile organisms, such as fish and crustaceans, to avoid the affected area and may destroy benthic invertebrates that are unable to relocate to areas with more acceptable oxygen levels.

Effects of eutrophication are particularly detrimental in areas that are nitrogen-limited, such as wetlands (Morris, 1991) and in estuaries with very low tidal exchange that are unable to clear deposited nutrients from enclosed areas through “flushing” action (Clement et al., 2001).

Acidification

In some waterways and forests, the oxidized nitrogen compounds can be associated with acidification of water bodies due to deposition as nitric acid. When NO_x contacts a strong oxidant in the atmosphere, it may form nitric acid (HNO_3) and deposit to land or water bodies in precipitation. Previous conclusions have placed the blame of water body acidification squarely on the atmospheric deposition of sulfuric acid (H_2SO_4). However, as sulfur emissions have been reduced greatly in the last several decades, acidification has failed to follow as was once hypothesized. This underscores the importance of other forms of acid deposition to water bodies, especially nitric acid. The acidification of freshwater lakes has been shown to be a major problem for some areas in the northeastern United States. In Florida, 23 percent of lakes and 39 percent of streams have been diagnosed as acidic. Acidic lake waters can cause species decline and loss of biodiversity by mobilizing some trace metals such as aluminum, which can be toxic when absorbed by fish and other aquatic organisms (Water Resources Atlas of Florida, 1998).

Atmospheric Transport of Nitrogen Compounds

As stated previously, the major sources of NO_x and ammonia to the atmosphere are fossil fuel combustion and fertilizer production and use, respectively. Approximately 21 Tg N yr^{-1} of nitrogenous compounds are initially emitted directly to the atmosphere as gases (Galloway, 1998). In addition, Smil (1998) approximated that, of the nearly 120 Tg N yr^{-1} applied directly to

agricultural land, an estimated 6% is volatilized as NH_3 and 6% is volatilized as NO_x and N_2O . Distribution and redeposition of these compounds is of important consideration when their deleterious effects on natural ecosystems as described previously are considered.

Generally, these compounds are transported to areas directly downwind of their emissions sources. However, under favorable conditions these emissions may react to form species with residence times on the order of days, so distribution may be on a regional scale (Galloway, 1998).

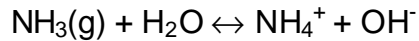
Most NO_x and ammonia is injected into the lower troposphere, or the lowest 1000 to 2000 m of the earth's atmosphere. Once there, they can be transported by wind, mixed by turbulent motion, converted to other compounds through chemical and physical processes, and may ultimately be deposited to the earth's surface or directly to the surface of waterways (Illinois State Water Survey, 2002).

Properties of Atmospheric Nitrogen Compounds

NH_3 and NH_4^+

Whether emitted from agricultural practices or from industrial processes, ammonia is typically released very near the land surface. Once in the atmosphere, NH_3 may deposit in its original chemical form very near its point of origin. Asman and Jaarsveld (1992) estimated that 46% of the NH_3 that was emitted was transported 50 km or less from its source.

Alternately, NH₃ may undergo the following reaction in the presence of water:

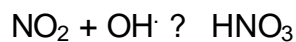


allowing ammonium to form certain aerosol species with completely different transport and deposition properties.

Once in an aerosol form such as NH₄NO₃ or (NH₄)₂SO₄, ammonium may be transported much greater distances than the emitted ammonia because of the longer residence time of these fine particles. This is especially true in urban areas with high emissions of SO₂ and NO_x that may be available to react with NH₄⁺ (Lawrence et al., 2000). Analysis by Warneck, (1988) showed that the atmospheric residence time of NH₃ ranged from 1-5 days, while Aneja et al. (1988) determined the atmospheric residence time of ammonium to be 1-15 days. Fern (1998) estimated that ammonium aerosols may travel thousands of kilometers from their sources crossing not only regional, but national borders before being deposited.

NO_x

Although NO_x is emitted to the atmosphere via the combustion of fossil fuels primarily as NO and NO₂, these species are readily oxidized in the natural atmosphere to NO₃⁻ aerosols, or may react with the hydroxyl radical to form nitric acid as shown in the following reaction from Seinfeld and Pandis (1998):



Once again, the chemical form of these species greatly affects the residence time of nitrogen in the atmosphere. NO_3^- aerosol has a residence time on the order of days, and can be transported across basin boundaries. $\text{HNO}_3(\text{g})$, however, has a relatively short residence time and is transported only short distances downwind of emission source (Lawrence et al., 2000).

Deposition Processes

The processes by which reactive nitrogen species are removed from the atmosphere and transferred to terrestrial or aquatic ecosystems are known as deposition processes. Deposition is the sink by which all of these compounds may enter water systems and thereby contribute to such problems as eutrophication and acidification. The magnitude of this deposition is described in terms of deposition flux, and measurements of deposition flux are important in understanding the linkage between various nutrient pathways and their effects on impaired ecosystems.

There are two main mechanisms for deposition of pollutants from the atmosphere: dry deposition and wet deposition. The dominant removal method for specific pollutants depends on factors such as the chemical solubility of the species and the particle diameters of aerosols.

Dry Deposition

Dry deposition processes of gases and aerosols occur constantly, regardless of the meteorological patterns at a particular time. Dry deposition flux

is relative to the concentration of gas or particles in the air as well as the atmospheric turbulence, chemical properties of the pollutant relative to its deposition surface, and the physical properties of the deposition surface. These variables can be described by a single variable known as the deposition velocity (Seinfeld and Pandis, 1998).

For nitrogen species such as NH_4^+ and NO_3^- particles, as well as HNO_3 and NH_3 gases, dry deposition flux is determined at monitoring stations by measuring ambient concentrations of pollutants and calculating the appropriate deposition velocity. The breadth of variables associated with calculation of these deposition velocities makes estimation of dry deposition less reliable than wet deposition. With respect to reactive nitrogen species, dry deposition is most important in deposition of NH_3 close to emission sources. However, as chemical reactions transform NH_3 to NH_4^+ species, wet deposition processes become more important (Lawrence et al., 2000).

Wet Deposition

Unlike dry deposition, wet deposition only occurs when precipitation reaches the earth's surface. Therefore, wet deposition flux is the accumulation of only a few hours of precipitation per year, whereas dry deposition occurs constantly. Precipitation acts to naturally scrub the atmosphere of pollutants and deposit them on either the terrestrial or aquatic ecosystem or to the surface of vegetation. This scrubbing action is essential to the chemical balance of the

atmosphere. As discussed previously, however, if excess nitrogen enters sensitive ecosystems through this pathway, harmful effects may result.

Wet deposition of nitrogen may be associated with condensed water in the form of rain, snow, sleet, hail, or even fog, and can be incorporated into these micrometeors through three main mechanisms. First, particles may act as cloud condensation nuclei when the atmosphere is supersaturated with water vapor. This process is known as nucleation scavenging, or washout, and is most efficient with aerosols having particle diameters from 0.1-10 μm . Less efficient is the process of diffusion. In this process, atmospheric gases, such as HNO_3 , are absorbed into water droplets via Brownian diffusion processes. Incorporation of pollutants by diffusion can either occur as an in-cloud process or occur below-cloud when precipitation is actively falling. Lastly, impaction, or scavenging, is a below-cloud process that occurs when precipitation is actively falling. In this process water droplets contact either aerosols or gases on their descent and the particles or gases are impacted into the body of the precipitation droplet (Brasseur et al., 1999). This process is also known as rainout and its rate has been shown to be dependent upon the intensity of the rainfall. The highest scavenging rates are associated with the most intense rainfall, and as a precipitation event progresses, there are fewer water-soluble pollutants in the atmosphere to be removed by the precipitation (Inglis and Choularton, 2000).

In addition to these three main mechanisms of wet deposition which are dependent upon precipitation actively falling from clouds and reaching the earth's surface, wet deposition may also occur when fog or cloud droplets containing

pollutants come into direct contact with the ground or water surfaces, or with vegetation that may extend into the atmosphere (Illinois State Water Survey, 2002). These processes may dominate for the deposition of pollutants to remote regions at high altitudes such as mountains. This acid fog has been implicated in the degradation of forests in many areas of the Appalachian Mountains.

Vegetation is compromised by the weakening of leaves, loss of soil nutrients, and release of toxic metals in the soil for uptake by the flora (EPA, 2003b).

Wet deposition is commonly measured using a collection bucket equipped with a moisture sensor that is only uncovered during a precipitation event.

Rainwater concentrations of chemical compounds or ions are measured directly and results are reported as units of concentration. Nitrate and ammonium are the nitrogen-containing species that are typically analyzed when wet deposition fluxes are measured.

Patterns of dry and wet deposition vary greatly geographically and meteorologically, but in general, dry deposition of NO_x is positively correlated with wet deposition of NO_3^- , and dry deposition of NH_3 is positively correlated with wet deposition of NH_4^+ (Lawrence, 2000).

Deposition to Tampa Bay

Tampa Bay is an important resource for the more than two million people in the area it borders. Though the seagrasses of the bay have begun an upturn in their numbers, in order to account for the expected increase in the human population in the next 20 or 30 years, the Tampa Bay Estuary Program has identified that it must find new means of reducing nitrogen introduction into the

bay. By identifying and reducing alternative pathways by which nitrogen is entering Tampa Bay, TBEP hopes to “hold the line” at the current level of nitrogen input in order to meet the seagrass repopulation goals that they have set.

During the 1970s the major culprit of seagrass decline was demonstrated to be nutrient-laden discharge from wastewater treatment plants. These plants have since been modified to include advanced wastewater treatment which greatly reduces the concentration of nutrients entering the bay from this source.

More recent studies have demonstrated the increased importance of atmospheric deposition as a source of nitrogen to Tampa Bay waters. Zarbock et al. (1996) has estimated that nearly one-third, or 1,100 metric tons of nitrogen per year, of all nitrogen that enters Tampa Bay is a result of direct atmospheric deposition to the surface of the bay. Poor et al. (2001) calculated the annual average nitrogen deposition rates to Tampa Bay during the period of 1996-1999. The average total nitrogen flux to the Bay waters was estimated to be 7.3 kg/ha/yr.

Tampa Bay Nitrogen Emission Sources

EPA emissions inventory indicates several large sources of NO_x and NH₃ in the areas adjacent to Tampa Bay. The second and third largest sources of NO_x in Florida identified by the EPA AirData website (<http://www.epa.gov/air/data/index.html>) are coal-fired electric utility plants located in Hillsborough County. The TECO Big Bend and TECO Gannon power plants were the largest local emission sources of NO_x, releasing 31,000 and

29,000 metric tons per year, respectively, according to EPA's 1999 data (EPA, 2003a).

Six of the top ten emissions sources of ammonia emissions in Florida in 1999 were fertilizer manufacturing facilities to the south and east of Tampa in Polk and eastern Hillsborough counties. The two largest sources are located in Polk County, and are both owned by IMC. The Nichols Plant emitted approximately 1300 metric tons of ammonia, while the New Wales Plant recorded 450 metric tons of ammonia emissions (EPA, 2003c). The top local emissions sources of NO_x and ammonia are displayed graphically in Appendix A.

Previous Nitrogen Wet Deposition Studies

Galloway et al. (1983) determined, by using ship-board precipitation collectors, the background concentrations of nitrogen compounds in rainfall over the Atlantic Ocean. Galloway's results showed background average concentrations in the Northern Hemisphere to be 5.5 ueq/L for nitrate, and 3.2 ueq/L for ammonium. Another study found that the volume-weighted average concentrations of nitrate and ammonium in marine air arriving at a coastal Portugal study site were 7.9 ueq/L and 19.2 ueq/L, respectively (Casimiro, 1991). Additionally, Avila et al. (1999) found that marine-originating precipitation at their collection site in NE Spain received volume-weighted average concentrations of 16.8 ueq/L for nitrate and 20.1 ueq/L for ammonium, These concentrations were less than the average concentrations for all of the rainfall events that they

sampled which had concentrations of 20.7 ueq/L and 22.9 ueq/L for nitrate and ammonium, respectively.

Concentrations of nitrogen-containing ions analyzed in precipitation from a mixture of urban and rural collection sites around the Korean Peninsula averaged 19.3 ueq/L for nitrate and 32.6 ueq/L for ammonium (Lee, 2000), and Cerón et al. (2002) found that the concentrations of nitrate and ammonium were 53.4 ueq/L and 5.54 ueq/L, respectively, for a coastal area of the Yucatan peninsula.

Studies have also been conducted immediately downwind of known nitrogen emissions sources to quantify the source's effect on rainfall chemistry in close proximity. Alastuey et al. (1999) found that nitrate and ammonium concentrations in rainfall collected beside a coal-fired electric utility plant in NE Spain were 27.2 ueq/L and 80 ueq/L, respectively.

These studies suggest that, in general, concentrations of ammonium and nitrate in precipitation collected from air masses which have recently traversed expanses of water are lower than those from land-originating trajectories.

Previous nitrogen flux studies by Poor et al. (2001) for the Tampa area revealed that the average annual nitrogen flux rates for nitrate and ammonium were 2.40 kg/ha/yr and 1.74 kg/ha/yr, respectively, with an average 5-7 mm rainfall event contributing 0.026 kg/ha to Tampa Bay. Additionally, dry deposition was reported for this study and found to have a magnitude similar to that found for the wet deposition. Poor et al. also reported that ammonium nitrogen fluxes from other NADP sites in Florida showed lower deposition rates than the results from Tampa. Average yearly ammonium nitrogen deposition was 0.8 kg/ha/yr

from Chassahowitzka, 1.3 kg/ha/yr from Sarasota, and 1.2 kg/ha/yr from the Kennedy Space Center. Nitrate nitrogen fluxes were determined to be similar between Tampa and the other three collection sites. The nitrate nitrogen flux was 2.1 kg/ha/yr at Chassahowitzka, 2.3 kg/ha/yr at Sarasota, and 2.8 kg/ha/yr at the Kennedy Space Center.

METHODS

Site Description

The precipitation data evaluated in this study was obtained from the Tampa Bay Atmospheric Integrated Research Monitoring Network (AIRMoN) station (FL18). This station is located on the eastern end of the Gandy Bridge, which connects Hillsborough to Pinellas County across Old Tampa Bay. The site is situated immediately adjacent to the bay at approximately 27.85 degrees north latitude and 82.55 degrees west longitude (Fig. 3).

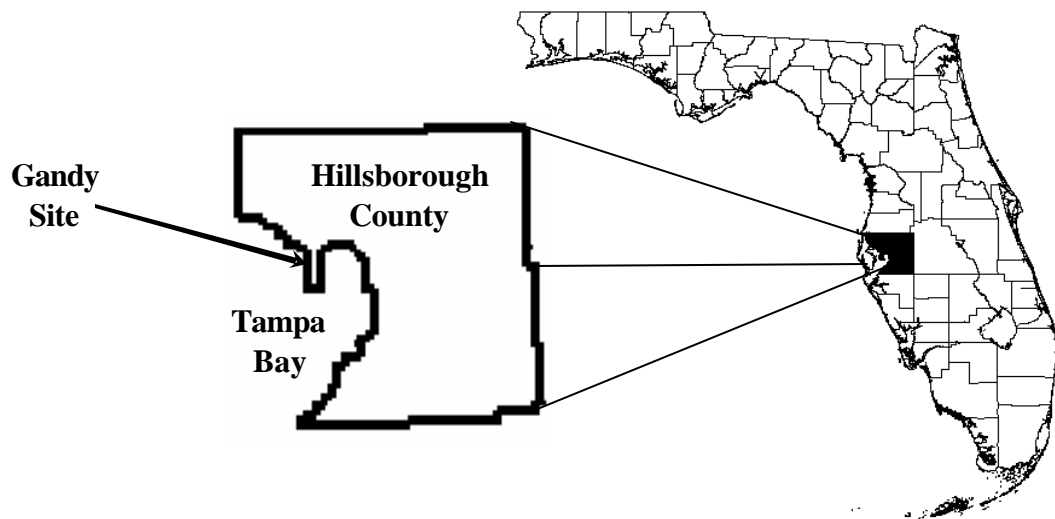


Figure 3. Location of AIRMoN data collection site in Hillsborough County, FL.

AIRMoN Network Description

Information for this study was obtained from the chemical evaluation of rainfall collected via the National Atmospheric Deposition Program (NADP) Atmospheric Integrated Research Monitoring Network (AIRMoN) wet deposition collection system. The AIRMoN network was instituted in 1996 to address several shortcomings of the previously established NADP network. Among them, AIRMoN collects and packages samples on a daily basis, instead of a weekly basis as the traditional NADP network does. This serves multiple purposes. First, by collecting samples daily and chilling them until chemical evaluation, nutrient-containing species such as ammonium and nitrate can be more accurately evaluated. Also, single-day sampling allows the coupling of precipitation data with specific meteorological events in order to more accurately determine source origins and trends.

The AIRMoN precipitation collection system utilizes an Aerochem Metrics Model 301 wet/dry precipitation collector in order to obtain samples only when precipitation is falling. A photograph of the Aerochem Metrics Model 301 wet/dry precipitation collector is shown in Figure 4. A mobile lid covers a clean sample bucket until precipitation is detected by a wetness sensor. When precipitation is detected, the mobile cover is automatically removed exposing the bucket until precipitation is no longer detected. The bucket is then recovered until the next precipitation event.

During a rainfall event, while the Aerochem Metrics sampler bucket is uncovered, it collects both wet and dry deposition. However,

Beverland et al. (1997), demonstrated that the proportion of dry deposition compared to wet deposition during this time period was insignificant provided the sample collection bucket was well covered prior to and after the rainfall event.



Figure 4. Photograph of Aerochem Metrics Model 301 wet/dry precipitation collector.

Precipitation depth at the Gandy Bridge AIRMoN site is measured by a collocated National Weather Service “stick” gauge and a secondary Belfort ® 5-780 recording rain gauge.

Each morning, between 8:00 am and 10:00 am, any precipitation that fell in the previous twenty-four hours was retrieved from the Aerochem Metrics sampler and refrigerated by technicians of the Environmental Protection Commission of Hillsborough County (EPCHC). Samples were sent, on a weekly basis, to the NADP Central Analytical Laboratory at the Illinois State Water Survey in Champaign, Illinois for analysis.

Precipitation samples were analyzed for pH and conductivity in the both the field and in the lab, and major ion concentrations including chloride, nitrate, sulfate, sodium, ammonium, potassium, calcium, and magnesium were analyzed in the lab. Samples were initially evaluated on-site and given a quality rating of either “A”, “B”, or “C”. “A” quality samples were of the highest quality and contained nothing but water, all protocols were followed, and there was no indication that there were issues affecting the quality of the sample. Samples with a quality rating of “B” were of unknown quality and may have contained a contaminant such as insect or plant matter. “B” quality samples may have also been contaminated due to sampling errors. Samples with a quality rating of “C” were of the lowest quality. They were either collected over an undefined time period, found to contain bird droppings, or had some other indication that the quality was compromised.

Analytical and quality control methods for data collection and laboratory analyses can be accessed through the NADP web site at <http://nadp.sws.uiuc.edu/> (NADP, 2002).

Data Acquisition

The chemical analysis data for the Tampa AIRMoN site was accessed via the NADP/AIRMoN website for the period of 1 August 1996 through 30 December 2000. Ion data obtained for this paper was accessed in units of microequivalents per liter (ueq/L). Laboratory-determined pH was used as the representative pH of the rainfall samples, and pH results were converted and

utilized as the concentration of hydrogen ions reported in ueq/L for the remainder of this paper. Daily precipitation data with missing chemical analysis data or with a quality rating of “C” were excluded from the analysis prior to further evaluation. Samples with incomplete chemical evaluations were generally found to be representative of very low rainfall events, thereby representing only the smallest contribution of nitrogen to Tampa Bay.

Air Mass Trajectories

Air mass back trajectories for specific daily rainfall events were derived using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by the National Oceanographic and Atmospheric Administration’s Air Resource Laboratory (NOAA/ARL). The HYSPLIT model utilizes two-hour archived meteorological data to plot the trajectory of an air mass along a three-dimensional grid. The model uses both Lagrangian and Eulerian calculations in order to plot trajectory results in a graphical output that reports both space and time results for the calculation of air mass trajectory (Draxler, 1997). The HYSPLIT model was accessed via the World Wide Web at <http://www.arl.noaa.gov/ready/hysplit4.html> (NOAA-ARL, 2002).

Twenty-four hour back trajectories were computed from the time of each precipitation event. Precipitation times were determined by reviewing NEXRAD National Mosaic Reflectivity Images and surface observations from the Tampa International Airport, which is located approximately five miles north of the Gandy Bridge Site (NCDC, 2001). For precipitation events that spanned several hours,

HYSPLIT back trajectories were computed for approximately every two hours over the course of the event. Among these trajectories, the dominant direction was chosen as representative of the entire event. Back trajectories were calculated using altitudes between 250 and 1000 meters above mean sea level in order to illustrate a representative cross-section of the mixing layer.

Trajectory Classification

Back trajectories were classified based on the path of the precipitating air mass during the 24-hours before reaching the Gandy site. The events were categorized into six different trajectory types based on the direction in which the air mass spent the majority of the previous day. The trajectory classifications were defined as Cape, Bahamas, Cuba, Gulf, Panhandle, and Tampa (Figure 5).

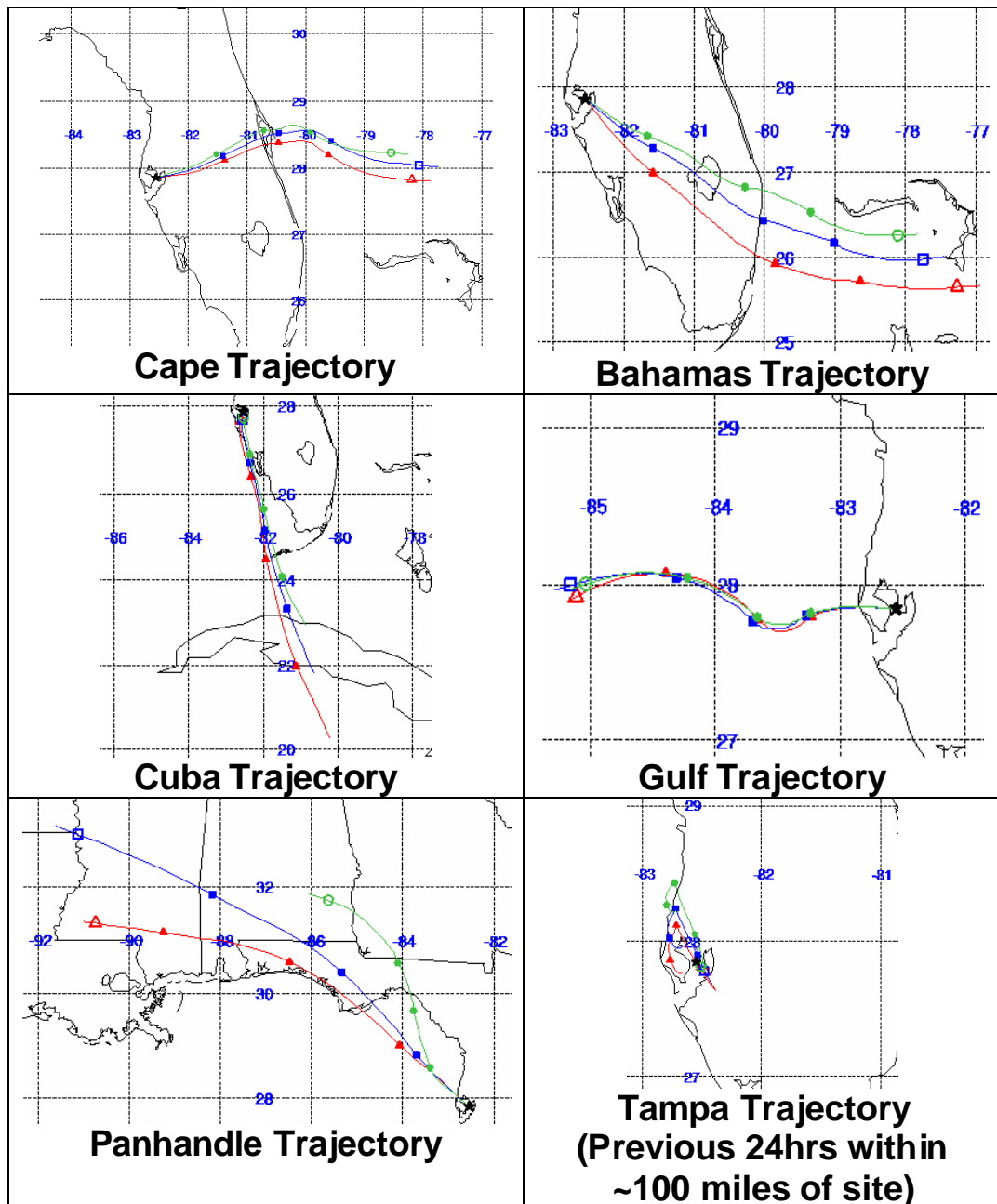


Figure 5. Examples of HYSPLIT air mass trajectory categories used in classifying rainfall events. Derived from Earls (2001).

Daily AIRMoN rainfall data for which no HYSPLIT trajectory information were available was removed from the set resulting in a total of 292 complete

records for the time period of August 1996 through December 2000.

Furthermore, data collected on 30 December 1997 and 20 December 2000 were excluded from further evaluation because sodium, chloride, and magnesium concentrations were found to be an order of magnitude or greater more than the average concentrations for those ions, thereby indicating likely contamination by sea spray. The AIRMoN dataset used for the remainder of the analyses is located in Appendix B.

Data Analysis

All general statistical analysis calculations were performed using the Microsoft Excel 2000® data analysis add-in and calculation functions of Microsoft Access 2000®.

Ion Balance Calculation

Before ion results were analyzed individually, the quality and completeness of the chemical analysis was confirmed by performing an ion balance calculation for all precipitation chemistry samples as described in Lee et al. (2000). Cation concentrations were summed for each individual rainfall sample and reported in units of microequivalents per liter (ueq/L). Next, the same summation was performed with the anions from each sample. Cation total concentrations were then compared with anion totals by performing regression analysis of the total microequivalents per liter of all anions as a function of the total microequivalents per liter of all cations. The R^2 value and the slope of the

regression line for this regression were evaluated to determine the fitness of the ion chemistry data.

Non-Sea Salt Sulfate Calculation

Since sulfate ion is contributed to atmospheric deposition by both anthropogenic emission sources and natural sea salt sources, and this study is interested primarily with the deposition of anthropogenically derived pollutants, the total concentration of sulfate in the precipitation samples was thus converted to a non-sea salt (nss) fraction. This conversion was accomplished by comparing the standard sea salt ratio of sulfate to sodium (0.121) to the ratio of sulfate to sodium present in the precipitation samples. The excess sulfate concentration contributing to sulfate to sodium ratios greater than 0.121 was determined to be anthropogenically derived and was thus classified as non-sea salt sulfate (nssSO_4^{2-}) (Alastuey et al., 1999). The non-sea salt fraction of the sulfate concentration will therefore be included in further calculations associated with this study. The formula for this conversion is provided in Equation 1 .

$$[\text{nss SO}_4^{2-}] = [\text{total SO}_4^{2-}] - [\text{Na}^+] * (0.121)$$

Equation 1

The average sea salt fraction accounted for 3.2% of the total sulfate concentration for the data set.

Histograms

Distribution of ion concentrations and precipitation totals were determined by creating histograms for each ion as well as precipitation depth data. Histograms were used in determining whether the data were normally distributed or if data distributions were skewed.

General Statistics

General statistical results were reported for each ion species as well as precipitation data. Precipitation data was analyzed for mean, standard deviation, and median depths and reported in units of millimeters (mm). Means, standard deviations, medians, and volume-weighted averages (VWA) were calculated for ion concentration data and reported in units of ueq/L. Volume-weighted averages for all ions were calculated using the following equation:

$$VWA = \frac{\sum_{i=1}^n C_i R_i}{\sum_{i=1}^n R_i}$$

Equation 2

where C is the concentration of each ion collected during rainfall events in ueq/L, and R is the rainfall depth of each event in millimeters (mm).

Correlations

Correlation calculations were next run among ion concentration data for all analyzed ion species. Pearson's correlation coefficients (r) were determined and

used to determine the relative tendency among ions to be deposited in concert with one another. The Pearson's correlation results were used to evaluate the potential for ions to be deposited either as chemical compounds or as individual species that may be derived from a similar emission source.

Multiple Regression Analysis

Multiple regression analysis was next used to determine the possible forms in which nitrate and ammonium were deposited by comparing all other analyzed anion concentrations with ammonium and all cation concentrations with nitrate concentrations. In addition, a regression analysis was also calculated for non-sea salt sulfate because of its tendency to deposit with nitrate and ammonium ions.

Principle Component Analysis

A principle component analysis (PCA) was conducted for the entire ion data set to determine which ions varied in conjunction with one another. These principle components were also used to seek ions either emitted from the same source or transported together. The SAS® statistical analysis program was used to perform the principle component analysis calculations.

Nitrogen Flux Calculations

In order to calculate the nitrogen flux deposited by wet deposition to the collection site, the ion concentration data first had to be converted into the form

of milligrams per liter (mg/L). This was accomplished by the use of the following equation:

$$\frac{mg}{L} = \left(\frac{ueq}{L} \right) \left(\frac{eq}{10^6 ueq} \right) \left(\frac{mol}{eq} \right) \left(\frac{g}{mol} \right) \left(\frac{10^3 mg}{1g} \right)$$

Equation 3

The nitrogen flux contributions of both ammonium and nitrate were then calculated for each event using the equation:

$$F_{wet} = C_{rain} D \cdot 10^{-2}$$

Equation 4

where C_{rain} is the concentration of nitrogen in rainfall in mg/L, D is the depth of the rainfall in mm, and F_{wet} is the flux of nitrogen in kg-N/ha/d.

The average yearly nitrogen flux rate for the entire data set was calculated and compared with the fluxes from previous studies conducted in other regions. Standard measurement errors were then calculated using the percent relative standard deviations (%RSD) determined by Poor et al. (2001). According to this study, the RSD for nitrate wet deposition was 40%, and the RSD for ammonium wet deposition was 41%. Therefore, by using the equation:

$$S_{i,wet} = F_{i,wet} \cdot RSD$$

Equation 5

where i is defined as either ammonium or nitrate, the average annual flux error for the total data set was determined.

Statistical Analysis Sorted by Trajectory

The precipitation event data was segregated according to the HYSPLIT trajectory classifications. Volume-weighted average concentrations were calculated for all ions according to trajectory classification. Similar studies have been conducted by Harrison et al. (2000) and Lucey et al. (2001) to analyze the relationship between precipitation chemistry and air mass trajectory. Ammonium and nitrate nitrogen fluxes were averaged for the entire data set and also for each individual trajectory classification for comparison.

Trajectory Chemical Classification

The type of air mass influencing the ion concentrations at the monitoring site during a precipitation event may have terrestrial, as well as marine, characteristics. Because of the potential for mixed-influence air masses from any trajectory due to the large water bodies surrounding the state, specific chemical signatures were chosen for marine, terrestrial local combustion, terrestrial aged combustion, and mixed terrestrial air masses in order to give a better idea of their specific contributions to nitrogen deposition beyond simply the physical trajectory that the air mass has traveled. Nitrate and ammonium nitrogen fluxes were therefore calculated for each classification. The parameters used in defining these air masses are presented below.

Marine Air Masses

Marine-classified air masses were defined as having a $[\text{Ca}^{2+}]:[\text{Na}^+]$ ratio less than 0.2. This ratio was chosen based on the high contribution of sodium from marine water bodies and the major contribution of calcium from crustal sources in this geographic area. Because the Gandy Bridge collection site is not located immediately adjacent to the Gulf of Mexico, as air masses passed over some land area to arrive at the site, additional calcium was collected by the air mass from the terrestrial environment. Therefore, the ratio of 0.2 was chosen for marine air masses based on the distribution of the data at hand.

Terrestrial Air Masses

Terrestrial air masses were classified as having a $[\text{Ca}^{2+}]:[\text{Na}^+]$ ratio greater than 0.5 indicating only events with the strongest source of crustal calcium and the least influence of sea salt. These air masses were subdivided based upon combustion influences. Local combustion events were defined as high in acid ($\text{H}^+ > 80$ ueq/L) while aged combustion sources were defined based upon a H^+ concentration < 80 ueq/L and a $[\text{NH}_4^+]:[\text{SO}_4^{2+}]$ ratio greater than 0.25 indicating air masses that have had sufficient time to chemically mature. Terrestrial source air masses that could not be classified into either local or aged combustion were designated as mixed terrestrial.

Mixed Marine/Terrestrial

Air masses without distinctly marine or terrestrial ion chemistry were classified as mixed marine/terrestrial and analyzed as such.

Tests for Statistical Significance

Differences between average nitrogen fluxes for physically-classified trajectories as well as between chemically-classified air masses were examined for statistical significance by utilizing the Kruskal-Wallis ANOVA test. The Kruskal-Wallis ANOVA test is a rank-order test and was chosen to test for significance of differences between classified data sets due to its usefulness in analyzing more than two data sets non-normal distributions. For significance, a confidence level of 95% was chosen. Similar tests for significant differences in ion species means between air mass categories using the Kruskal-Wallis ANOVA test were documented by Russell et al. (1998).

Tropical System Rainfall Deposition

Archived tropical system data obtained via the Terrapin Associates Hurricane and Tropical Storm Tracking website (<http://hurricane.terrapin.com>) was utilized to determine which precipitation events were likely associated with tropical storms and hurricanes. Tropical events that were determined to have potentially influenced the Tampa Bay area, due to the proximity of the storm path, were investigated further via the NEXRAD National Reflectivity Images to determine events that actually contributed precipitation to the area and on which

dates those precipitation events occurred. The tropical precipitation dates were cross-referenced with the sample data from the Gandy Bridge Site and common events were evaluated. These precipitation events were chosen because they represented mainly marine air that had collected primarily over open water, and were essentially “pristine” with respect to anthropogenic pollutants. Ion chemistry data from these events was compiled and volume-weighted average concentrations were calculated. This data was compared with volume-weighted average data from all non-tropical events to approximate a background concentration of ammonium and nitrate in precipitation for this region.

Nitrate and ammonium fluxes were then calculated for tropical and non-tropical data. Results for this calculation were also tested for significance by using the Kruskal-Wallis test with a confidence level of 95%.

Comparison of Pollutant Levels

Galloway et al. (1983) determined that nitrate and ammonium background concentrations from rainfall collected over the Atlantic Ocean were 5.5 ueq/L and 3.2 ueq/L, respectively. These results were compared with the average concentrations of nitrate and ammonium from the entire data set, the concentrations from the Gulf HYSPLIT trajectories, the concentrations from events chemically-classified as marine, and concentrations from events associated with tropical systems. The Gulf, marine, and tropical event classifications were expected to represent the cleanest air masses determined by each of the classification methods used in this study.

RESULTS

Ion Balance Calculation

An ion balance test was performed to test the quality and completeness of the data, and is shown in Figure 6. The graph of the total anion microequivalents per liter versus the total cation microequivalents per liter for each rainfall event had a linear relationship with a slope of 1.08, and correlation (r) of 0.98 indicating a tight regression. However, as the ion totals increase, the residual values between the regression line and the actual data points increase slightly, and culminate with the point expressing the highest ion loading showing a greater contribution of anions than cations. By looking at this specific event, it was determined that an excess of chloride ion was the likely cause of the variability. This may have been a result of sample variability, inaccuracies in the testing, or an unmeasured ion contributing significantly to the event on the date that this sample was taken. It was decided that this event should remain in the data set for further testing since its influence on such a large data set was determined to be minimal. Overall, the ion balance graph demonstrates that the major contributing ions in the wet deposition have been accounted for, and that the results of the testing were unbiased.

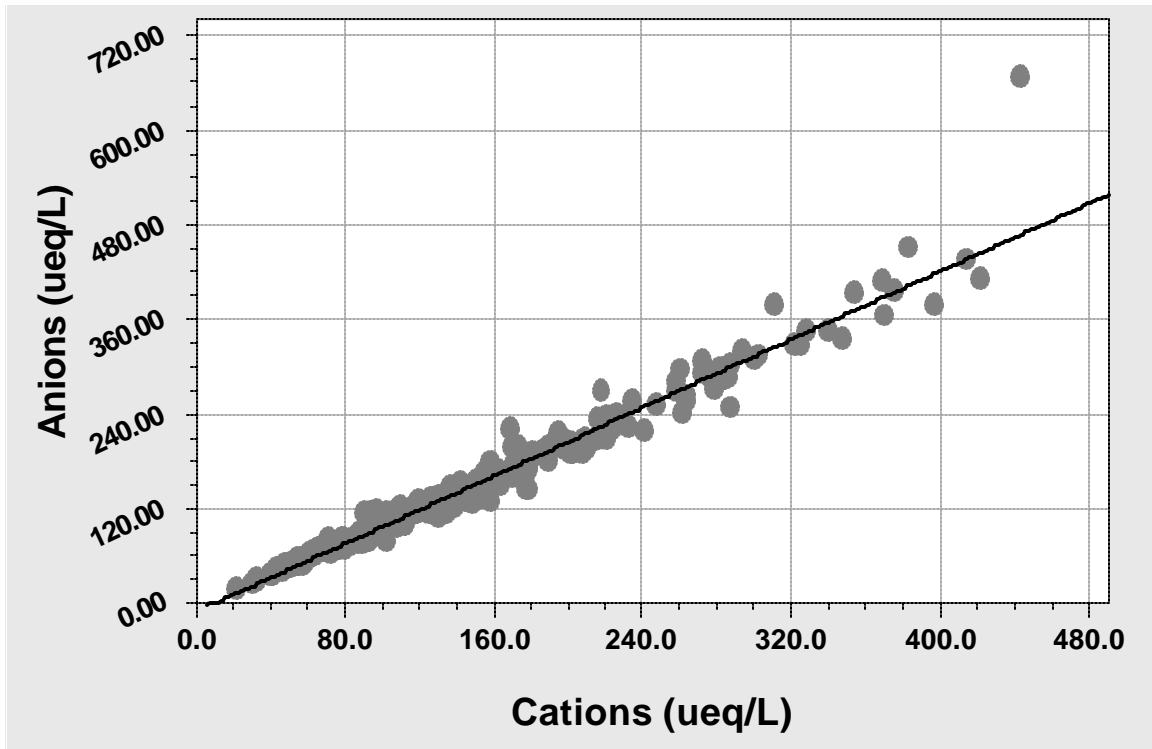


Figure 6. Total concentration of anions plotted against the total concentration of cations for each event.

Histograms

Histograms for all precipitation depth and ion concentration data were calculated and are presented in Figures 7 through 16.

Histograms of precipitation depth demonstrated that wet deposition events during the selected time period consisted primarily of small amounts of rainfall. The highest frequency of events consisted of the lowest precipitation depths and the general trend was toward decreasing frequency as rainfall depth increased. This was likely due to the sporadic and isolated rainfall that is common to the Tampa Bay area during the wet season.

The examination of histograms for ion concentrations showed the data distribution to be positively skewed in all cases. There was a large variability in the data set in the upper concentrations making the data non-normal and therefore subject only to statistical analysis appropriate to data having non-normal distributions. These results were consistent with results obtained by Inglis and Choularton (2000) which demonstrated that as the duration of a rainfall event becomes longer, the pollutants available for scavenging become scarcer. This leads to decreases in pollutant concentrations as precipitation depth increases.

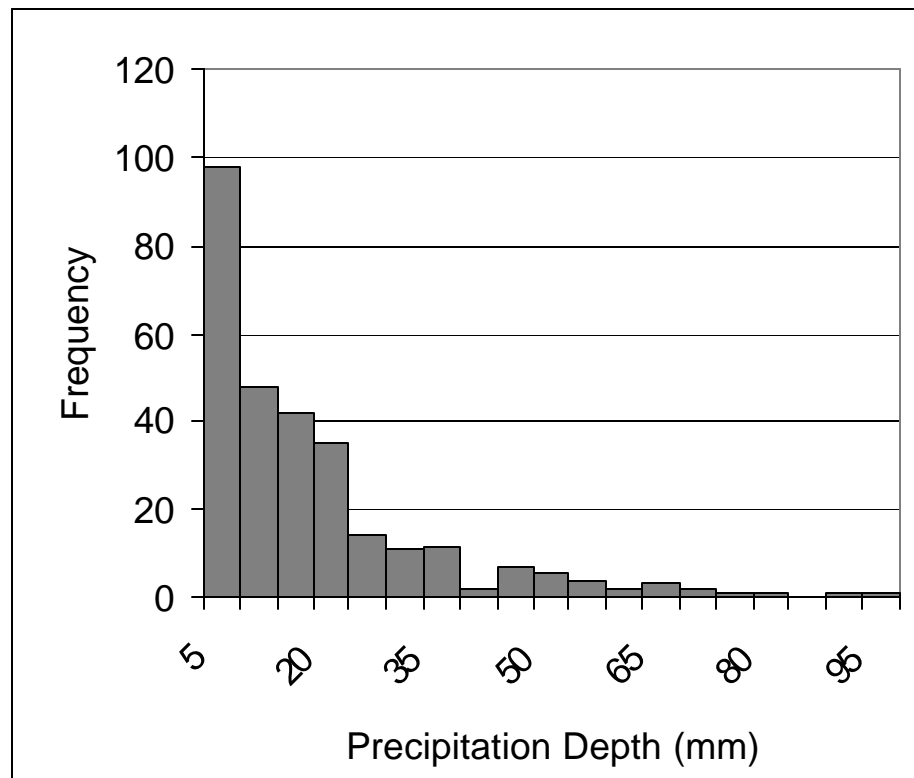


Figure 7. Histogram of distribution of precipitation depth among all events.

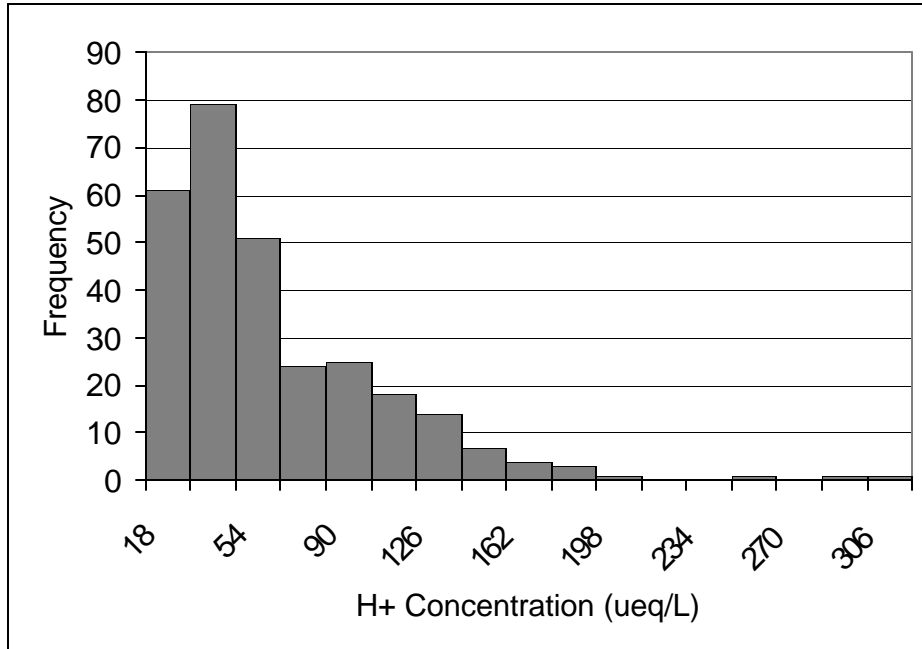


Figure 8. Histogram of distribution of hydrogen ion concentration among all events.

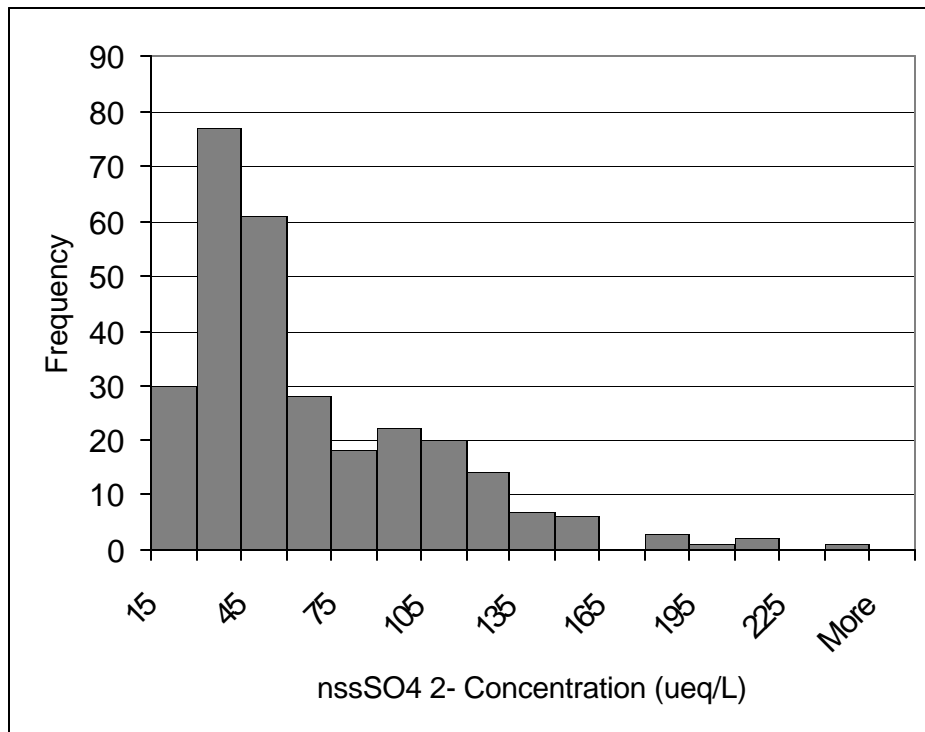


Figure 9. Histogram of distribution of non-sea salt sulfate concentration among all events.

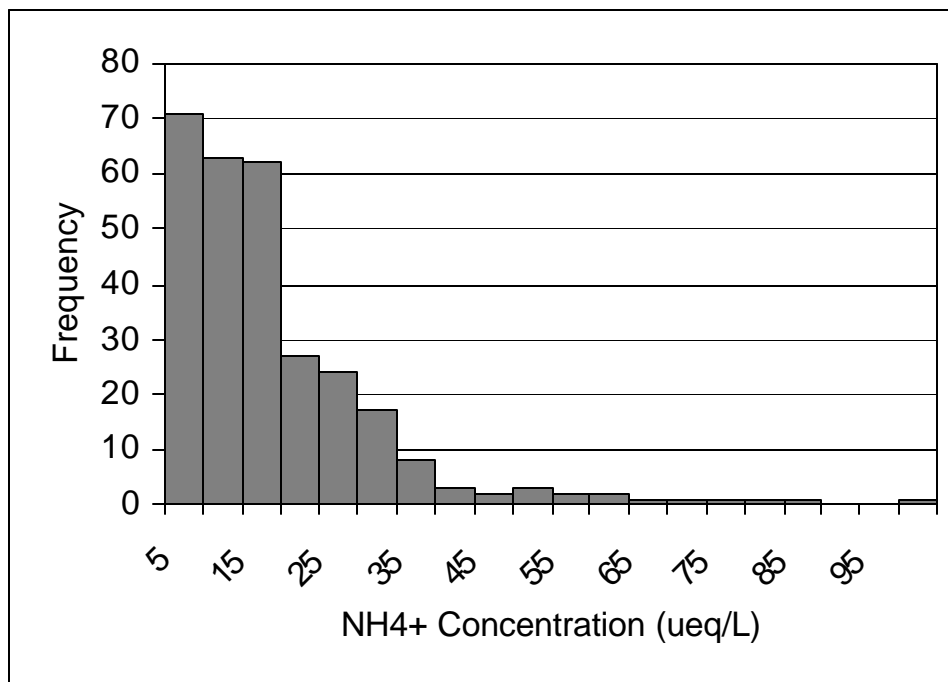


Figure 10. Histogram of distribution of ammonium concentration among all events.

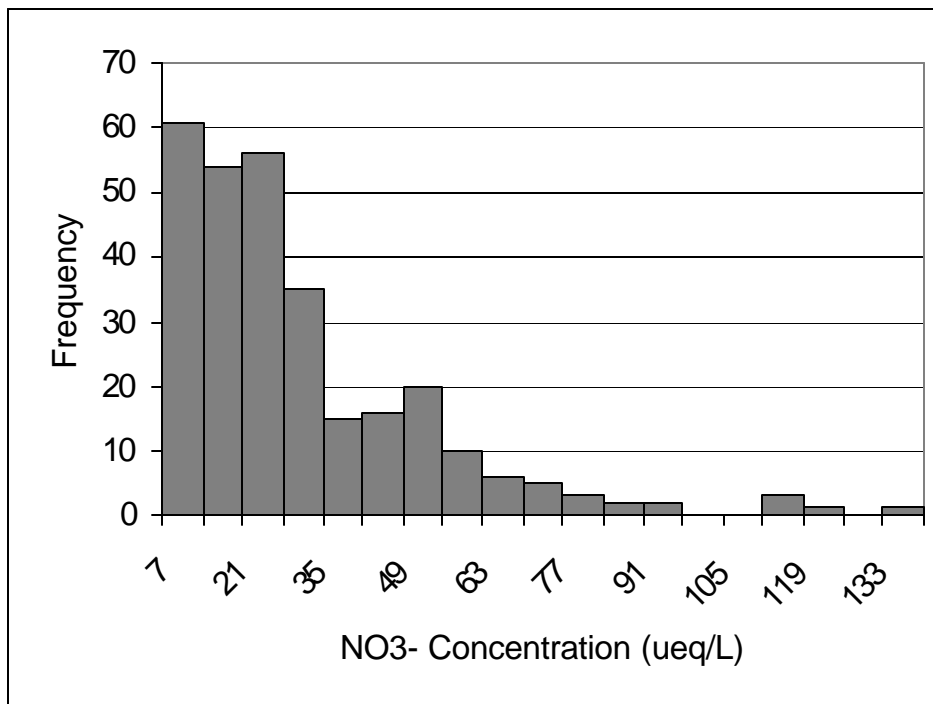


Figure 11. Histogram of distribution of nitrate concentration among all events.

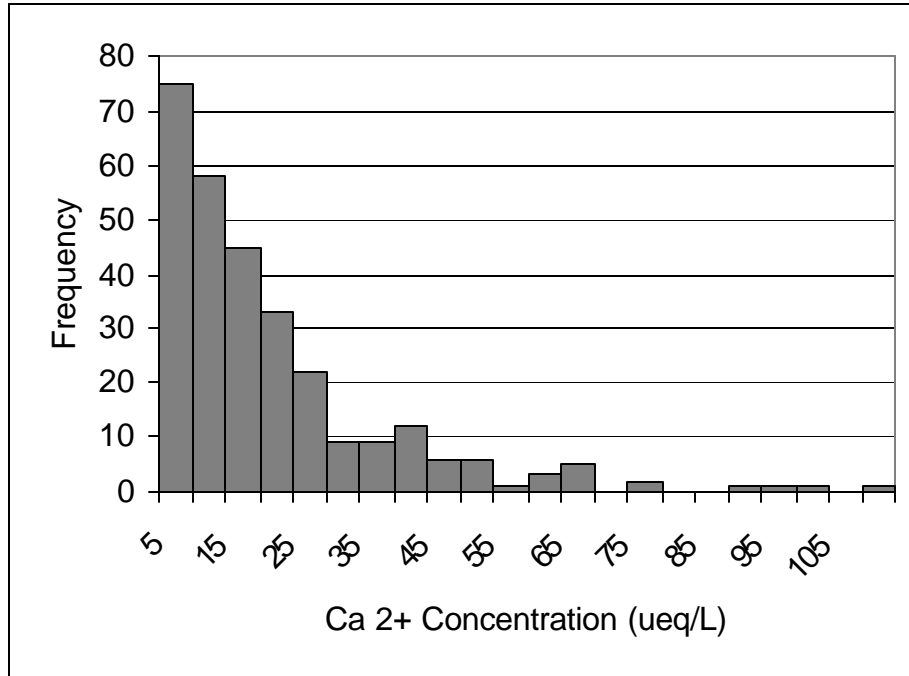


Figure 12. Histogram of distribution of calcium concentration among all events.

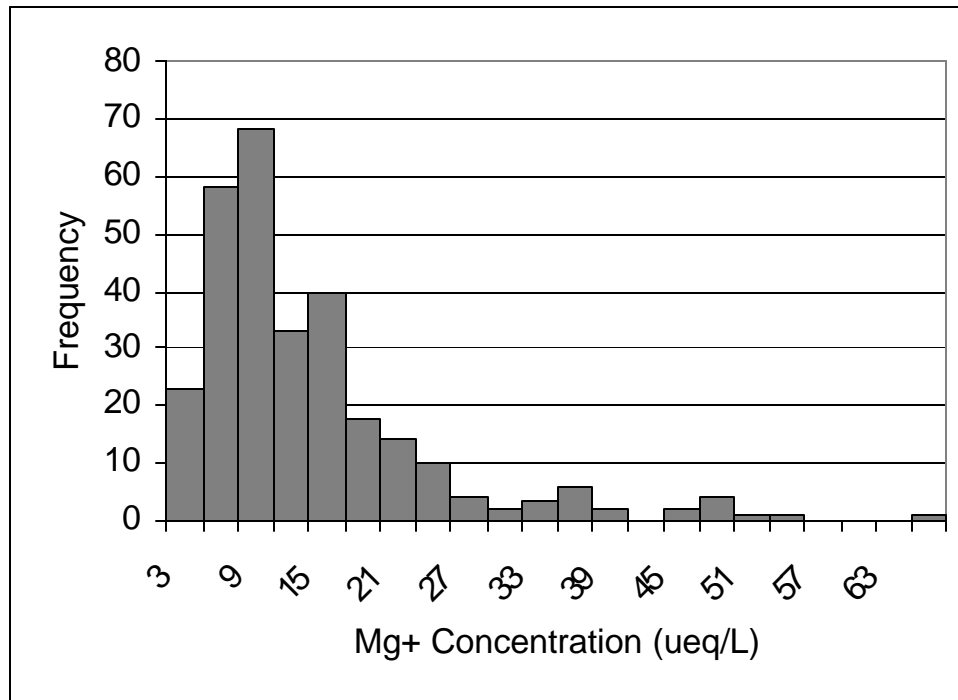


Figure 13. Histogram of distribution of magnesium concentration among all events.

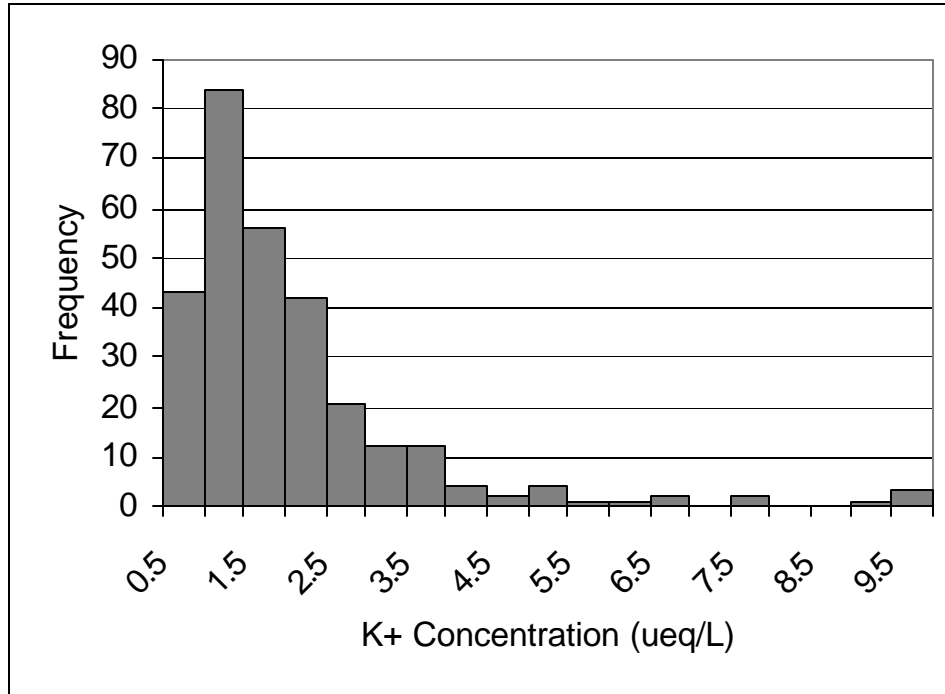


Figure 14. Histogram of distribution of potassium concentration among all events.

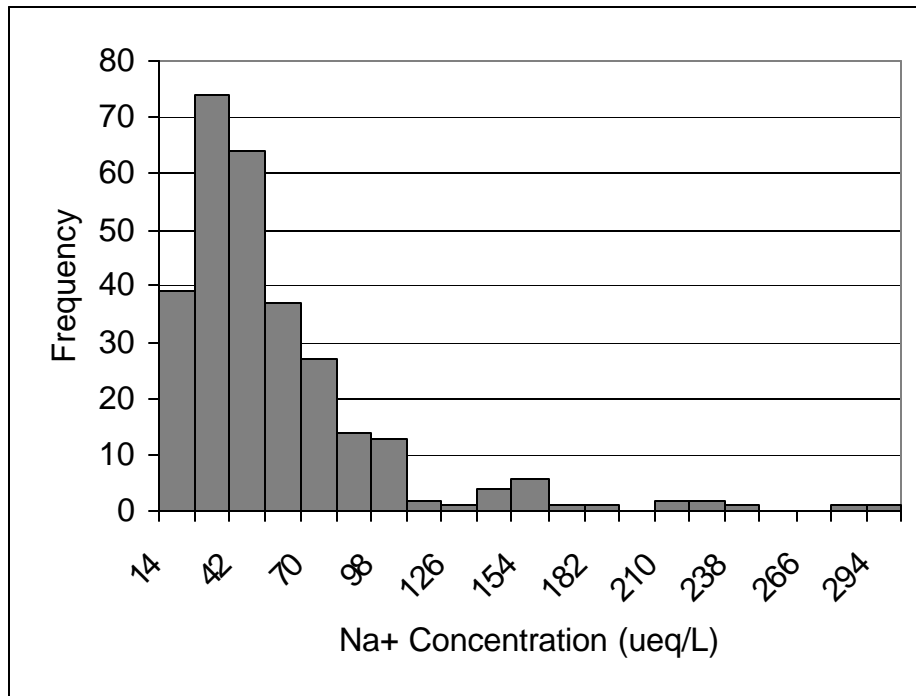


Figure 15. Histogram of distribution of sodium concentration among all events.

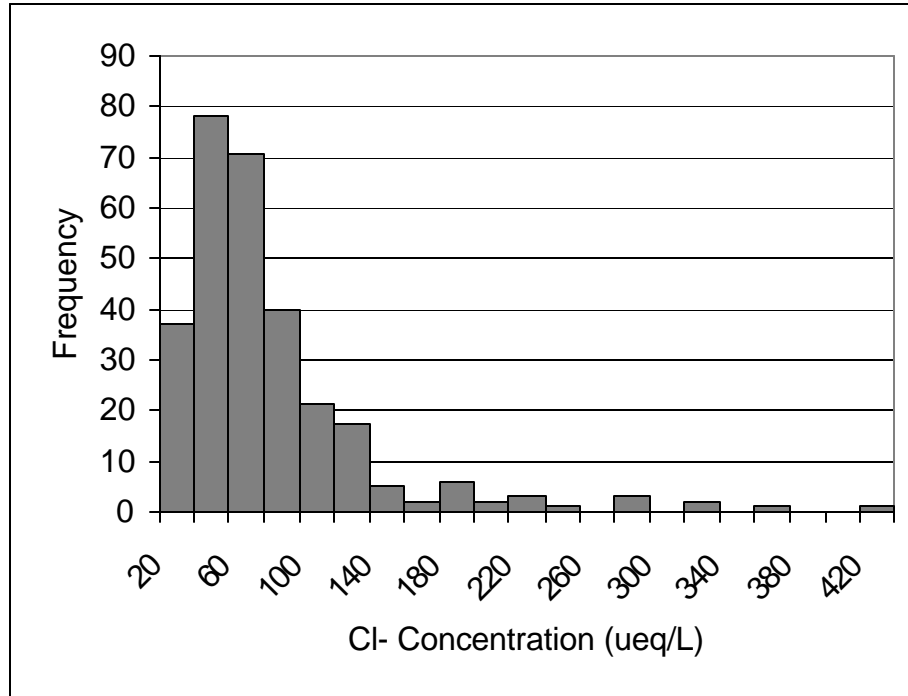


Figure 16. Histogram of distribution of chloride concentration among all events.

General Statistics

General statistics for precipitation and all ion data are shown in Table 3.

As demonstrated by the histograms, the general statistics show the large amount of variability that exists in the upper extremes of the data. The standard deviations for all data are nearly equal to the magnitudes of the means, and the mean values are larger than the medians for all cases.

The highest average ion concentration was seen for chloride at 63.2 ueq/L, and was likely the result of the proximity of the collection site to the saline waters of Tampa Bay. The next highest average concentrations were observed in non-sea salt sulfate and hydrogen ions at 52.6 ueq/L and 51.6 ueq/L, respectively. The high concentration of non-sea salt sulfate seen along with high

concentrations of hydrogen ions is typically associated with anthropogenic combustion sources seen in urban areas.

With regards to nitrogen-contributing ions deposited at the collection site, the average concentration of nitrate ions was 24.1 ueq/L, while that of ammonium was calculated at 14.7 ueq/L.

Volume weighted averages for all ions were always less than their means. VWA concentrations for nitrate and ammonium were 15.4 ueq/L and 10.5 ueq/L, respectively.

Table 3. Mean, standard deviation, median, and volume weighted average (VWA) values of precipitation depth and ion concentration calculated for all events.

	Mean	Standard Deviation	Median	Volume Weighted Average
Precipitation (mm)	14.8	16.5	9.9	N/A
H ⁺ (ueq/L)	51.6	44.4	37.0	41.2
nssSO ₄ ²⁻ (ueq/L)	52.6	40.4	38.6	38.4
NH ₄ ⁺ (ueq/L)	14.7	14.0	10.8	10.5
NO ₃ ⁻ (ueq/L)	24.1	21.8	17.3	15.4
Ca ²⁺ (ueq/L)	17.2	17.5	11.5	9.3
Mg ⁺ (ueq/L)	12.0	10.0	8.9	9.3
K ⁺ (ueq/L)	1.6	1.5	1.2	1.1
Na ⁺ (ueq/L)	47.3	42.8	35.2	38.0
Cl ⁻ (ueq/L)	63.2	56.3	47.0	48.8

Trajectory Results

The percent distribution of rainfall events by HYSPLIT trajectory classification is shown in Figure 17. The highest percentage of the 290

precipitation events arrived via the Cuba trajectory from the south of the collection site. There were a total of 82 events that originated from a Cuba trajectory that accounted for 28% of all data. The Bahamas trajectory accounted for the second highest percentage of the trajectories with 23%, and was followed by the Gulf-originating events with 20% of the total. Cape trajectories contributed to 13% of the data points, while Tampa and Panhandle-classified trajectories were split evenly at 8% each.

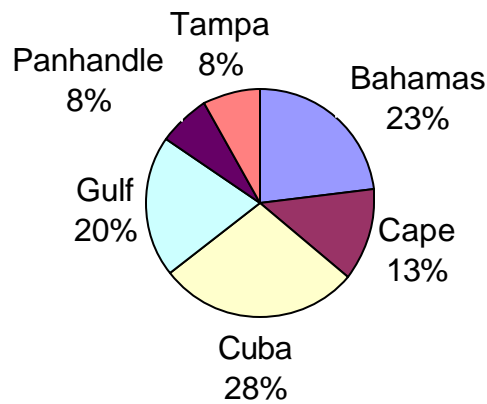


Figure 16. Distribution of precipitation events amongst HYSPLIT trajectories.

Ion Correlation

The ion correlation matrix for the tested ions is shown in Table 4. The greatest degree of correlation was seen between sodium and magnesium (0.97), chloride and magnesium (0.95), and sodium and chloride (0.95). Potassium was also shown to have a high degree of correlation with sodium, chloride, and magnesium with correlation coefficients of 0.86, 0.88, and 0.88, respectively.

These strong correlations between sodium, chloride, magnesium, and potassium were as predicted, since the major source of contribution for all of these items is known to be sea water. The close relationship among them is preserved because of the proximity of the collection site to Tampa Bay.

Non-sea salt sulfate concentration had a strong correlation to hydrogen ion concentration with a correlation coefficient of 0.91, indicating that they were likely deposited as sulfuric acid particles. Deposition of sulfuric acid is a strong signal of a nearby urban combustion source. Nitrate had a close correlation with both non-sea salt sulfate (0.84) and with hydrogen ion (0.82) indicating that it was likely emitted along with these two anthropogenic ions, and possibly originated from a similar local combustion source.

Ammonium ions were moderately correlated with calcium (0.64), non-sea salt sulfate (0.63), and nitrate (0.60). This indicates that ammonium may be transported from distant sources. Since the primary source of calcium in the region is from terrestrial soils, high wind days that are capable of making calcium-containing particles airborne are likely the greatest contributors to increases in calcium ions in wet deposition samples. At the same time, ammonium sulfate in the form of a very minute particle may be transported great distances when the wind speed is high.

Table 4. Correlation matrix of ion concentrations for all data.

	H^+ (ueq/L)	nss SO_4^{2-} (ueq/L)	NH_4^+ (ueq/L)	NO_3^- (ueq/L)	Ca^{2+} (ueq/L)	Mg^+ (ueq/L)	K^+ (ueq/L)	Na^+ (ueq/L)	Cl^- (ueq/L)
H^+ (ueq/L)	1								
nss SO_4^{2-} (ueq/L)	0.91	1							
NH_4^+ (ueq/L)	0.39	0.63	1						
NO_3^- (ueq/L)	0.82	0.84	0.60	1					
Ca^{2+} (ueq/L)	0.29	0.56	0.64	0.62	1				
Mg^+ (ueq/L)	-0.06	0.08	0.22	0.17	0.41	1			
K^+ (ueq/L)	-0.03	0.12	0.33	0.25	0.44	0.88	1		
Na^+ (ueq/L)	-0.11	-0.01	0.15	0.09	0.31	0.97	0.86	1	
Cl^- (ueq/L)	0.01	0.14	0.28	0.19	0.41	0.95	0.88	0.95	1

Multiple Regression Analysis

Multiple regression analysis was used to further determine with which ions nitrate and ammonium were deposited, and therefore, verify from which potential emission sources the nitrogen compounds entering Tampa Bay originated. The results of the multiple regression analysis between nitrate and all cations are shown in Table 5. The best multiple regression for nitrate was seen with calcium and hydrogen ions. Nitrate and hydrogen, both common in urban areas such as Tampa, were most likely deposited as nitric acid in rainfall. The association of nitrate with calcium is likely because both ions have sources that originate over land, though calcium is typically from natural sources and nitrate typically originates from man-made processes.

Results of the ammonium multiple regression analyses can be seen in Table 6. The best regression for ammonium was seen with chloride and nitrate

ions. Ammonia gas in the atmosphere reacts with both nitric acid and hydrochloric acid to form ammonium nitrate and ammonium chloride, respectively, and can travel large distances before deposition.

A regression analysis was also documented for non-sea salt sulfate since, though it does not contribute nitrogen, it is of anthropogenic origin and is commonly associated with the other ions of interest. This regression analysis only showed significance of non-sea salt sulfate with hydrogen ions, and therefore, concurs with previous analyses that the majority of non-sea salt sulfate is deposited as sulfuric acid.

Table 5. Results of multiple regression analyses for nitrate.

Regression Statistics					
Multiple R	0.93				
R Square	0.87				
Adjusted R Square	0.87				
Standard Error	7.48				
Observations	290				
ANOVA					
	df	SS	MS	F	Significance F
Regression	2	105630	52815	944	1E-126
Residual	286	15997	55.9		
Total	288	121627			
	Coefficients	Standard Error	t Stat	P-value	
Intercept	-0.92	0.60	-1.53	0.13	
Ca (ueq/L)	0.53	0.053	10.14	7.5E-21	
H+ (ueq/L)	0.28	0.009	29.13	1.38E-87	

Table 6. Results of multiple regression analysis for ammonium.

Regression Statistics					
Multiple R	0.84				
R Square	0.70				
Adjusted R Square	0.70				
Standard Error	8.37				
Observations	290				
ANOVA					
	df	SS	MS	F	Significance F
Regression	2	46366	23183	331	4.2E-75
Residual	286	20053	70.1		
Total	288	66419			
	Coefficients	Standard Error	t Stat	P-value	
Intercept	0.19	0.63	0.31	0.76	
NO3 (ueq/L)	0.52	0.025	21.0	9.7E-60	
Cl (ueq/L)	0.05	0.006	8.33	3.4E-15	

Table 7. Results of multiple regression analysis for non-sea salt sulfate.

Regression Statistics					
Multiple R	0.98				
R Square	0.96				
Adjusted R Square	0.96				
Standard Error	9.78				
Observations	290				
ANOVA					
	df	SS	MS	F	Significance F
Regression	1	671655	671655	7025	8E-204
Residual	287	27441	95.6		
Total	288	699097			
	Coefficients	Standard Error	t Stat	P-value	
Intercept	3.21	0.707	4.5	8.17E-06	
H+ (ueq/L)	0.85	0.010	83.8	8E-204	

Principle Component Analysis

Principle component analyses (PCA) for ion concentrations are shown in Table 8. Four factors were found explaining 95% of the total variance in ion results. Factor 1, which explained 51% of the variance, had high ion concentrations of magnesium, potassium, sodium, and chloride, and was likely the result of strong sea salt laden rainfall. Factor 2 explained 32% of the variance and had high concentrations of hydrogen, sulfate, and nitrate indicating the strong influence of a nearby combustion source. Factor 3, which explained 8% of the variation in the data, showed only high concentrations hydrogen ions. However, this factor also showed strong negative variances with calcium and ammonium. This may be the result of the difference in ions deposited on a strong wind day compared with a calm wind day. Hydrogen is likely building on calm days from local emissions, while ammonium and calcium may only be transported into the area from distant sources on days with a high wind velocity. Lastly, factor 4 explains 4% of the variance in the data and includes only local sources of ammonium.

Table 8. Eigenvalue and eigenvector results of the principle component analysis for all data.

Eigenvalues of the correlation matrix

Factor	Eigenvalue	Difference	Proportion	Cumulative
1	4.56	1.65	0.51	0.51
2	2.91	2.16	0.32	0.83
3	0.76	0.40	0.08	0.91
4	0.36	0.16	0.04	0.96

Eigenvectors

Ion	Factor 1	Factor 2	Factor 3	Factor 4
H ⁺	0.19	0.46	0.51	0.07
SO ₄ ²⁻	0.32	0.40	0.20	0.07
NH ₄ ⁺	0.29	0.25	-0.56	0.70
NO ₃ ⁻	0.30	0.40	0.12	-0.23
Ca ²⁺	0.34	0.16	-0.55	-0.66
Mg ⁺	0.38	-0.32	0.11	-0.06
K ⁺	0.39	-0.26	-0.01	0.08
Na ⁺	0.35	-0.36	0.18	0.02
Cl ⁻	0.39	-0.29	0.14	0.10

Annual Nitrogen Flux Results

The nitrate nitrogen flux for this study was calculated to be 2.1 kg/ha/yr, while the nitrogen flux for wet-deposited ammonium was 1.4 kg/ha/yr. These results were slightly less than the results seen for the same area presented in Poor et al. (2001). Poor found nitrate nitrogen flux to be 2.4 kg/ha/yr and ammonium nitrogen flux to be 1.7 kg/ha/yr. This discrepancy was probably due to the loss of data in this study for which ion chemistry data was incomplete or those for which HYSPLIT trajectories were not available.

A comparison with nitrogen flux data from previous studies showed that the annual average nitrogen flux for this analysis was generally less than results associated with precipitation in the northeastern United States. Luo et al. (2002)

found that, along the Connecticut coastline of Long Island Sound, nitrate nitrogen fluxes averaged 8.0 kg/ha/yr and ammonium nitrogen fluxes averaged 2.8 kg/ha/yr. A subsequent study by Luo et al. (2003) showed average inland and coastal nitrate and ammonium nitrogen fluxes throughout Connecticut to average 8.2 kg/ha/yr and 2.7 kg/ha/yr, respectively. Whitall et al. (2003) found that nitrate and ammonium nitrogen fluxes for the Neuse River Estuary in North Carolina averaged 3.5 kg/ha/yr each. It is well-recognized that high nitrogen fluxes are prevalent in the New England area of the U.S. associated with large electric utility sources, while along the Mid-Atlantic States, such as North Carolina, nitrogen emissions from agriculture contribute to these high fluxes.

In contrast Townsend (1998) found that the average nitrate nitrogen flux and the average ammonium nitrogen flux to the Gulf of Maine were 2.3 kg/ha/yr and 1.2 kg/ha/yr, respectively. These results were very similar to the results obtained in this study of the Tampa Bay area.

Average Standard Error for Flux Calculations

The calculations of standard error for flux measurements in this study are shown in Table 9. The results reveal that, for the 3.5 kg/ha/yr total nitrogen flux, there is an average annual nitrogen flux standard error of 1.4 kg/ha/d. The largest source of error for this type of measurement can be attributed to the variability in rainfall collection (%RSD=40).

Table 9. Standard error measurements for nitrate and ammonium nitrogen flux.

	RSD %	Avg. Annual N-flux (kg/ha/yr)	Avg. Annual Flux Standard Error (kg/ha/yr)
S NO ₃ ⁻	40	2.1	0.84
S NH ₄ ⁺	41	1.4	0.57
Total	40	3.5	1.4

Volume Weighted Concentration Averages by Trajectory

Volume weighted average (VWA) ion concentrations for all ions classified by trajectory are shown in Figures 18 through 26. Hydrogen ion concentrations were greatest in air masses originating from the Tampa trajectory and had a VWA concentration of 75.7 ueq/L followed by the Cape and Bahamas trajectories with VWA concentrations of 67.3 ueq/L and 57.2 ueq/L, respectively. This indicates that the highest concentrations of acid compounds are from sources located immediately within the local area, or from sources that have been transported from the eastern or southern regions of Florida. Cuba, Gulf, and Panhandle trajectories, all of which contact the Gulf of Mexico prior to arrival at Tampa Bay, showed the lowest VWA concentrations of acid compounds deposited.

VWA concentrations for non-sea salt sulfate showed a pattern similar to hydrogen ions. Cape, Tampa, and Bahamas trajectories showed the highest concentrations of non-sea salt sulfate at 66.7 ueq/L, 65.3 ueq/L, and 53.7 ueq/L, respectively. This was expected because of the good correlation of non-sea salt sulfate with hydrogen ions as shown in the previous analysis.

Ammonium wet deposition was dominated by air masses arriving from the Cape trajectory with a VWA concentration of 21.7 ueq/L. Bahamas and Tampa, which also originate over land, had the next highest concentrations of 13.6 ueq/L and 12.2 ueq/L, respectively. Ammonium is likely transported great distances across the peninsula from agricultural operations and fertilizer manufacturers located in the middle of the state, and is then deposited to Tampa Bay with precipitation.

Nitrate wet deposition was dominated by air masses originating within the Tampa Bay area. Similar to non-sea salt sulfate and hydrogen ion concentrations, Tampa trajectories contributed the highest concentrations of nitrate with a VWA of 30.4 ueq/L, followed by the Cape and Bahamas trajectories with concentrations of 28.4 ueq/L and 20.6 ueq/L, respectively. This demonstrates that emissions from within the Tampa Bay region contribute to the rainfall events with the highest concentrations of nitrate.

Calcium was primarily deposited from air masses originating from the Cape trajectory and had a VWA concentration of 19.8 ueq/L. Cuba and Gulf trajectories showed the lowest calcium concentrations with 6.3 ueq/L and 8.7 ueq/L, respectively. Since calcium is primarily emitted from crustal sources, this result was as expected.

Magnesium, potassium, sodium, and chloride all demonstrated their highest concentrations associated with air masses originating from Gulf trajectories, followed by air masses of Panhandle origin. Gulf and Panhandle trajectories both traverse rather large expanses of the Gulf of Mexico prior to

arrival in Tampa Bay and collect large concentrations of these sea salt ions. The lowest ion concentrations were deposited in association with Tampa trajectories, and were likely the result of the slow wind speed of these air masses. These calm winds were not as likely to aerosolize vast quantities of sea salt as is possible from other air mass trajectories.

Similar ion concentration distributions were observed by Norman et al. (2001) in India. In this study, anthropogenic pollutants were observed at higher concentrations from air mass trajectories that traveled across the continental land mass than from marine trajectories. Also similar, was the fact that Norman et al. found air masses local to the collection site experienced the highest concentrations of anthropogenic pollutants.

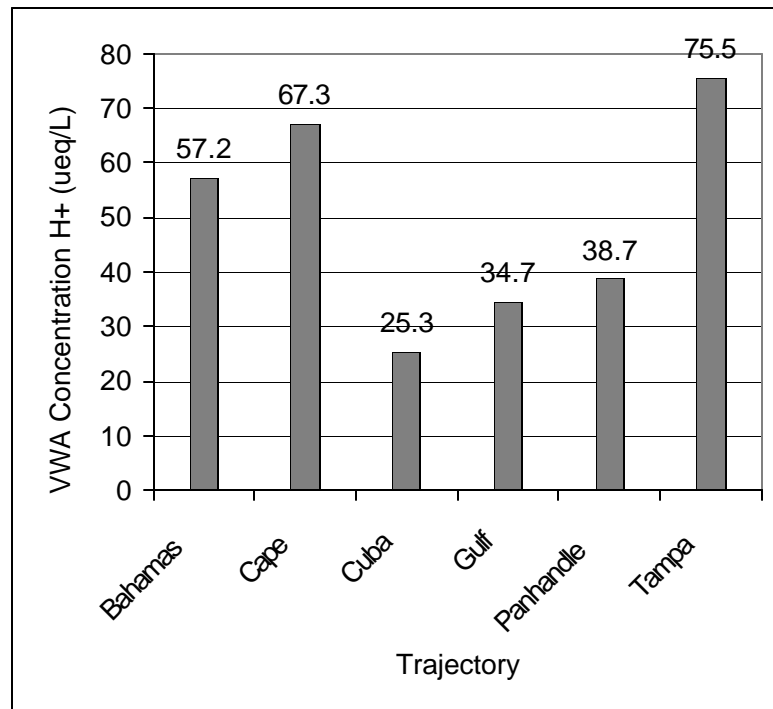


Figure 18. Volume weighted average hydrogen ion concentration classified by HYSPLIT trajectory.

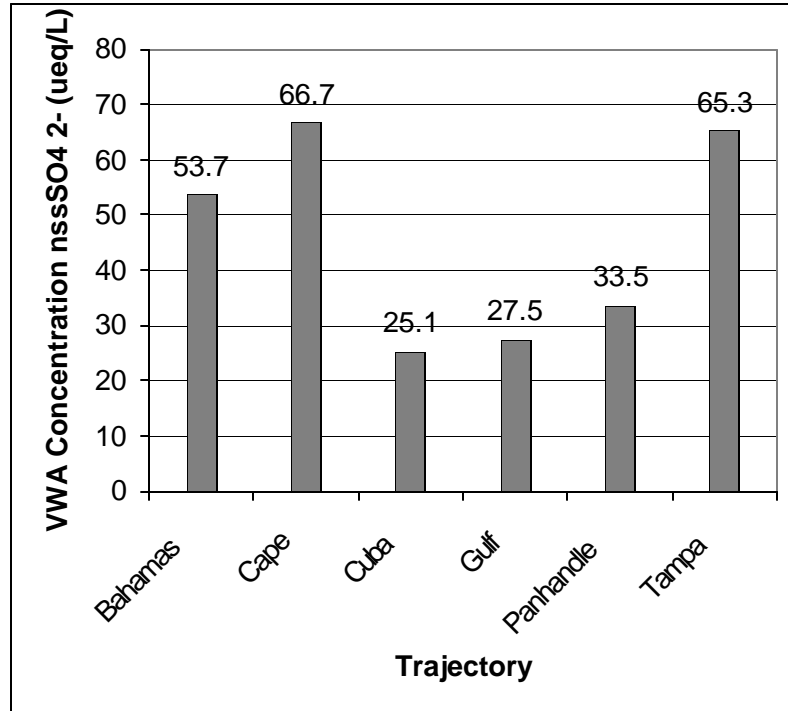


Figure 19. Volume weighted average non-sea salt sulfate concentration classified by HYSPLIT trajectory.

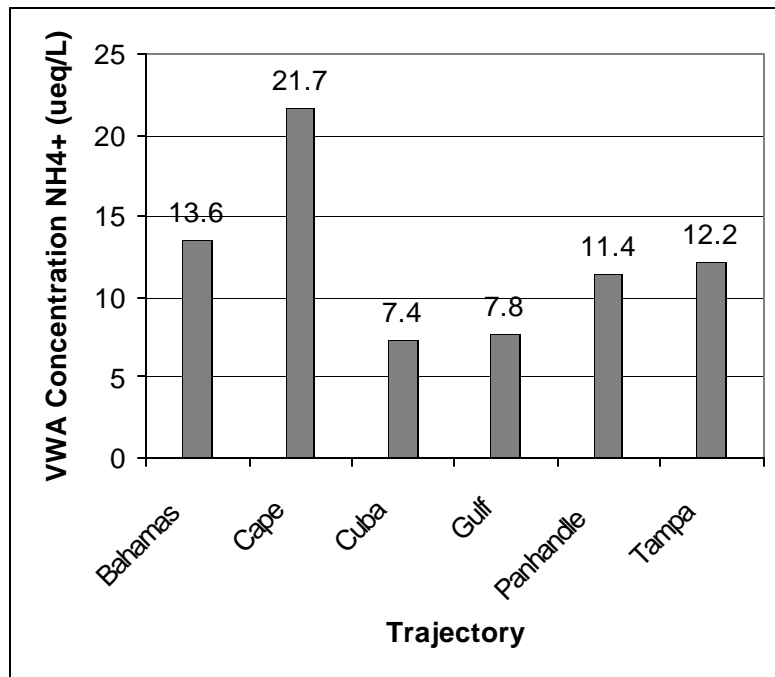


Figure 20. Volume weighted average ammonium concentration classified by HYSPLIT trajectory.

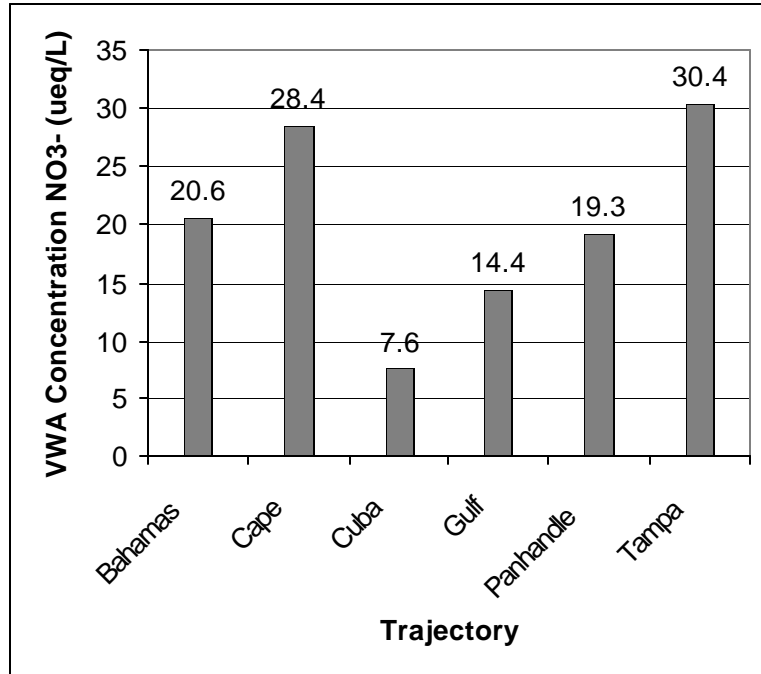


Figure 21. Volume weighted average nitrate concentration classified by HYSPLIT trajectory.

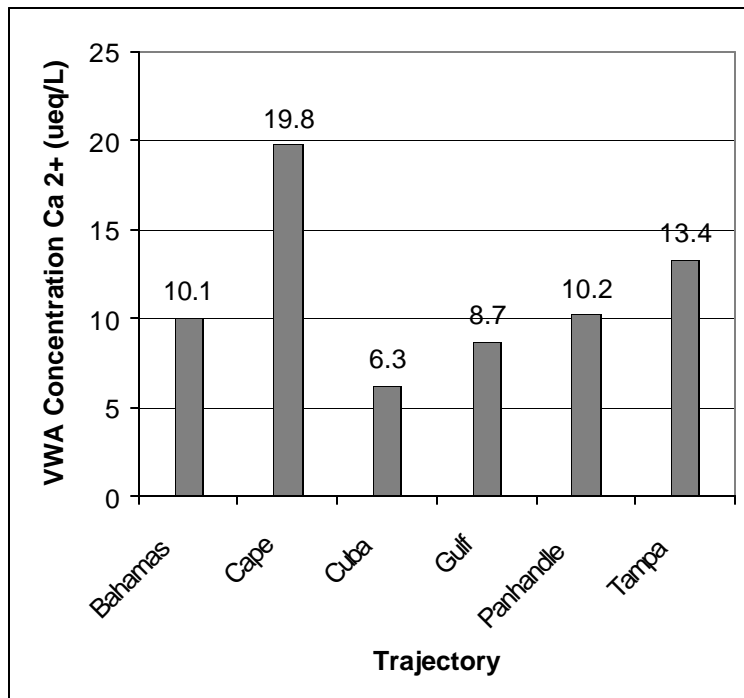


Figure 22. Volume weighted average calcium concentration classified by HYSPLIT trajectory.

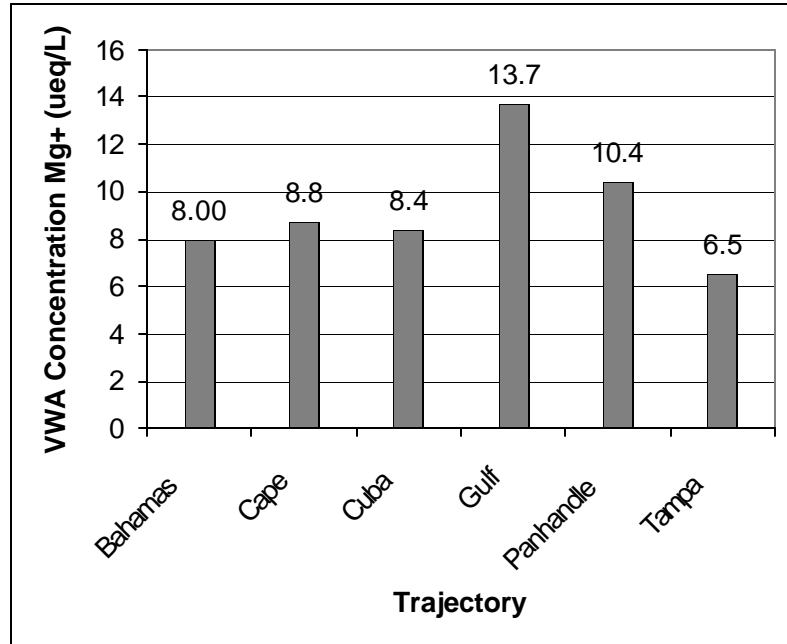


Figure 23. Volume weighted average magnesium concentration classified by HYSPLIT trajectory.

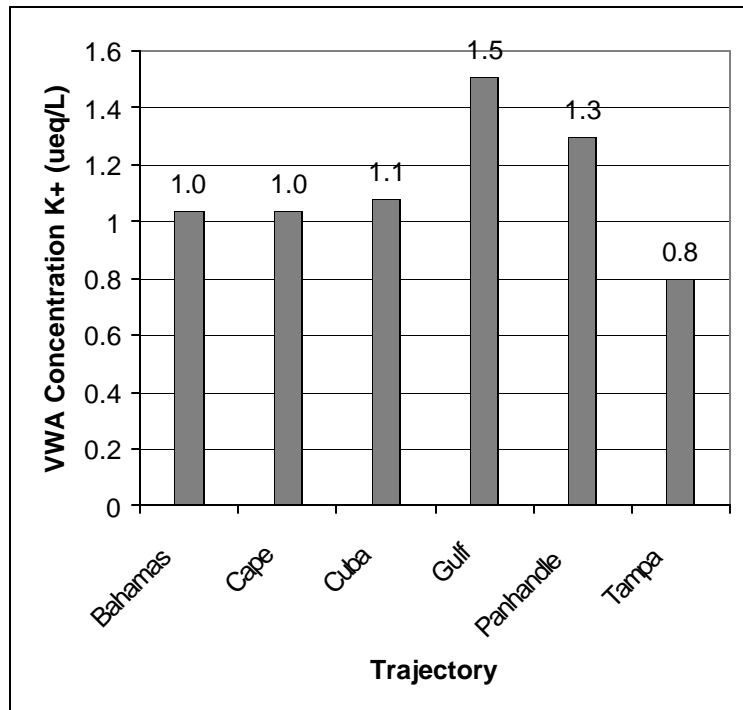


Figure 24. Volume weighted average potassium concentration classified by HYSPLIT trajectory.

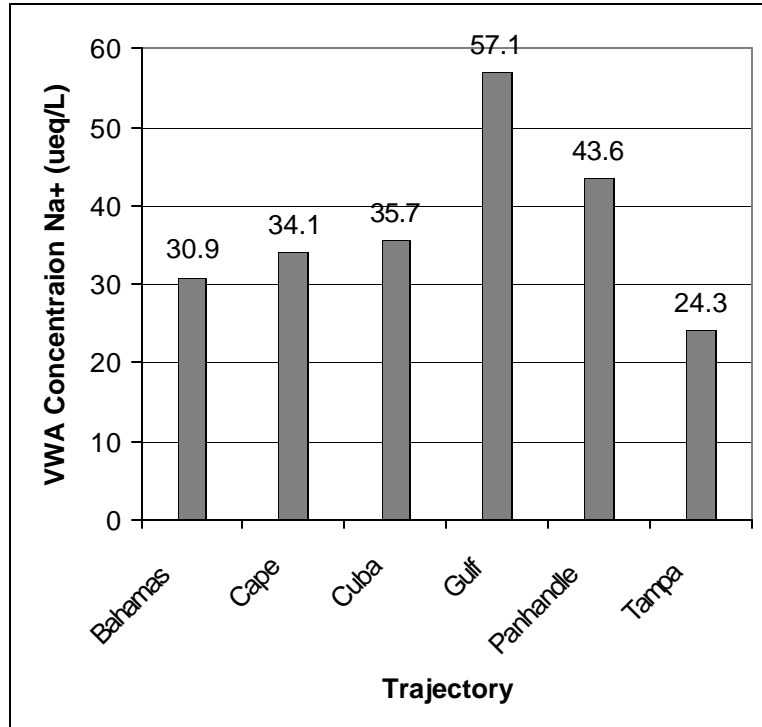


Figure 25. Volume weighted average sodium concentration classified by HYSPLIT trajectory.

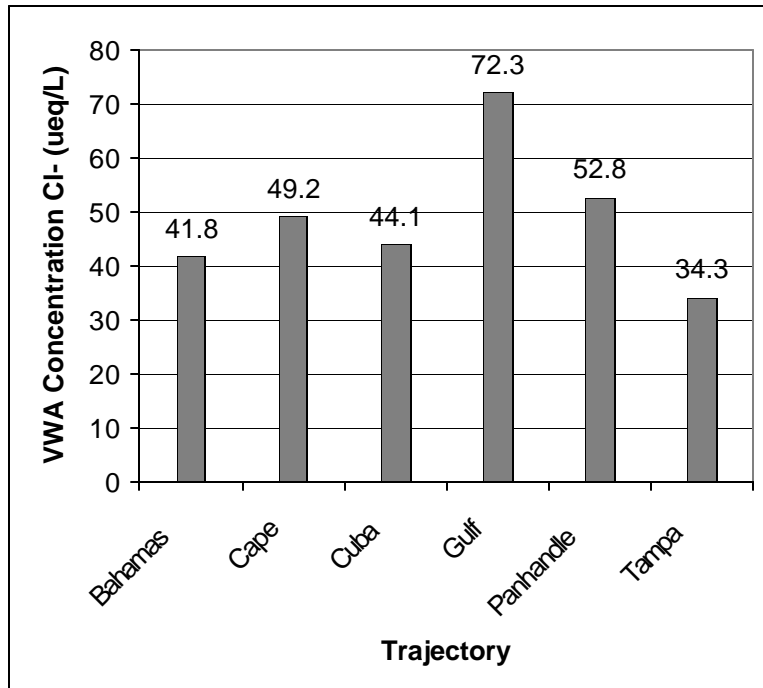


Figure 26. Volume weighted average chloride concentration classified by HYSPLIT trajectory.

Trajectory-Sorted Nitrogen Flux Calculations

The average daily nitrogen fluxes sorted by trajectory are shown in Figure 27. Nitrate N flux for all data averaged 0.032 kg/ha/d. The highest average nitrate nitrogen flux was from the Tampa-originating trajectories. These trajectories, which were typically slow moving and always remained within 100 miles of the collection site, had an average nitrate N flux of 0.050 kg/ha/d. Cuba and Gulf trajectories showed the lowest average daily nitrate N fluxes with 0.022 and 0.026 kg/ha/d, respectively. In general, the air masses that remained over land prior to arrival at the collection site had the highest average nitrate nitrogen fluxes.

Ammonium N-flux tended to follow the same pattern of higher values from land-based trajectories than from marine trajectories. The highest average ammonium nitrogen flux was from the Bahamas trajectory and deposited an average of 0.027 kg/ha/d. This was followed by air masses from the Cape and Cuba trajectories which deposited average nitrogen fluxes of 0.026 kg/ha/d and 0.022 kg/ha/d in the form of ammonium. The lowest average ammonium nitrogen flux resulted from precipitation events from the Gulf trajectory with an average flux of 0.014 kg/ha/d. This demonstrated that ammonia emissions from the Gulf of Mexico or from areas across the Gulf are of less importance than Florida-based sources. Of note is the observation that the ammonium N-flux for the Tampa-based trajectory was below the average of 0.0212 kg/ha/d at 0.020 kg/ha/d showing that ammonium is being transported more from distant sources than from local sources.

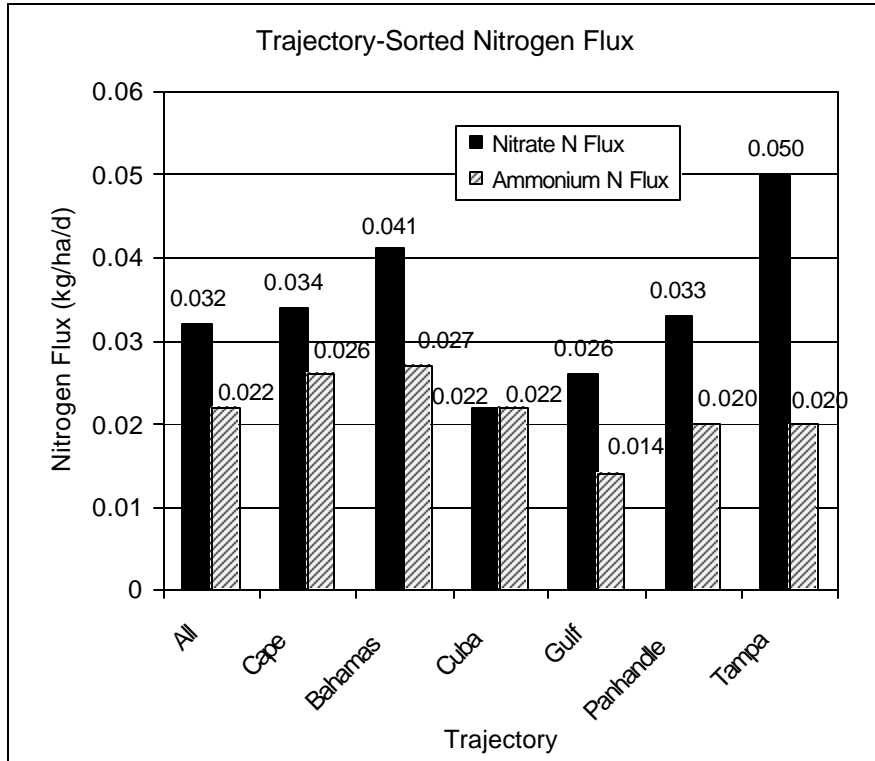


Figure 27. Average ammonium and nitrate nitrogen flux classified by HYSPLIT trajectory.

Kruskal-Wallis Analysis of Nitrogen Flux Results

Because data were determined to have a non-normal distribution, the Kruskal-Wallis ANOVA test was chosen as the acceptable method to compare nitrogen wet deposition flux between air mass trajectories. The Kruskal-Wallis ANOVA test is a rank sum test that compares three or more sets of data to determine if a significant difference exists between them. A significant difference was chosen as having a confidence level of at least 95%.

Results of the Kruskal-Wallis ANOVA test for the difference in deposition of nitrate nitrogen flux between air mass trajectories are shown in Table 9. The Kruskal-Wallis statistic was found to be 8.72 with a p-value of 0.121. Therefore,

the nitrate nitrogen flux between trajectories did not represent a statistically significant difference at the 95% confidence level. This may have been due to increased nitrate levels in all air masses as they passed through the Tampa area to the collection site which is located in the center of the urban area.

Table 10. Results of Kruskal-Wallis ANOVA test for significant difference of nitrate nitrogen flux between air mass trajectories.

n	290		
Trajectory	n	Rank sum	Mean rank
Bahamas	67	10474.5	156.34
Cape	38	5551.0	146.08
Cuba	82	11242.0	137.10
Gulf	58	7352.0	126.76
Panhandle	22	3497.0	158.95
Tampa	23	4078.5	177.33
Kruskal-Wallis statistic	8.72		
p	0.121		

The results of the Kruskal-Wallis ANOVA test of average ammonium nitrogen flux between air mass physical trajectories are shown in Table 10. The Kruskal-Wallis ANOVA test did show a statistically significant difference of ammonium nitrogen fluxes between air mass trajectories at the 95% confidence level. The Kruskal-Wallis statistic was calculated as 11.59 and the p-value was found to be 0.041.

Table 11. Results of Kruskal-Wallis ANOVA test for significant difference of ammonium nitrogen flux between air mass trajectories.

	n	290	
Trajectory	n	Rank sum	Mean rank
Bahamas	67	10509.0	156.85
Cape	38	5975.5	157.25
Cuba	82	12826.0	156.41
Gulf	58	6697.0	115.47
Panhandle	22	3197.5	145.34
Tampa	23	2990.0	130.00
Kruskal-Wallis statistic	11.59		
p	0.041		

Chemical Classification of Rainfall Events

The distribution of physical trajectories amongst chemical classifications is shown in Table 12. The marine chemical classification was dominated by the Gulf and Cuba HYSPLIT trajectories. These results are in keeping with the results determined previously that indicate the least influence of urban sources associated with air masses originating from these regions. The local combustion chemical signature was found to encompass primarily Tampa and Bahamas physical trajectories. These results were also as expected, since many local sources of urban air pollution are emitted from Tampa sources, and since the largest coal-fired power plant in the state of Florida is located to the southwest of the Tampa area. Air mass trajectories from the Cape region were primarily associated with the aged combustion chemical signature indicating influence of urban sources to the east across the state. These sources had time to react and

form the chemical species associated with aged combustion before deposition to Tampa Bay.

Table 12. Distribution of physical trajectory events among chemically-classified events.

	Total	Marine	Local Combustion	Aged Combustion	Mixed Terrestrial	Mixed Marine/Terrestrial
Bahamas	67	13	14	8	4	28
Cape	38	2	9	13	3	11
Cuba	82	39	0	7	3	33
Gulf	58	31	1	6	4	16
Panhandle	22	8	3	5	0	6
Tampa	23	3	12	1	1	6

Average nitrate and ammonium nitrogen fluxes calculated based on chemical classification of rainfall events are shown in Figure 28. The figure shows that the nitrate nitrogen flux was dominated by the local combustion events as expected. The average nitrate nitrogen flux for local combustion events was calculated as 0.060 kg/h/d. This nitrate nitrogen flux contribution from local combustion sources was nearly twice the average nitrate nitrogen flux contribution of 0.032 calculated for the entire data set. This result is in agreement with the close association that was observed between nitrate, hydrogen, and non-sea salt sulfate ions in previous calculations.

Marine-classified events demonstrated an average nitrate nitrogen flux of 0.023 kg/ha/d and an average ammonium nitrogen flux contribution of 0.018 kg/ha/d which were both lower than the average fluxes calculated for the entire data set of 0.032 kg/ha/d for nitrogen and 0.022 kg/ha/d for ammonium.

Once again, since there are no large anthropogenic nitrogen sources located in the marine environment, this result was expected.

Aged combustion sources showed average ammonium and nitrate nitrogen fluxes equal to 0.026 kg/h/d. This was the only instance in which ammonium nitrogen deposition was as high as the nitrate nitrogen deposition. These results may be due to the distance that ammonium can travel once it has reacted with other species to form extremely fine particles, and the high association of nitrate with acid species associated with local combustion.

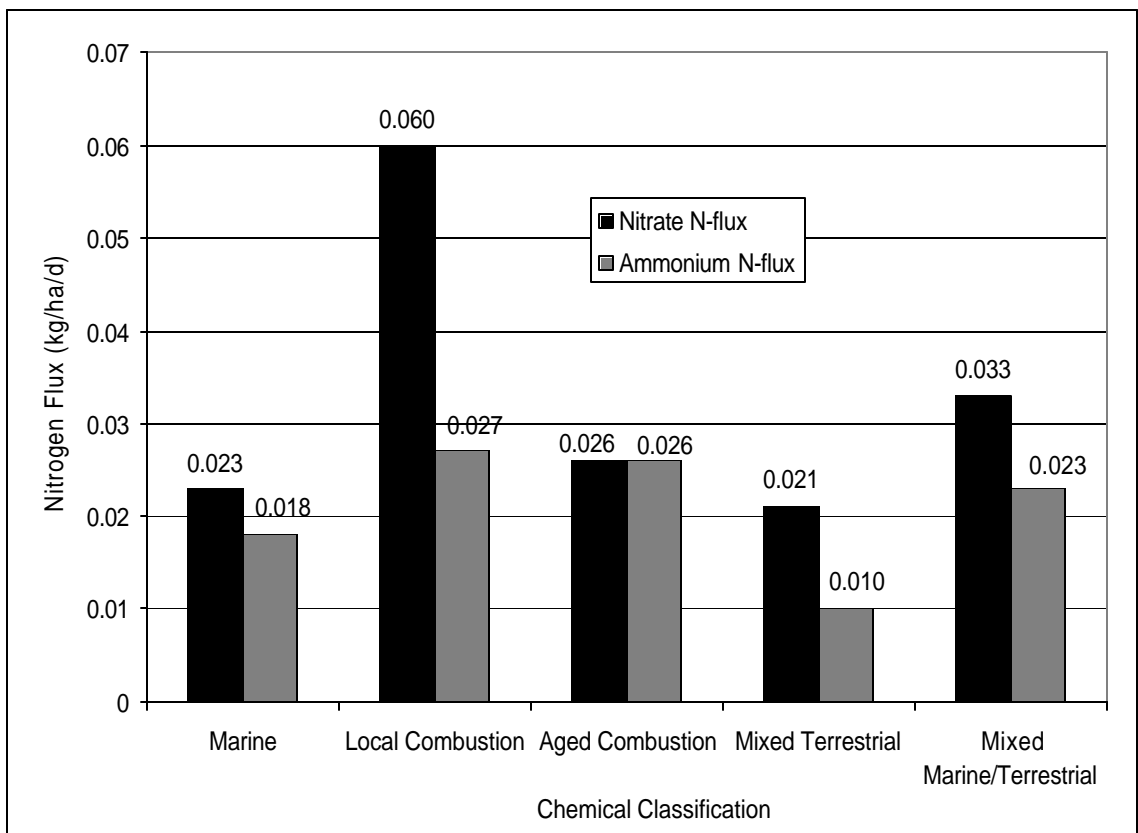


Figure 28. Average ammonium and nitrate nitrogen flux per event classified by chemical data.

Kruskal-Wallis Analysis of Chemically-Classified Data

The Kruskal-Wallis ANOVA test of chemically-classified nitrogen fluxes revealed that both nitrate and ammonium nitrogen fluxes exhibited statistically significant differences between chemical classifications. Results from the Kruskal-Wallis analysis for nitrate nitrogen flux are shown in Table 12, and results for ammonium nitrogen flux are shown in Table 13. These calculations resulted in p-values of 0.022 and 0.023 for nitrate and ammonium, respectively, and therefore indicated that there was a significant difference between chemically-defined classifications at the chosen 95% confidence interval.

Table 13. Kruskal-Wallis analysis of nitrate N-flux data among chemically-classified rainfall events.

n	290		
Classification	n	Rank sum	Mean rank
Local Combustion	39	7152.0	183.38
Aged Combustion	40	5459.5	136.49
Mixed Terrestrial	15	1792.5	119.50
Mixed Marine/Terrestrial	100	14816.0	148.16
Marine	96	12975.0	135.16
Kruskal-Wallis statistic	11.42		
p	0.022		

Table 14. Kruskal-Wallis analysis of ammonium N-flux data among chemically-classified rainfall events.

	n	290		
	Classification	n	Rank sum	Mean rank
	Marine	96	13456.5	140.17
	Local Combustion	39	6308.5	161.76
	Aged Combustion	40	6666.0	166.65
	Mixed Terrestrial	15	1328.0	88.53
	Mixed Marine/Terrestrial	100	14436.0	144.36
	Kruskal-Wallis statistic	11.34		
	p	0.023		

Nitrogen Deposition from Tropical Systems

Fifteen precipitation events from the Gandy Bridge Site data set were determined to be the result of tropical storms or hurricanes. The volume weighted average ion concentrations for tropical events are shown in Figure 29 along with the average ion concentrations for the set of non-tropical data for comparison.

The figure shows that higher VWA concentrations of anthropogenic pollutant ions were seen in non-tropical events than in tropical events including nearly three times higher nitrate concentrations for the non-tropical events. Higher concentrations of sea salt components, including sodium and chloride, were seen among tropical systems compared to events of non-tropical origin. These results were as hypothesized since the origin of tropical systems, by definition, is over the Atlantic Ocean or Gulf of Mexico in this region of the world. Because of the lack of anthropogenic sources of air pollutants located in areas

where tropical storms and hurricanes originate, as well as the purging associated with the large quantities of rainfall in these systems, the data sample consisted of precipitation that collected less of the anthropogenic pollutants before depositing rainfall in the Tampa Bay area. The small amount of nitrate and ammonium that were deposited by the tropical systems was determined to be a reasonable estimation of the background concentrations of both nitrate and ammonium for this region.

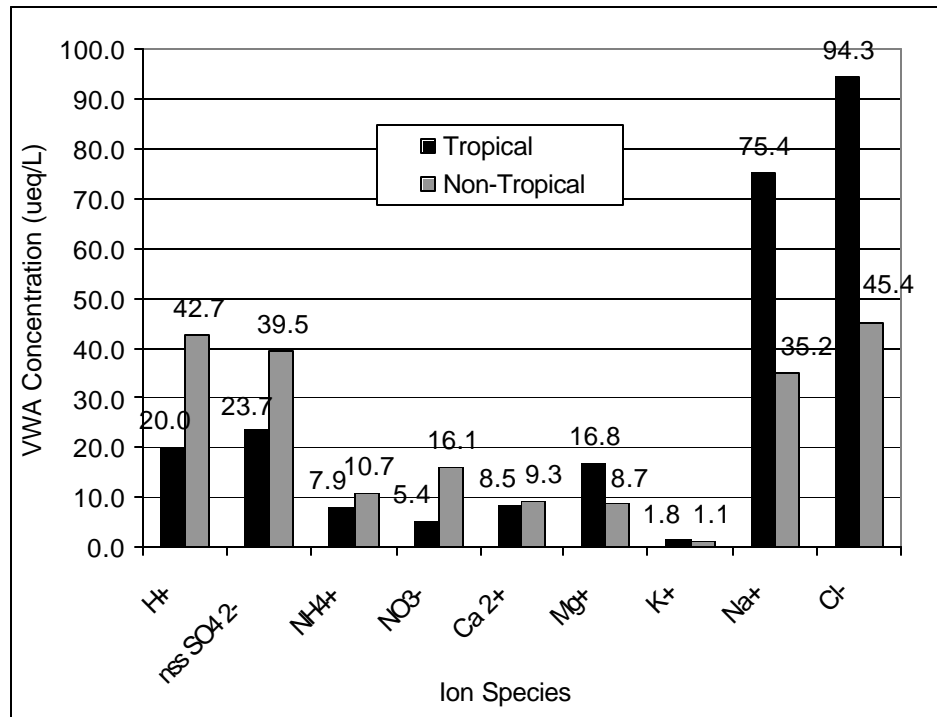


Figure 29. Volume weighted average ion concentrations calculated for both tropical and non-tropical precipitation events.

Average nitrate and ammonium nitrogen fluxes for tropically-influenced data were 0.015 kg/ha/d and 0.022 kg/ha/d, respectively, while non-tropical data

had average nitrate and ammonium concentrations of 0.033 kg/ha/d and 0.22 kg/ha/d, respectively.

Kruskal-Wallis Analysis of Tropical and Non-Tropical Data

Kruskal-Wallis ANOVA tests were performed on both the nitrate and ammonium nitrogen flux data between tropical and non-tropical events. The results for nitrogen nitrate flux are shown in Table 15. The p-value for this calculation was 0.090. Therefore, the flux difference found between these classifications was not significant at a confidence interval of 95%.

Table 16 displays the Kruskal-Wallis analysis results for ammonium fluxes characterized as tropical or non-tropical. Results indicate that there is also not a significant difference between ammonium nitrogen fluxes among these classifications. The calculated p-value for this test was 0.71.

Table 15. Kruskal-Wallis ANOVA test of nitrate N flux between tropical and non-tropical events.

n	290		
Trajectory	n	Rank sum	Mean rank
Tropical	15	1646.0	109.73
Non-Tropical	275	40549.0	147.45
Kruskal-Wallis statistic	2.88		
p	0.090		

Table 16. Kruskal-Wallis ANOVA test of ammonium N flux between tropical and non-tropical events.

n	290		
Ammonium	n	Rank sum	Mean rank
Tropical	15	2298.5	153.23
Non-Tropical	275	39896.5	145.08
Kruskal-Wallis statistic	0.13		
p	0.71		

Data Comparison

Table 14 compares ammonium and nitrate concentration results calculated in this study with background concentrations determined by Galloway et al. (1983). The marine chemically-classified air masses, the classification with the lowest concentrations of these two ion species, deposited rainfall with concentrations approximately twice the values determined by Galloway. These results demonstrate that even the “freshest” air masses arriving at Tampa Bay are depositing considerably higher concentrations of nitrate and ammonium than the backgrounds for the region. Also, by comparing the results of the Gulf HYSPLIT trajectory with the results from the other classifications for events that are less influenced by anthropogenic pollutants, it can be inferred that a significant plume of urban nitrogen pollution may exist over the Gulf of Mexico that is being transported back into the Tampa Bay region as air masses move from west to east. Additionally, emissions from Mexico or the southern United States may be carried to Tampa in these instances. Though the Gulf HYSPLIT

trajectories were expected to contribute the lowest nitrate and ammonium concentrations because they originated over open water with presumably the least influence of anthropogenic nitrogen emissions sources, the air masses originating from the Cuba trajectory may have actually been less influenced by anthropogenic emissions. Nitrate and ammonium concentrations from precipitation events originating from the Cuba trajectory were 12.5 ueq/L and 9.9 ueq/L, respectively.

Table 17. Comparison of nitrate and ammonium concentration results from “clean” precipitation events calculated in this study with backgrounds determined by Galloway.

	Marine	Tropical	Gulf	All Data	Galloway ^a
Nitrate (ueq/L)	11.5	13.7	21.6	24.1	5.5
Ammonium (ueq/L)	7.6	8.5	11.8	14.7	3.2

^a From Galloway et al. (1983).

CONCLUSIONS

According to HYSPLIT-calculated air mass trajectories, there appears to be a difference in the amount of nitrogen contributed to Tampa Bay between precipitation events traveling along various trajectories. Though the results obtained from average nitrate nitrogen flux measurements did not demonstrate a statistically significant difference between trajectories at the chosen confidence interval, the nitrate did appear to have a much greater average concentration and average nitrogen flux from Tampa or other Florida-originating trajectories than from trajectories traversing the Gulf of Mexico. These results reveal that the highest concentrations of nitrate are associated with sources near the bay itself, and that distant sources, such as those located in the rest of the continental U.S. or Mexico, contribute lower concentration of nitrate to the bay. These results also imply that reducing nitrate emissions from local combustion sources, such as coal-fired electric utilities or mobile sources, has the capacity to make a greater difference in reducing rainfall concentrations of nitrate to Tampa Bay than reductions in emissions from sources elsewhere.

Nitrate also showed strong associations with non-sea salt sulfate and hydrogen ion in correlation, regression, and PCA calculations as is typical of urban anthropogenic combustion sources. NO_x is likely emitted in conjunction

with non-sea salt sulfate and is likely deposited in the form of nitric acid. The close association with non-sea salt sulfate is another indication that nitrate collected in precipitation at the Gandy site is emitted from a local coal or oil-fired electric utility plants.

The repowering of the Tampa Electric Cooperative (TECO) Gannon power plant from coal to cleaner-burning natural gas should contribute to a significant reduction in the deposition of nitrate to Tampa Bay and help the Tampa Bay Estuary Program in its goals to revive the seagrass communities in the Bay. Also, the mandated reduction in sulfate emissions from the TECO Big Bend coal-fired power plant by addition of more efficient scrubber system should also reduce NO_x emissions as a byproduct and significantly reduce local nitrate deposition. The improvements to these two power plants are anticipated to lower the total atmospheric deposition of nitrogen to Tampa Bay by 12 tons per year over the next 10 years (TBEP, 2000).

Nitrogen deposited in the chemical form of ammonium did show a statistically significant difference in average fluxes between HYSPLIT trajectories. However, the nitrogen flux contributed in this form from the Tampa trajectory was less than the average ammonium nitrogen fluxes from the Bahamas or Cape trajectories. Results from the chemical trajectory analysis also indicated that ammonium deposition was greatest from distant emissions sources located across Florida to the east and southeast. These results were reasonable as there are several large fertilizer production facilities and large expanses of agricultural land located to east and southeast of the Gandy AIRMoN site.

In order to reduce the ammonium nitrogen wet deposition deposited to Tampa Bay, the most efficient method would involve controlling the fugitive ammonia emissions from these fertilizer production facilities, or by encouraging agricultural operations in this area to control reemission of ammonia from fertilizer application as well as from animal waste lagoons. Fertilizer application can be modified to include soil injection or other techniques that reduce the initial aerosolization of ammonia associated with fertilizer spreading, thereby reducing the reemission of ammonia from the soil after the fertilizer is applied. Also, encouraging the utilization of more efficient, covered animal waste lagoons that may be capable of generating power through the decomposition of animal waste materials could be useful means of reducing ammonia emissions from livestock operations.

The evaluation of ammonium concentration in rainfall deposited by tropical versus non-tropical weather systems also revealed that the average concentration of ammonium from tropical systems was lower than concentrations from the non-tropical events. This shows that high ammonium concentrations are not likely the result of natural marine sources or extremely distant ammonia sources, but are probably the result of either anthropogenic sources or terrestrial natural sources local to Florida.

In general, precipitation associated with marine air masses could be classified as “cleaner” with respect to anthropogenic pollutants than air masses that traveled across the Florida land mass. All data analysis in this study suggested that deposition of sodium, chloride, magnesium, and potassium were

associated with air masses that crossed the Gulf of Mexico prior to arrival at the collection site. This was evidenced by the general volume weighted average results as well as their association through regression and PCA analysis. The concentrations of sodium, chloride, magnesium, and potassium were also all shown to be greater with rainfall events that were associated with tropical events.

Future studies of interest would entail the use of AIRMoN daily rainfall collection sites located outside of the Tampa urban area. Results from these sites, potentially located to the east of the city in a rural area, or to the west over the Gulf of Mexico, could be coupled with AIRMoN data from the Gandy site and HYSPLIT trajectory data. By comparing the rainfall chemistry measurements in these proposed locations with the ion concentrations in samples collected at the Gandy site as an air mass moves across these reference points, a more precise understanding of the locations of nitrogen emission sources affecting Tampa Bay can be acquired.

In addition, after the repowering of the TECO Gannon power plant, and the addition of the updated scrubber system to the TECO Big Bend power plant, a similar study could also be performed to evaluate the potential change in nitrogen deposition. Results from a study of this type may be able to better anticipate the results that could be expected from future electric utility modifications in the Tampa Bay area or near other, similarly sensitive ecosystems.

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APPENDICIES

Appendix A – Source Locations



Figure 30. Map of the Tampa Bay area with the largest emissions sources of NO_x , ammonia, and SO_2 indicated along with the location of the Gandy Bridge AIRMoN site. (Reproduced from Poor et al., 2001)

Appendix B – Data Table

Table 18. List of Gandy Bridge AIRMoN Site ion data including HYSPLIT trajectories and classification of tropical system precipitation.

Date Off	Precip	H	nss SO4	NH4	NO3	Ca	Mg	K	Na	Cl	Tropical	Trajectory
	(mm)	(ueq/L)										
8/12/1996	10.922	26.51	30.44	6.1	12.9	11.98	12.43	1.36	47.85	57.54	No	Gulf
8/13/1996	17.272	17.52	26.7	7.21	12.42	17.96	20.16	2.15	78.73	97.03	No	Gulf
8/19/1996	16.002	66.59	72.04	24.39	31.77	34.93	38.59	4.14	156.6	206.19	No	Cape
8/20/1996	3.048	81.93	109.35	21.62	26.13	48.4	13.99	1.66	46.11	64.31	No	Cape
8/24/1996	1.016	49.37	83.31	25.5	45.8	89.82	17.12	1.92	58.29	83.49	No	Cape
8/27/1996	0.508	241.79	200.58	16.63	105.8	73.35	17.44	2.02	63.07	120.72	Yes	Tampa
9/2/1996	7.874	167.28	139.4	11.09	53.22	15.47	5.6	0.61	21.49	40.33	No	Cape
9/10/1996	1.778	63.6	76.78	8.87	15.16	19.46	4.11	0.33	10.7	17.77	No	Bahamas
9/11/1996	6.096	18.77	25.91	4.44	12.42	17.47	12.34	1.18	47.41	60.92	No	Bahamas
9/12/1996	16.51	31.87	29.63	2.77	8.87	4.99	6.58	0.82	26.97	34.98	No	Cuba
9/17/1996	4.826	30.44	38.6	9.98	12.1	11.98	14.32	1.84	52.63	61.49	No	Cuba
9/19/1996	0.508	16.73	30.73	31.05	27.42	21.46	24.52	2.97	104	121.85	No	Gulf
9/21/1996	5.08	15.61	32.54	11.64	20.81	26.95	7.16	0.56	15.01	18.05	No	Cape
9/22/1996	13.208	8.98	7.2	4.44	4.52	7.98	32.67	3.07	145.3	163.03	No	Gulf
9/23/1996	22.352	14.91	11.5	3.33	5.81	4.49	17.77	1.79	78.73	95.34	No	Gulf
10/3/1996	4.064	96.26	80.3	14.41	17.1	19.96	12.84	1.25	52.2	95.05	No	Bahamas
10/5/1996	0.254	45.02	68.46	27.72	22.1	35.93	17.03	2.28	72.64	104.64	No	Cape
10/6/1996	3.302	23.63	19.55	6.65	4.52	5.99	12.67	1.18	55.24	75.31	Yes	Cape
10/7/1996	14.224	43	37.51	8.87	3.71	2	3.46	0.38	15.22	27.64	Yes	Cape
10/8/1996	43.688	3.19	11.65	3.33	2.42	18.46	63.36	6.39	280.6	349.19	Yes	Gulf
10/18/1996	16.51	27.76	25.39	8.32	6.94	3.99	1.23	0.18	5.22	7.9	No	Cape
11/3/1996	2.032	15.97	33.75	11.09	16.61	40.92	17.36	1.87	65.25	92.52	No	Gulf
11/8/1996	7.62	5.17	10.12	6.65	5.97	10.98	6.09	1.2	24.79	28.77	No	Bahamas
11/9/1996	1.27	7.14	17.07	7.76	5.48	13.97	28.06	2.86	118.8	152.88	No	Gulf
11/22/1996	0.254	59.35	54.85	9.98	26.45	26.45	21.97	2.43	87	113.67	No	Gulf
11/26/1996	16.002	14.91	18.03	9.42	5	6.99	9.46	1.3	42.02	48.51	No	Cuba
12/2/1996	18.542	6.36	6.55	2.77	3.39	5.99	10.12	1.07	42.19	50.77	No	Cuba
12/8/1996	43.942	16.73	13.67	5.54	5.16	3.99	13	1.36	53.94	65.16	No	Gulf
12/15/1996	0.508	63.6	56.12	12.75	24.19	18.46	35.22	3.71	150.5	186.44	Yes	Panhandle
1/10/1997	10.414	9.41	17.56	9.98	8.55	20.46	34.97	3.48	140.5	170.36	No	Cuba
1/14/1997	1.016	30.44	27.72	9.98	25.64	14.47	14.98	1.64	60.03	68.82	No	Cape
1/26/1997	8.89	7.3	18.48	7.21	6.29	16.97	12.1	3.38	40.02	54.44	No	Cuba
2/9/1997	2.286	3.83	35.9	14.41	13.06	64.87	14.81	1.64	52.63	77.85	No	Gulf
2/11/1997	1.27	51.69	74.46	24.39	20	22.95	8.15	1.69	29.88	40.62	No	Panhandle
2/15/1997	12.446	13.29	17.06	10.53	6.77	10.48	11.68	1.64	48.28	58.67	No	Cuba
3/14/1997	16.002	3.66	18.93	12.75	10.81	28.44	10.12	1.38	32.8	40.62	No	Panhandle

3/21/1997	16.256	44	55.02	8.87	10.32	15.47	8.64	1.07	32.23	40.9	No	Cuba
3/22/1997	0.762	47.14	36.99	4.44	19.68	9.98	6.99	2.2	29.84	38.64	No	Gulf
3/31/1997	5.842	48.24	31.45	13.31	9.19	5.49	8.31	1.48	36.02	61.77	No	Gulf
4/8/1997	14.478	29.07	49.9	23.84	18.22	24.45	21.39	2.58	88.3	99.85	No	Cuba
4/12/1997	7.366	11.05	8.03	7.21	10.48	6.99	8.56	1.1	36.84	50.77	No	Bahamas
4/15/1997	32.766	38.32	33.95	14.97	14.19	3.99	3.54	0.9	15.4	18.9	No	Panhandle
4/24/1997	15.494	23.63	28.09	13.31	12.74	20.46	46.9	5.68	211.8	261.47	No	Cuba
4/26/1997	54.61	43	46.87	29.38	16.61	11.48	33.49	4.53	142.7	177.42	No	Bahamas
4/27/1997	13.462	13.6	8.02	6.65	4.52	2.99	6.25	1	26.66	31.87	No	Bahamas
4/28/1997	0.762	24.18	35.64	36.59	42.26	45.41	49.37	7.19	207.9	261.47	No	Cuba
4/29/1997	32.004	23.63	15.58	18.85	9.19	7.98	15.63	3.02	65.68	82.36	No	Cuba
5/12/1997	15.494	80.06	89.92	30.49	26.29	12.48	6.58	1.13	26.06	27.92	No	Bahamas
5/13/1997	19.812	22.56	19.61	4.99	5.48	1.5	2.14	0.41	10.05	11.85	No	Bahamas
5/29/1997	5.334	17.52	18.73	13.31	14.03	21.46	20.57	2.38	87.87	106.62	No	Cape
5/31/1997	1.778	73.02	116.38	68.74	38.87	35.93	8.64	2	29.32	49.36	No	Cape
6/10/1997	2.285	113.09	135.45	62.09	27.9	38.42	16.79	1.23	69.6	137.36	No	Cape
6/14/1997	6.349	56.68	63.13	4.44	16.45	19.96	21.97	2.3	90.87	116.21	No	Gulf
6/24/1997	16.001	91.93	74.73	18.85	41.29	19.46	6.42	1.94	22.49	37.23	No	Tampa
6/26/1997	3.556	135.97	116.02	14.41	48.38	16.97	10.86	1.43	44.37	57.54	No	Bahamas
7/3/1997	3.556	31.87	23.09	4.44	20.32	6.99	12.51	1.25	55.24	62.9	No	Panhandle
7/5/1997	5.333	108	97.12	22.73	49.03	11.98	4.28	0.69	14.75	20.59	No	Panhandle
7/6/1997	40.386	85.79	79.55	10.53	16.13	2.5	2.8	0.31	11.92	18.05	No	Cuba
7/7/1997	17.272	31.15	27.63	6.1	10.64	2.5	7.24	0.74	31.45	38.92	No	Cuba
7/11/1997	2.285	100.79	91.58	17.19	53.06	30.94	33.41	3.63	143.1	183.34	No	Tampa
7/12/1997	62.483	58	35.47	10.53	20.97	7.49	3.04	0.54	13.14	21.72	No	Gulf
7/15/1997	20.32	156.11	110.54	21.62	71.93	18.46	2.96	0.43	10.48	24.82	No	Tampa
7/16/1997	3.047	142.38	123.01	14.41	58.71	29.44	7.24	1.02	27.88	39.49	No	Tampa
7/19/1997	1.27	149.09	103.33	15.52	67.9	16.97	18.43	2.23	76.99	98.72	No	Bahamas
7/20/1997	24.637	62.15	56.39	12.75	16.45	3.99	4.2	0.61	17.49	23.98	No	Cuba
7/21/1997	14.224	31.15	21.22	4.44	10.32	3.99	8.89	1	38.02	46.82	No	Cuba
7/22/1997	9.143	69.73	49.44	4.44	19.68	4.49	1.97	0.26	6.13	8.74	No	Tampa
7/23/1997	10.16	124	94.51	24.95	67.09	21.46	3.7	0.49	12.22	31.59	No	Bahamas
7/25/1997	8.889	83.84	76.74	13.86	24.19	9.98	8.39	0.92	35.19	48.51	No	Bahamas
7/28/1997	0.254	76.46	86.84	17.74	48.38	36.43	20.49	2.02	68.73	78.69	No	Cuba
8/2/1997	2.286	41.06	48.04	6.1	24.35	18.46	10.53	1.23	41.8	50.77	No	Tampa
8/3/1997	4.826	9.19	23.49	2.22	14.35	22.46	19.5	1.59	63.94	77.57	No	Bahamas
8/4/1997	11.43	27.13	24.6	5.54	13.71	7.98	9.55	1.15	39.28	47.1	No	Cuba
8/5/1997	4.572	39.21	20.74	5.54	16.45	12.97	13.74	1.79	57.42	90.82	No	Cuba
8/6/1997	25.654	25.91	15.44	8.87	11.45	3.99	6.25	0.67	27.23	35.26	No	Gulf
8/8/1997	5.08	46.07	41.43	4.99	14.68	3.49	2.96	0.56	13.79	17.21	No	Gulf
8/12/1997	0.762	167.28	145.32	24.39	63.71	34.93	11.6	1.79	39.67	72.21	No	Tampa
8/17/1997	43.434	103.14	94.82	11.09	19.35	6.49	4.69	0.54	18.27	27.64	No	Bahamas
8/22/1997	6.858	49.37	23.71	5.54	24.03	6.99	19.5	2.12	94.83	115.08	No	Gulf

9/2/1997	3.556	76.46	79.01	20.51	23.22	12.97	6.25	0.67	24.97	42.31	No	Cape
9/4/1997	0.508	45.02	108.57	17.19	24.35	64.87	12.34	1.51	42.24	51.62	No	Gulf
9/14/1997	6.096	56.68	73.61	19.96	29.03	29.94	4.11	0.87	12.88	22.56	No	Tampa
9/18/1997	16.002	105.54	119.08	33.26	57.58	74.35	12.59	1.59	34.54	60.92	No	Cape
9/24/1997	14.732	42.02	57.46	23.84	22.1	24.95	8.31	1.02	25.84	41.18	No	Bahamas
9/26/1997	19.812	9.19	8.9	1.66	3.06	4.49	4.44	0.51	19.4	23.13	No	Cuba
9/28/1997	19.558	6.97	8.83	2.22	1.94	2.99	6.34	0.72	28.53	33.85	No	Cuba
10/2/1997	1.524	68.15	41.55	14.41	41.45	17.96	9.46	3.09	35.15	39.49	No	Panhandle
10/17/1997	6.096	32.62	55.01	33.82	43.87	46.91	11.27	1.56	40.98	60.36	No	Cape
10/19/1997	6.096	37.45	37.02	3.33	5.64	4.49	6.42	0.61	26.19	35.82	No	Tampa
10/25/1997	2.54	83.84	90.27	28.83	43.55	15.97	24.44	2.69	107.4	117.62	No	Cuba
10/28/1997	50.8	16.35	13.26	2.22	3.39	3.49	10.86	1.07	48.72	51.62	No	Cuba
11/1/1997	47.752	17.52	17.64	5.54	5.16	3.99	8.56	0.84	36.63	41.74	No	Cuba
11/7/1997	16.51	46.07	33.15	7.76	20	4.49	7.08	1.02	30.58	34.13	No	Tampa
11/13/1997	4.064	15.97	14.75	6.1	7.9	5.49	5.68	0.77	24.4	27.36	No	Bahamas
11/14/1997	61.976	14.91	15.42	1.66	1.45	3.49	5.6	0.69	24.01	28.77	No	Cuba
11/30/1997	11.43	29.75	32.26	3.33	3.71	2.5	3.54	0.74	15.57	19.46	No	Bahamas
12/1/1997	5.334	34.95	38.53	6.1	4.84	4.49	3.95	0.95	17.14	20.87	No	Cuba
12/4/1997	30.48	25.91	18.41	4.44	6.45	2.5	7.98	1.02	33.67	40.62	No	Bahamas
12/10/1997	5.334	25.91	31.24	10.53	11.77	12.48	7.32	0.95	30.88	34.98	No	Cuba
12/11/1997	62.738	19.65	21.98	4.44	5.64	3.99	7.73	1.15	33.45	39.77	No	Cuba
12/12/1997	15.24	35.76	39.64	5.54	6.61	4.99	8.39	1.38	37.23	44	No	Cuba
12/13/1997	76.2	34.95	23.9	2.22	2.26	2.99	1.89	0.2	8.96	21.44	No	Cuba
12/14/1997	25.4	25.91	22.72	2.22	3.06	3.49	3.46	0.38	15.27	19.18	No	Cuba
12/25/1997	13.97	22.05	19.53	7.21	10.32	8.98	15.8	2	69.16	75.59	No	Cuba
12/26/1997	20.066	24.74	17.73	6.1	8.39	3.99	11.68	1.64	51.33	56.41	No	Gulf
12/27/1997	94.488	23.63	23.88	7.76	3.55	2.99	5.27	0.84	22.92	29.62	No	Cuba
12/28/1997	1.016	51.69	53.39	19.96	17.58	15.97	47.23	9.05	204	237.78	No	Gulf
12/29/1997	1.016	85.79	61.92	16.63	51.61	9.48	22.14	2.51	92.22	109.44	No	Gulf
1/7/1998	7.62	65.08	77.46	20.51	43.71	47.41	22.05	4.12	73.95	94.49	No	Bahamas
1/8/1998	32.004	20.58	15.29	4.44	3.87	4.99	10.62	1.13	47.41	53.03	No	Cuba
1/15/1998	1.778	20.11	19.46	13.86	17.1	9.98	19.42	2.33	87	96.75	No	Bahamas
1/16/1998	13.208	8.98	13.91	4.44	5.48	6.99	7.57	0.95	33.01	40.33	No	Cuba
1/23/1998	25.908	11.57	7.48	3.33	3.87	2	2.39	0.33	10.44	12.41	No	Bahamas
1/24/1998	12.192	7.14	4.89	1.11	1.77	1.5	2.14	0.33	9.48	11.28	No	Bahamas
1/25/1998	0.762	73.02	106.16	55.99	55.96	18.96	14.24	3.27	58.72	59.51	No	Gulf
2/3/1998	48.514	23.09	28.64	7.21	5.97	12.97	7.32	0.95	26.58	36.39	No	Bahamas
2/7/1998	3.048	49.37	40.89	6.65	30.64	24.45	22.88	2.43	92.22	113.11	No	Panhandle
2/28/1998	7.366	29.75	34.36	16.63	14.84	16.47	18.43	2.99	77.43	91.95	No	Cuba
3/1/1998	24.638	45.02	38.86	19.96	15.48	9.98	13.17	2.4	53.94	69.67	No	Cuba
3/9/1998	27.178	16.73	22.97	21.07	11.13	13.47	16.54	3.04	68.29	79.82	No	Cuba
3/19/1998	46.482	15.26	10.96	4.44	7.74	3.99	4.69	0.56	19.53	23.69	No	Cuba
3/20/1998	53.34	33.38	32.13	7.76	8.55	3.99	4.53	0.82	18.4	23.41	No	Cuba

5/1/1998	4.572	14.57	38.9	37.14	18.22	39.92	17.36	3.04	65.68	80.11	No	Cuba
5/5/1998	1.016	46.07	93.52	76.5	110.96	99.3	38.92	6.16	130.5	159.08	No	Gulf
5/6/1998	20.828	23.09	27.78	22.73	14.84	13.97	6.91	1.36	25.05	33.28	No	Gulf
5/29/1998	57.15	29.75	23.85	1.66	5.48	2.5	3.46	0.38	14.57	17.77	No	Bahamas
5/31/1998	2.54	271.29	228.24	26.61	117.41	19.96	8.31	1.07	27.19	44.57	No	Tampa
6/25/1998	22.86	76.46	80.9	29.94	31.29	16.97	7.08	1.28	23.18	31.59	No	Bahamas
6/28/1998	14.224	89.83	59.43	17.74	25.8	8.98	11.11	1.28	49.15	53.87	No	Panhandle
7/7/1998	13.716	115.73	81.01	22.73	35.64	7.49	3.13	0.67	8.48	12.41	No	Cape
7/8/1998	73.152	40.13	28.28	10.53	14.03	5.49	6.09	0.74	24.36	31.59	No	Cuba
7/10/1998	42.418	33.38	24.17	3.33	9.68	3.99	5.68	0.59	23.92	30.18	No	Gulf
7/11/1998	26.924	11.05	5.74	1.11	4.19	2.5	7.9	0.77	33.45	41.18	No	Panhandle
8/7/1998	25.4	83.84	65.94	8.32	29.68	16.47	4.94	0.51	17.75	37.23	No	Tampa
8/8/1998	22.86	69.73	60.8	10.53	24.68	9.48	5.1	0.46	15.44	22.85	No	Bahamas
8/9/1998	10.922	56.68	45.02	8.87	21.45	5.49	4.61	0.59	18.53	25.39	No	Bahamas
8/10/1998	19.05	83.84	71.51	11.64	28.06	4.49	2.22	0.23	7.83	13.82	No	Cape
8/17/1998	11.684	167.28	144.62	11.64	65.48	19.46	8.23	0.82	28.27	44.85	No	Bahamas
8/18/1998	3.556	159.75	129.77	18.29	54.67	17.47	5.02	0.74	15.05	47.1	No	Bahamas
8/19/1998	9.398	52.9	53.63	19.4	20.97	10.48	4.44	0.69	16.22	19.74	No	Bahamas
8/20/1998	1.27	31.87	46.08	38.25	28.22	7.49	5.6	0.84	21.84	31.87	No	Bahamas
8/29/1998	1.016	149.09	168.12	53.22	77.09	44.41	9.55	1.51	25.01	51.9	No	Bahamas
8/31/1998	24.384	139.13	133.62	55.44	72.09	33.43	13.82	1.92	43.41	76.44	No	Bahamas
9/2/1998	10.668	18.77	23.28	8.87	10.81	11.48	6.67	0.92	26.1	32.15	Yes	Panhandle
9/3/1998	9.906	16.73	44.11	13.86	17.26	28.44	44.43	4.81	179.2	217.75	Yes	Cuba
9/4/1998	6.35	12.12	9.4	1.66	5	3.99	13	1.48	56.55	68.26	Yes	Cuba
9/6/1998	0.508	100.79	136.58	19.4	46.45	44.91	22.05	2.84	82.65	114.52	No	Bahamas
9/9/1998	15.24	46.07	40.21	3.88	17.74	7.49	4.03	0.43	16.96	22.56	No	Bahamas
9/10/1998	6.858	74.72	65.24	4.44	23.55	8.98	8.89	0.97	38.97	53.03	No	Tampa
9/16/1998	5.588	6.36	8	6.1	6.29	5.99	2.63	0.28	11.31	14.1	No	Bahamas
9/18/1998	6.35	27.76	31.31	4.44	5.64	3.49	5.76	0.67	25.14	30.46	No	Cuba
9/19/1998	19.812	27.13	25.95	1.66	3.23	2.5	5.43	0.46	23.01	28.21	No	Cuba
9/20/1998	32.004	11.57	11.36	2.22	2.42	2.99	5.92	0.54	26.53	31.31	No	Cuba
9/21/1998	16.51	38.32	59.16	14.97	6.94	8.98	8.15	0.87	34.15	39.77	No	Cuba
11/5/1998	28.956	19.65	23.82	22.73	4.52	3.99	1.56	0.26	6.18	19.74	Yes	Cuba
12/14/1998	10.668	7.47	23.81	4.44	5	19.96	14.07	1.74	64.81	64.31	No	Cuba
12/27/1998	1.778	63.6	72.59	7.21	23.55	16.47	34.64	3.22	93.52	161.9	No	Gulf
12/30/1998	9.652	16.35	18.43	3.88	6.45	5.49	10.86	1.13	33.49	54.16	No	Gulf
1/3/1999	21.082	12.12	17.06	4.44	3.87	10.48	34.97	3.56	129.2	172.62	No	Cuba
1/10/1999	13.97	17.92	16.7	4.99	6.45	7.49	20.08	2.25	87.43	105.49	No	Gulf
1/24/1999	49.276	5.54	3.38	1.66	2.26	2.99	6.75	0.74	30.58	35.82	No	Cuba
3/1/1999	4.318	4.61	44.3	12.75	18.39	45.91	31.76	3.48	74.38	126.36	No	Gulf
3/22/1999	0.508	76.46	106.79	14.97	46.13	36.43	45.92	4.96	132.7	210.98	No	Gulf
4/18/1999	9.652	46.07	70.09	23.28	35.48	35.93	20.32	2.58	60.9	84.62	No	Panhandle
4/29/1999	0.508	8.78	76.43	42.69	44.84	55.39	46.9	9.49	220.1	302.09	No	Gulf

4/30/1999	18.288	62.15	47.81	18.85	27.1	5.49	4.61	0.59	17.83	28.21	No	Panhandle
5/8/1999	10.668	12.4	48.8	31.05	21.13	43.91	18.43	3.09	56.11	81.52	No	Cuba
5/10/1999	1.27	100.79	93.41	52.67	83.06	39.92	20.32	5.01	53.94	110.29	No	Tampa
5/11/1999	6.096	81.93	93.98	21.07	43.06	30.44	13.08	1.82	21.75	57.54	No	Tampa
5/15/1999	10.16	18.77	28.78	13.31	12.26	15.97	25.1	2.94	82.21	129.47	No	Gulf
5/20/1999	2.794	100.79	123.98	26.06	60.32	63.37	17.44	2.46	49.15	80.11	No	Tampa
5/22/1999	33.782	98.5	91.88	24.39	41.61	17.96	13.08	1.71	39.1	77.85	No	Bahamas
5/31/1999	0.508	118.42	169.39	70.41	68.54	62.38	32.91	8.65	150.5	412.37	No	Cape
6/16/1999	67.31	91.93	76.78	11.64	32.9	7.49	3.62	0.41	12.48	19.74	No	Bahamas
6/17/1999	29.464	36.6	37.64	5.54	11.77	8.98	8.56	0.9	34.8	42.87	No	Cuba
6/18/1999	55.88	20.58	19.76	2.22	4.52	2.5	3.46	0.36	13.92	18.05	No	Cuba
6/19/1999	36.576	27.76	33.64	16.63	9.84	2.99	1.56	0.18	5.92	9.87	No	Cuba
6/20/1999	17.78	68.15	73.09	28.83	14.84	5.49	4.2	0.46	15.4	33.28	No	Cape
6/24/1999	2.794	124	108.92	19.4	62.09	33.43	18.51	2	61.77	102.67	No	Cape
6/26/1999	3.81	113.09	120.61	31.6	50	48.9	7.82	0.84	21.88	47.39	No	Tampa
7/1/1999	25.4	17.12	17.94	6.65	17.74	14.97	9.05	0.92	35.84	42.87	No	Cuba
7/2/1999	11.176	74.72	64.83	14.97	18.71	5.99	3.54	0.38	13.09	24.26	No	Cuba
7/4/1999	1.016	27.76	40.67	26.61	16.45	13.97	6.01	0.51	25.23	34.98	No	Bahamas
7/5/1999	16.256	36.6	31.55	9.42	6.61	3.99	3.87	0.41	16.31	28.49	No	Bahamas
7/10/1999	14.986	24.74	24.25	2.22	9.84	9.48	14.89	1.56	45.67	73.34	No	Bahamas
7/15/1999	6.35	121.18	105.58	13.31	45.16	13.47	4.69	0.41	22.23	25.67	No	Bahamas
7/16/1999	2.286	50.52	35.71	10.53	24.35	11.98	6.91	0.51	28.4	34.98	No	Bahamas
7/18/1999	2.032	105.54	118.45	41.02	30.97	64.87	12.18	1.48	39.71	108.31	No	Cape
7/19/1999	14.224	76.46	76.68	11.09	19.03	8.98	8.64	0.84	33.93	46.26	No	Bahamas
7/27/1999	7.874	73.02	76.43	8.87	32.74	24.95	7.32	0.79	27.4	41.74	No	Gulf
7/31/1999	14.732	40.13	32.7	6.1	22.26	6.99	10.04	0.97	39.5	50.77	No	Gulf
8/1/1999	18.288	115.73	96.24	11.09	42.74	11.48	6.91	0.74	25.45	35.26	No	Gulf
8/6/1999	0.762	66.59	76.62	9.98	45.32	38.42	8.06	0.84	30.97	38.36	No	Cuba
8/7/1999	28.702	11.31	8.94	2.22	3.71	4.49	4.11	0.36	17.31	21.15	No	Cuba
8/11/1999	8.128	38.32	39.72	7.76	14.68	14.97	9.63	1	36.49	48.51	No	Gulf
8/12/1999	21.082	25.91	18.06	4.44	11.29	4.99	11.44	1.15	43.5	60.92	No	Panhandle
8/15/1999	3.556	187.69	175.6	29.38	57.9	9.48	11.85	1.25	21.75	59.8	No	Cuba
8/17/1999	1.016	110.52	180.12	98.13	57.58	91.82	14.48	0.31	25.66	106.05	No	Bahamas
8/18/1999	19.304	28.41	29.35	9.42	21.77	20.96	2.8	0.26	8.7	13.26	No	Cape
8/19/1999	11.684	108	92.75	17.74	37.58	12.48	9.96	1	42.19	58.67	No	Cape
8/21/1999	10.16	32.62	43.61	19.4	14.19	10.98	9.71	1.02	42.24	53.03	No	Cuba
8/22/1999	85.09	45.02	43.73	3.88	7.9	3.49	3.04	0.2	11.96	14.67	No	Tampa
8/23/1999	10.16	37.45	28.57	1.66	15.64	3.49	4.28	0.43	18.57	22.85	No	Tampa
8/24/1999	6.096	66.59	68.59	2.22	16.29	8.98	5.84	0.69	25.05	35.54	No	Cuba
9/2/1999	11.684	15.26	13.14	2.77	10.48	13.97	9.05	0.9	39.41	46.26	No	Cape
9/6/1999	4.064	36.6	26.22	3.33	20.16	7.98	12.84	1.41	46.54	68.82	No	Gulf
9/7/1999	30.734	34.15	24.72	2.77	10.97	3.99	11.11	1.1	41.76	58.67	No	Gulf
9/8/1999	12.446	27.13	17.54	3.88	14.03	7.49	14.56	1.48	39.15	76.72	No	Gulf

9/9/1999	1.524	31.15	23.82	5.54	46.29	34.43	8.89	0.9	37.15	40.62	No	Gulf
9/12/1999	19.05	105.54	89.68	27.72	41.29	14.47	5.27	0.51	21.14	30.74	No	Cape
9/15/1999	7.366	0.75	8.29	3.88	7.26	24.45	14.4	1.59	67.42	74.46	Yes	Cape
9/18/1999	19.812	54.13	51.02	12.75	18.39	10.48	3.54	0.38	13.66	17.49	Yes	Bahamas
9/20/1999	24.13	26.51	32.34	10.53	5.16	3.49	2.72	0.41	11.53	14.39	Yes	Bahamas
9/21/1999	48.26	24.18	23.44	3.88	1.94	1	2.55	0.26	11.05	14.67	Yes	Cuba
9/26/1999	23.368	16.35	10.28	4.44	9.03	3.49	1.65	0.15	6.22	7.33	No	Cuba
9/27/1999	0.762	80.06	129.3	29.38	15.97	38.92	9.13	0.92	39.54	57.54	No	Bahamas
9/29/1999	2.794	56.68	95.6	48.79	45	44.91	13.58	2.12	3.22	78.98	No	Cape
11/25/1999	4.318	27.76	23.99	28.83	44.67	20.96	8.97	1.18	39.19	40.62	No	Cape
12/7/1999	1.016	15.26	35.52	13.86	19.51	22.46	11.03	1.28	26.53	55.57	No	Cuba
12/14/1999	5.588	10.08	24.64	4.99	4.68	15.47	5.68	0.72	23.53	29.05	No	Cuba
12/18/1999	12.954	29.75	25.9	7.76	9.19	3.49	1.48	0.18	6.18	13.82	No	Cape
12/19/1999	7.112	30.44	27.11	13.31	3.23	3.49	1.65	0.15	6.52	25.39	No	Cape
12/23/1999	1.016	14.91	36.81	11.09	25.8	57.39	9.22	0.92	36.49	41.18	No	Gulf
12/28/1999	3.556	38.32	31.82	11.09	25	4.99	12.01	1.28	29.58	57.54	No	Gulf
1/7/2000	11.938	21.55	31.97	18.85	14.35	16.97	8.31	1.56	33.49	41.74	No	Bahamas
1/11/2000	33.02	12.4	12.39	4.44	5.32	2.99	4.77	0.56	21.53	23.69	No	Cuba
1/24/2000	10.668	31.87	31.01	12.75	13.39	4.99	8.23	1.25	36.23	40.05	No	Cuba
1/25/2000	3.302	22.56	22.73	1.11	5.81	4.99	8.8	0.92	39.32	45.41	No	Cuba
2/2/2000	0.508	21.55	50.26	26.61	28.71	26.95	4.44	1.66	20.01	21.15	No	Panhandle
2/15/2000	5.842	20.58	28.41	9.98	9.84	11.98	13.25	1.51	43.93	65.44	No	Cuba
3/17/2000	1.016	20.58	45.86	20.51	27.74	51.9	51.84	7.16	269.7	304.91	No	Cuba
3/28/2000	14.732	18.77	25.22	11.64	13.87	13.97	21.07	2.51	75.47	102.67	No	Gulf
4/9/2000	4.064	22.05	21.31	7.21	18.22	9.48	9.87	1.53	38.97	43.44	No	Gulf
4/14/2000	5.842	16.73	18.41	7.76	10.64	12.48	7.73	1.53	33.67	38.08	No	Bahamas
6/12/2000	10.16	27.76	53.8	21.62	9.19	24.95	6.75	1.1	25.1	29.33	No	Cape
6/13/2000	1.016	129.85	142.05	47.12	85.8	38.42	15.63	2	61.55	83.77	No	Bahamas
6/15/2000	3.302	94.07	124.65	28.27	44.84	29.94	14.56	1.38	53.94	55.28	No	Bahamas
6/18/2000	22.86	38.32	43.34	15.52	10.81	7.49	8.48	0.77	35.84	43.44	No	Bahamas
6/19/2000	1.27	108	103.07	31.6	53.22	21.46	21.15	2.23	94.61	104.08	No	Bahamas
6/20/2000	33.528	91.93	81.98	29.94	53.54	21.96	14.89	1.56	60.68	67.13	No	Tampa
6/21/2000	18.542	91.93	72.1	14.97	39.35	8.48	3.54	0.36	13.31	17.21	No	Gulf
6/24/2000	36.576	38.32	34.76	9.98	13.71	3.99	2.14	0.28	8.7	11	No	Gulf
6/26/2000	29.21	36.6	31.58	14.41	16.61	4.99	3.13	0.36	12.57	14.67	No	Bahamas
6/27/2000	11.684	47.14	39.01	8.87	22.58	8.98	6.42	0.77	25.23	31.87	No	Bahamas
6/28/2000	2.794	58	56.45	16.63	28.22	8.98	5.27	0.77	23.92	29.62	No	Bahamas
6/30/2000	3.048	47.14	77.22	14.41	31.13	33.43	19.09	2.97	75.9	84.62	No	Cuba
7/1/2000	30.988	26.51	17.08	4.99	12.26	4.99	9.46	1.15	42.93	48.51	No	Panhandle
7/2/2000	1.016	142.38	112.01	19.96	36.13	20.46	8.39	2.74	32.75	85.75	No	Panhandle
7/5/2000	4.064	115.73	96.73	13.86	54.35	13.97	7.24	1.15	28.27	36.1	No	Bahamas
7/9/2000	18.542	103.14	88.46	19.4	29.03	7.49	7.49	0.95	31.19	36.39	No	Gulf
7/10/2000	40.64	103.14	94.3	34.37	40.97	11.48	11.03	1.33	50.02	62.9	No	Cape

7/14/2000	10.414	62.15	49.82	14.97	27.26	11.48	15.06	1.71	71.77	79.82	No	Gulf
7/16/2000	40.132	45.02	35.1	11.64	26.29	10.48	17.61	2	81.56	91.95	No	Panhandle
7/21/2000	1.524	132.87	111.4	46.57	71.77	38.92	13.99	2.1	53.29	59.8	No	Panhandle
7/24/2000	31.75	48.24	34.47	11.64	22.74	5.49	7.73	0.92	33.49	38.64	No	Gulf
7/25/2000	18.542	25.91	16.47	2.22	14.03	2.99	6.5	0.69	29.1	35.26	No	Cuba
7/27/2000	30.988	110.52	94.31	13.31	40.64	9.48	3.87	0.56	13.83	16.64	No	Bahamas
7/31/2000	1.778	135.97	98.31	22.73	89.03	25.45	12.51	1.76	49.59	58.67	No	Bahamas
8/1/2000	46.482	37.45	30.29	4.99	7.42	2.99	3.13	0.36	12.92	17.49	No	Bahamas
8/3/2000	1.524	297.46	200.75	10.53	126.77	24.45	7.98	1.15	27.27	78.41	No	Tampa
8/10/2000	1.778	76.46	112.52	22.18	110.8	108.78	16.21	2.23	50.89	58.1	No	Cape
8/12/2000	4.064	81.93	83.55	6.1	35.48	17.96	25.02	2.46	95.91	104.36	No	Gulf
8/13/2000	67.564	8.19	4.68	1.11	3.55	2.5	7.41	0.74	35.28	40.33	No	Gulf
8/23/2000	2.54	73.02	69.24	9.98	38.71	22.95	4.44	0.95	14.57	23.69	No	Cape
8/28/2000	2.794	49.37	42.47	26.61	21.61	9.98	3.29	0.84	12.05	14.67	No	Bahamas
8/29/2000	16.764	31.87	40.81	9.42	20.16	13.97	8.39	1.1	37.89	49.08	No	Panhandle
8/30/2000	9.906	59.35	50.61	21.62	37.9	13.97	15.06	1.92	66.99	75.03	No	Panhandle
9/1/2000	2.032	36.6	38.21	18.29	48.06	33.93	21.23	9.08	92	96.75	No	Cuba
9/2/2000	0.762	17.12	10.35	2.77	19.35	12.97	14.32	1.66	67.64	77.57	No	Gulf
9/5/2000	1.778	96.26	93.46	7.21	29.35	11.48	7.49	0.84	27.75	29.62	No	Gulf
9/6/2000	3.302	33.38	28.96	2.22	12.58	7.49	13	1.36	61.77	70.23	No	Cuba
9/7/2000	3.302	60.73	44.32	12.2	27.58	10.98	2.72	0.84	10.53	14.67	No	Cuba
9/8/2000	3.81	45.02	29.73	2.22	21.93	2.99	2.63	0.38	12.4	15.23	No	Cape
9/16/2000	5.08	27.76	31.12	4.44	12.42	10.98	4.94	0.64	23.31	26.51	Yes	Tampa
9/17/2000	52.832	13.91	16.76	5.54	3.06	3.99	5.02	0.72	23.27	28.77	Yes	Cuba
9/18/2000	16.51	5.67	13.7	3.88	2.74	15.47	43.78	4.99	229.2	268.52	Yes	Cuba
9/20/2000	15.24	42.02	46.84	12.75	17.42	12.48	15.22	1.71	65.46	73.05	No	Bahamas
9/21/2000	7.62	81.93	85.08	10.53	20.32	8.98	12.34	1.51	55.68	62.62	No	Bahamas
10/4/2000	1.27	38.32	45.57	11.09	27.26	21.46	6.34	1.46	22.62	33.57	No	Cape
10/21/2000	7.112	29.75	96.32	80.39	44.51	55.89	8.64	1.59	31.62	46.26	No	Cape
11/10/2000	3.048	21.55	31.08	9.98	11.61	11.48	16.29	2.02	71.77	79.54	No	Cuba
11/15/2000	10.414	18.77	18.76	3.88	7.9	5.49	8.06	0.87	35.97	40.9	No	Gulf
11/18/2000	5.08	23.09	23.19	8.87	13.06	8.48	13.41	1.46	61.33	70.52	No	Panhandle
11/26/2000	13.97	27.76	29.99	11.09	7.74	2.99	5.35	0.72	25.75	30.46	No	Cuba
11/27/2000	6.096	41.06	42.7	12.2	18.06	6.49	9.79	1.15	48.07	58.95	No	Gulf
12/13/2000	6.858	34.95	42.84	8.32	7.42	4.99	2.8	0.33	12.44	14.39	No	Cuba
12/16/2000	2.032	27.76	59.74	24.39	39.67	44.91	14.81	3.58	51.76	58.1	No	Bahamas
12/17/2000	7.112	10.8	11.29	1.66	3.39	3.99	13.17	1.3	63.29	70.8	No	Gulf
12/28/2000	7.62	40.13	44.12	10.53	7.26	4.49	9.05	1.1	38.06	50.21	No	Bahamas
12/29/2000	1.778	39.21	41.91	11.64	15.97	13.97	27.32	3.25	130.3	163.03	No	Gulf