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Period: 11/01/06 – 08/31/07

* Data for internal use only as the results and interpretations are still preliminary.

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1. Comparison of the Ambient Levels of NO, NO_2, O_3:

Based on the US EPA data, the major anthropogenic source of NOx (NO + NO_2) in the ambient air in the United States is motor vehicles emissions, accounting for ~55% of the total emissions. Additional sources of NOx include electric utilities (~22%) and other industrial, commercial, and residential sources. Nitrogen oxides form from fossil fuel burning at high temperature, as in a combustion process, which is a pre-cursor molecule of the ground-level ozone. Therefore, high concentrations of NOx and O_3 are expected to be found often in the areas heavily impacted by traffic and power plants, although these nitrogen containing species can come from natural sources and be transported over long distances by prevailing winds.

According to the preliminary data we collected and analyzed at the Meadowlands site, mainly focused on the spring 2007, the NO, NO_2 and O_3 levels at this location are within the ranges of both primary and secondary standards by NAAQS. The daily and 8-hours averages of NO, NO_2 and O_3 at the MERI site in spring were smaller than those listed urban sites and some other suburban sites (Table 1). The NO_2 concentration in Meadowland is relatively high and

even exceeds the NO concentration, which is different from other sites.

Table 1. Comparison of NO, NO_2 and O_3 in the Ambient air*

Sites	*NO(ppb)*	*NO_2 (ppb)*	*O_3 (ppb)*	*Sources*
EPA Standard 8-hours average	--	53	84	EPA website
MERI Site Daily Average	13.8	21.3	24.0	This work
MERI Site 8-hours Average	23.9	25.0	32.5	This work
Ramapo (May)			85	NJDEP
Flimington (May)			88	NJDEP
Chester (yearly, 2005)	4	11		NJDEP
Elizabeth (yearly, 2005)	42	32		NJDEP
Rural Site in UK	--	9.4	--	Clapp et al., 2001
Busy Street in UK	--	59	--	Clapp et al., 2001
Green Area of Buenos Aires City	240	65	30	Mazzeo et al., 2005
Southern California	2-76	23-38	9-81	Qin et al., 2004
Phoenix, Arizona	--	1-10	15-35	Gaffney et al., 2002

* Data may not be for direct comparison due to the different temporal measurement scales of the concentrations.

2. Effects of Weather Conditions on the Ambient Concentrations of NOx and O_3:

At a specific location, many factors may affect the ambient levels of atmospheric species of interest, and meteorological conditions sometimes play an important role on that aspect. To explore the role of meteorological effects on the ambient concentrations of NOx and O_3, we have focused on three meteorological parameters, the sky conditions, wind directions and speeds and relative humidity. Below are brief discussions on each.

(1) Effects of Sky Conditions:

Table 2 shows a brief comparison of the concentrations of NO, NO_2 and O_3 in clear, cloudy and rainy days. The NOx concentration was higher in rainy days but lower in clear day, and O_3 had the opposite trends with highest concentrations in clear days. The possible reason is that in clear days NOx could be easily spread out, and lowered NOx concentration together with higher O_3 concentrations could be an indicator of the VOC-sensitive characteristics at the MERI site. In VOC-sensitive region, O_3 would increase with VOC but decrease with NOx.

Table 2. Comparison of NOx and O_3 under Different Sky Conditions.

	Sky Conditions		
Species	***Clear days***	***Cloudy days***	***Rainy Days***
NO (ppb)	7.13	17.13	18.03
NO_2 (ppb)	16.02	21.40	26.32

NOx (ppb)	23.15	38.53	44.35
O_3 (ppb)	29.34	22.99	19.78

(2) Effects of Wind Directions and Speeds:

The wind direction may influence the concentrations of NOx and O_3. Figure 1 shows the concentration distributions of NOx and O_3 as a function of wind directions. Higher concentrations of NOx were primarily related to the west-south wind, indicating the possible high NOx discharge from the sources in that direction. For the O_3 levels, the south wind often led to higher O_3 concentrations, indicating the higher background O_3 from the south regions of the Meadowland site.

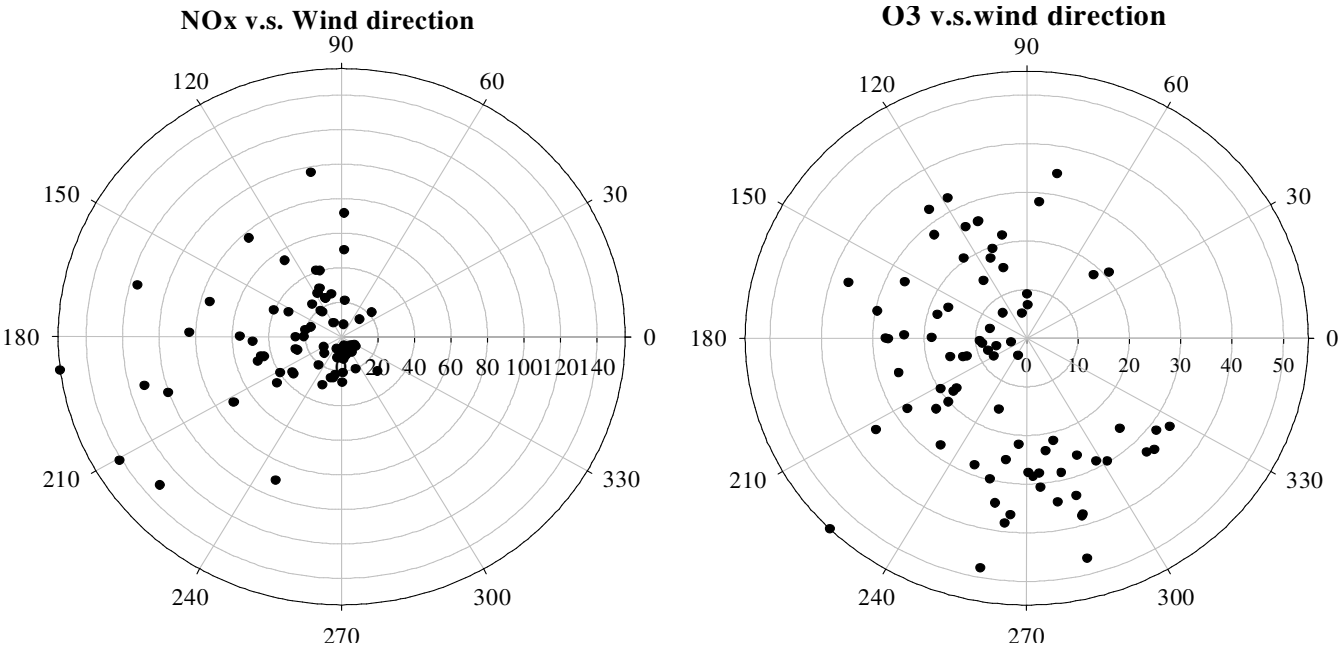


Figure 1. The influence of wind direction on NOx (left) and O_3 (right).

Wind speeds could also exert certain influences on the ambient levels of NOx and O_3 as shown in Figure 2. The NOx level decreased as the wind speed increased, indicating the dilution through wind mixing. The main sources of the NOx in this area seem local rather than regional. However, the O_3 concentration increased linearly with wind speed. The high speed wind could reduce the NOx level, and thus the O_3 production may increase because of the VOC-sensitive characters of the region.

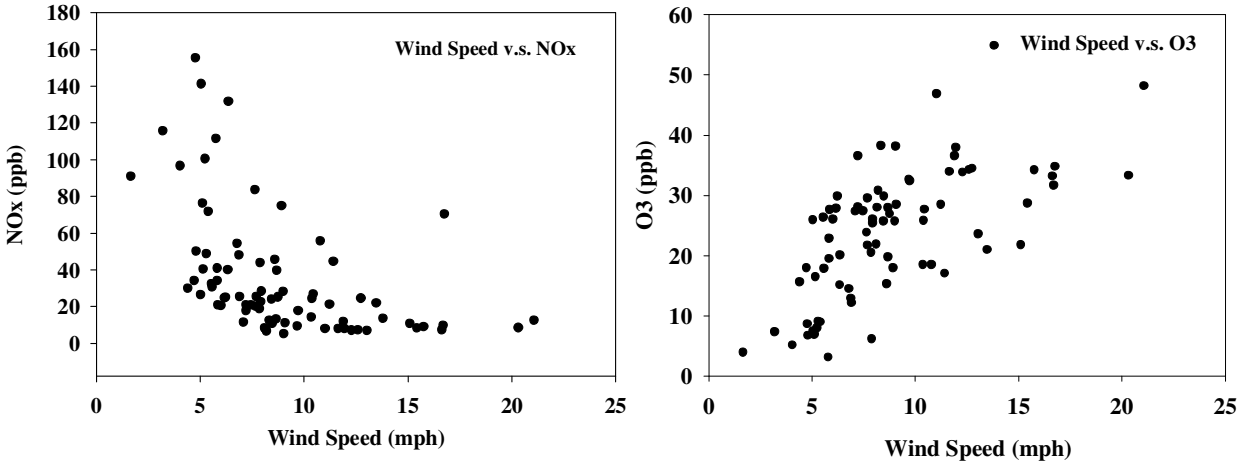


Figure 2. The correlations between wind speed and NOx (left) and O_3 (right).

(3) Effects of Relative Humidity:

Relative humidity (RH) may indirectly influence the ambient levels of NOx and O_3 level. The NOx concentrations increased positively with RH, and the O_3 levels increased negatively with RH (Figure 3). This could be explained partly by the fact that relative humidity is a measure of water in the air and high RH represents high possibility of precipitation events, and in rainy days, NOx concentration is higher and O_3 concentration is lower as discussed previously.

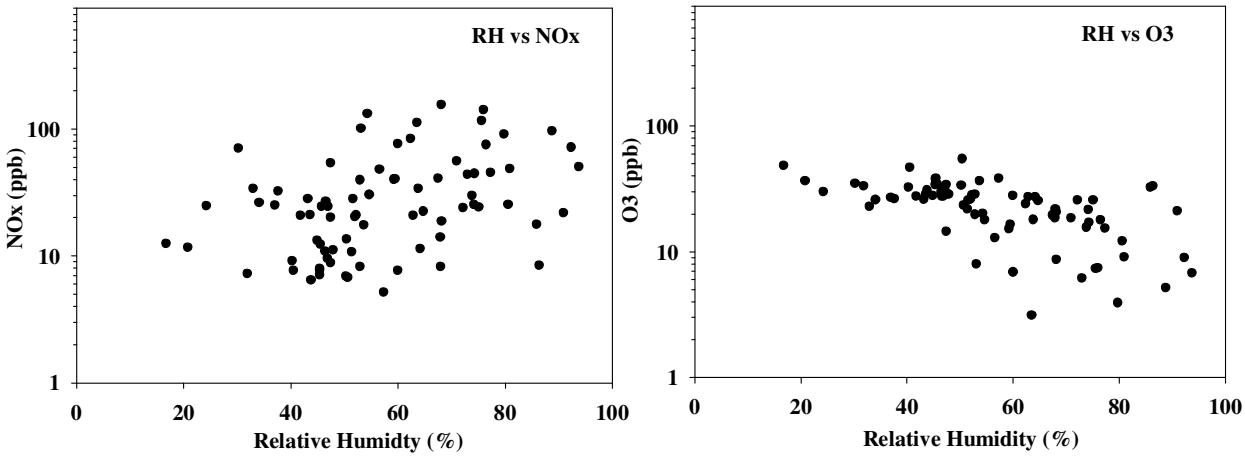


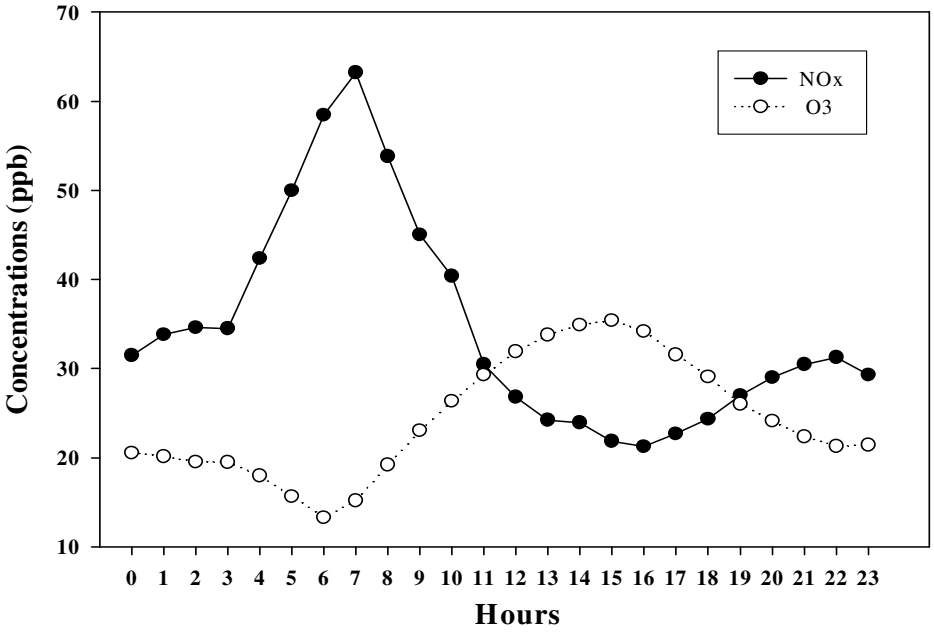
Figure 3. The correlations between Relative Humidity and NOx (left) and O_3 (right).

3. Diurnal Patterns of NOx and O_3:

During a day, the NOx was produced mainly in the morning hours, with most of NOx production occurred from 4am to 8am (Figure 4). This could be explained by a combined effect of the air boundary layer condition and the production of NOx from vehicles in the morning. In the early morning before the sunrise, the air mixing near the ground is limited due to the stable conversion layer, and thus NOx produced could be accumulated in the atmospheric boundary layer. In addition, the MERI site is very close to several highways, such as I95, Rt 3,

Rt 21 and Rt 1, and the high traffic hours in the morning could generate a tremendous mount of NOx, contributing in a great deal to the high loading of NOx in the ambient air.

On the other hand and in the contrary, O_3 was produced in the afternoon hours with peaks around 4pm. During the daytime, the photochemical reactions would dominate the inter-reactions among NOx and O_3, and such reactions would result in the production of O_3 and consumption of NOx, leading to the observed patterns that O_3 increases and NOx decrease during the day. In addition, the convection of air in daytime is high due to the thermal mixing; the NOx could be diluted greatly at the ground level, resulting in its concentration decrease. However, the difference in the O_3 concentration between lower and upper air is small, its concentration could not be affected much by convection. In the late afternoon, both photochemical reactions and convection of air mass decrease, and therefore the concentrations of NOx increase slightly with time. O_3 would react with NOx, producing certain secondary nitrogen-containing pollutants, such as N_2O_5, HNO_3 and NO_3, and thus the O_3 concentration



usually decreases with time at night.

Figure 4. Diurnal variations of averaged NOx and O_3 concentrations.

4. Estimate of Atmospheric Deposition of Nitrogen to the Meadowlands:

Nitrogen is a major nutrient that controls primary productivity in many aquatic environments, and atmospheric deposition is an important non-point source of nitrogen in these environments. Atmospheric nitrogen can be carried out by direct deposition pathways: wet deposition by precipitation and dry deposition by both aerosol particles and gases, and it can also enter a water body by indirect pathways through deposition first to the land surface and then subsequent runoff to the body of water. A large fraction of the atmospheric nitrogen input is found to be in the form of inorganic nitrogen, mainly nitrate (NO_3^-) and ammonium (NH_4^+). NO_3^- is formed through atmospheric reactions involving NO_x, and emission sources for NO_x include fossil fuel combustion from automobiles and industries, biomass burning, and other natural fixation processes. The formation of NH_4^+ is mainly by the gas-to-particle conversion reactions of NH_3 in the atmosphere, and NH_3 is primarily derived from agricultural waste, fertilizers, industrial emissions, and decomposition of organic substances.

Based on aerosol sampling at the MERI site during the spring, the atmospheric dry deposition fluxes (F_d) (mg m^{-2} $month^{-1}$) of nitrate can be calculated by the following equation:

$$F_d = (2.592 \times 10^4) * C_{air} * V_d$$

where C_{air} is the concentration of nitrate in the air (mg m^{-3}), V_d is the dry deposition velocity (cm s^{-1}), and 2.592×10^4 is a unit conversion factor. The average dry deposition velocities are assumed to be 0.34 cm s^{-1} for aerosol nitrate which was derived based on in situ measurements of aerosol particle-size distributions conducted at the New Jersey coast (Gao, 2002). The average concentration of nitrate in the ambient air over the Meadowlands is 1.81 ug/m3, and the

calculated value of nitrate dry deposition rate is 0.432 kg N $ha^{-1}year^{-1}$. Table 3 is a comparison of the atmospheric dry deposition rates of nitrate at different locations. The nitrate dry deposition rate at the MERI site is only lower than that of the Great Bay in NJ, but much higher than other sites listed; the relatively high concentration of NO_2 might be one of the possible reasons. The high dry deposition rate of nitrate at the MERI site makes it more important to study the characteristics of NOx-O_3 system as it relates to the high air-to-water flux of nutrient nitrogen, and consequently nutrient nitrogen may affect the eco-system in this region. Currently, we do not have data for wet deposition of nitrogen. The total deposition flux of nitrogen would increase if the wed deposition flux was included.

Table 3. Comparison of Nitrate Dry Deposition.

Sites	***Dry deposition Rates (kg N /ha .year)***	***References***
Meadowland	0.432	This work
Mullica River-Great Bay, NJ	0.874	Ayars & Gao, 2007
Washington Crossing, NJ	0.1	CASTNET
Wye, Maryland	0.05	AIRMoN
Hawaii	0.1	Carrillo et al, 2002.
Bridgeport and Hammonasset	0.12	Luo et al., 2002
Connecticut coastline	0.28	Luo et al., 2002

5. Relative Difference between particulate Nitrate and gas HNO_3:

The gaseous HNO_3 is the precursor of aerosol nitrate. It is mainly formed through the hydrolysis of N_2O_5, which is the product of NO_2 and NO_3, and the direct photochemical reactions between NO_2 and $HO^{\cdot}HNO_3$ could react with base species to produce aerosol nitrate and thus get the gas-phase nitrogen-containing species transferred to the aerosol phase.

HNO_3 is one of the typical acid species, which could lower greatly the pH of rain water, because of its high solubility. High level of HNO_3 would possible cause acid precipitations, which would possibly damage ecosystem and buildings. Aerosol nitrate mainly affect the ecosystem, serving as a nutrient. The excess of nitrate would cause the nitrogen eutrophication of lakes, farmlands and wetlands of nitrogen sensitive, bays and coastal areas.

Possibly an important part of the total atmospheric nitrogen deposition could be dry deposition by gas-phase nitrogen, in particular nitric acid gas. Recent studies from coastal New Jersey and North Carolina indicate that both oxidized N such as HNO_3 and reduced N such as NH_3 contribute significantly to the dry deposition of atmospheric nitrogen (CASTNET; Walker et al., 2004). Therefore, careful considerations of the contribution by nitric acid to the dry deposition, as well as wet deposition by precipitation, should be incorporated into future atmospheric N measurements for a better characterization of the total atmospheric nitrogen input to the Meadowlands aquatic eco-system.

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