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Journal of the Air & Waste Management Association Publication details, including instructions for authors and subscription information: <u>http://www.tandfonline.com/loi/uawm20</u>

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To cite this article: Nares Chuersuwan, Barbara J. Turpin & Charles Pietarinen (2000) Evaluation of Time-Resolved PM_{2.5} Data in Urban/Suburban Areas of New Jersey, Journal of the Air & Waste Management Association, 50:10, 1780-1789, DOI: <u>10.1080/10473289.2000.10464214</u>

To link to this article: <u>http://dx.doi.org/10.1080/10473289.2000.10464214</u>

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Evaluation of Time-Resolved PM_{2.5} Data in Urban/Suburban Areas of New Jersey

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ABSTRACT

Time-resolved data is needed for public notification of unhealthful air quality and to develop an understanding of atmospheric chemistry, including insights important to control strategies. In this research, continuous fine particulate matter (PM_{2.5}) mass concentrations were measured with tapered element oscillating microbalances (TEOMs) across New Jersey from July 1997 to June 1998. Data features indicating the influence of local sources and long-distance transport are examined, as well as differences between 1-hr maxima and 24-hr average concentrations that might be relevant to acute health effects. Continuous mass concentrations were not significantly different from filter-collected gravimetric mass concentrations with 95% confidence intervals during any season. Annual mean PM_{2.5} concentrations from July 1997 to June 1998 were 17.3, 16.4, 14.1, and 15.3 μ g/m³ at Newark, Elizabeth, New Brunswick, and Camden, NJ, respectively. Monthly averaged 24- and 1-hr daily

IMPLICATIONS

The promulgation of National Ambient Air Quality Standards (NAAQS) for atmospheric fine particles (PM2,) requires collection of 3 years of monitoring data to determine compliance and the development of a state implementation plan for areas found to be out of compliance. The measurements discussed here, made in advance of Federal Reference Method (FRM) monitoring, provide insights into compliance issues important in New Jersey. Specifically, compliance is in question for the annual standard, and consideration of transport and secondary aerosol formation will be important to the development of effective strategies to reduce PM25 concentrations in New Jersey. This work also illustrates temporal, spatial, and seasonal features of PM25 data that provide insights relevant to the development of effective PM25 State Implementation Plans (SIPs) elsewhere and insights into the effect of averaging time on data features.

maximum $PM_{2.5}$ concentrations suggest the existence of a high $PM_{2.5}$ (May–October) and a low $PM_{2.5}$ (November–April) season.

 $PM_{2.5}$ magnitudes and temporal trends were very similar across the state during high $PM_{2.5}$ events. In fact, the between-site coefficients of determination (R²) for daily $PM_{2.5}$ measurements were 84–98% for June and July. Additionally, during the most pronounced $PM_{2.5}$ episode, $PM_{2.5}$ concentrations closely tracked the daily maximum 1-hr O₃ concentrations. These observations suggest the importance of transport and atmospheric chemistry (i.e., secondary formation) to $PM_{2.5}$ episodes in New Jersey. The influence of local sources was observed in diurnal concentration profiles and annual average between-site differences. Urban wintertime data illustrate that high 1-hr maximum $PM_{2.5}$ concentrations can occur on low 24-hr $PM_{2.5}$ days.

INTRODUCTION

Epidemiologic studies showing statistically significant relationships between high concentrations of particulate matter and health effects1-3 have prompted the U.S. Environmental Protection Agency (EPA) to issue National Ambient Air Quality Standards (NAAQS) for airborne particles with aerodynamic diameters less than $2.5 \,\mu m \,(PM_{25})$. The NAAQS require annual average PM₂₅ concentrations to remain below 15 µg/m3 determined from a 3-year average of the annual mean concentrations, and 24-hr average PM_{2.5} concentrations to remain below 65 μ g/m³ determined from an average of the annual 98th percentile of 24-hr average concentrations for 3 successive years.⁴ While 24-hr average PM2.5 concentrations measured with Federal Reference Method (FRM) samplers must be used to determine compliance with NAAQS, continuous PM₂₅ concentrations could provide more valuable information to guide State Implementation Plan (SIP) development in nonattainment areas.

PM_{2.5} is a complex chemical mixture comprised of material emitted in particulate form (primary) and of particulate matter formed in the atmosphere (secondary) mainly through photochemical reactions and cloud processing. PM₂₅ concentrations and composition are affected by meteorology and by atmospheric chemistry that is dynamic on the time scale of minutes to hours. Diurnal variations of PM2 5 concentrations and meteorological data available from continuous measurement instruments can be used to study the contributions of primary and secondary particulate matter, as well as particulate matter generated locally and that transported from distant source regions. For example, Turpin and Huntzicker used diurnal profiles of organic and elemental carbon to estimate the contributions of primary emissions and secondary formation to organic carbon concentrations in the Los Angeles Basin.⁵ Information about diurnal, spatial, and episodic concentrations can also be used to investigate exposure patterns, and real-time automated data collection enables timely public notification in the event of "unhealthful" air quality.

In this paper, continuous PM_{2.5} concentrations and meteorological measurements are used to enhance the understanding of PM2 5 patterns in New Jersey, and to identify conditions that result in high PM_{2.5} concentrations. Data features that illustrate the contribution of local sources and long-distance transport and the effect of averaging time (i.e., 1- and 24-hr) on data features are discussed. New Jersey is the most densely populated and fifth smallest state in the United States, with roughly 1100 persons/mi² and a total area of ~7400 mi² (see Figure 1).⁶ It ranks tenth nationally in the manufacture of durable and nondurable goods. Approximately 20% of the land is classified as farmland, 35% is forest, and the rest is considered developed. New Jersey has a moderate climate with noticeable, but not unusually extreme or severe, changes in weather. It is centrally located in the northeast urban corridor extending from Washington, DC, to Boston, MA.⁷ The state borders New York to the north, Delaware to the south, the Atlantic Ocean to the east, and Pennsylvania to the west. New Jersey is out of attainment for O₃ (1-hr NAAQS), but meets the PM₁₀ standards.

METHODS

Measurement Methods

Continuous $PM_{2.5}$ mass was measured between July 1997 and June 1998 using tapered element oscillating microbalances (TEOMs). Each TEOM is equipped with a 2.5-µm cut point cyclone inlet. Ambient air is pulled through the cyclone at 16.7 L/min. The flow is then isokinetically split into a 3.0 L/min sample stream and a 13.7 L/min bypass flow. A hydrophobic Teflon-coated borosilicate glass fiber filter mounted on the narrow end of a hollow tapered



Figure 1. Sampling locations in New Jersey: Newark, Elizabeth, New Brunswick, and Camden.

tube collects particles in the 3.0 L/min sample stream at 50 °C. The narrow end of the tapered element oscillates in response to an applied electric field, and changes in oscillation frequency reflect changes in particle mass on the filter.^{8,9}

The instrument measures the oscillation frequency every 2 sec and uses the change in frequency to calculate the amount of $PM_{2.5}$ added to the filter. The TEOM stores 5-min, 1-, 8-, and 24-hr $PM_{2.5}$ averages in a computer connected to the system. The TEOM filter is held at 50 °C to minimize vapor condensation and evaporation¹⁰ and to provide a standard sampling condition. It should be noted that TEOMs can now be programmed to operate at 30 °C. However, such units were not available for this study. Losses of NH_4NO_3 , some semivolatile organic materials, and water from the collected particles are to be expected with operation at 50 °C, and these losses can result in underestimates of the $PM_{2.5}$ mass concentrations. However, high concentrations of NH_4NO_3 have not been measured in New Jersey.

Supplemental data were available from the state monitoring network and the New Brunswick Photochemical Assessment Monitoring (PAM) site. The New Jersey Department of Environmental Protection (NJDEP) provided hourly O_3 for Newark, Elizabeth, and Camden. Hourly meteorological data and O_3 were taken from a PAM site operated by NJDEP and Rutgers University and located ~0.5 mi from the New Brunswick TEOM. Table 1 lists parameters measured at the four sampling locations. Meteorological parameters include temperature, wind speed, wind direction, relative humidity (RH), and solar radiation at ground level. At the New Brunswick PAM site, temperature, wind speed, and wind direction profiles are measured from ground level to 1500 m. Daily means were calculated from 1-hr average data, monthly means from 24-hr averages, and annual means from monthly means.

For the purpose of quality control, filter-based mass measurements were collected concurrently with New Brunswick TEOM measurements. A 47-mm Teflon filter was installed in the 13.7 L/min TEOM bypass flow, and $PM_{2.5}$ was collected for 24-hr periods (1200–1200 hr EST) every third day from February 1998 to February 1999. Filter samples were collected at the ambient trailer temperature, approximately 20–25 °C. Filter handling and weighing protocols followed EPA's guidance for $PM_{2.5}$ mass measurement. Teflon filters were equilibrated at constant temperature (20–23 °C) and RH (30–40%) for at least 24 hr prior to weighing, and the weighing room temperature and RH after sample collection were within 2 °C and 5% RH of the precollection values.

A CAHN C-30 microbalance was used for filter weighing and was calibrated with a primary mass standard (200 mg) traceable to NIST mass standards at the beginning of each weighing session. The primary mass standard was reweighed at the end of each session as a validation check. Field blanks and laboratory blanks were also weighed for quality assurance purposes. Mass concentrations of PM_{2.5} were calculated by subtracting the postcollection weight from precollection weight and dividing by the volume of air that was pulled through the filter over the sampling

Table 1. Measurements with 1-hr resolution at each sampling location (July 1997–June 1998).

Parameters Measured	Newark	Elizabeth	New Brunswick	Camden
PM	\checkmark		\checkmark	
PM 10	\checkmark			\checkmark
NO	\checkmark		\checkmark	\checkmark
VOĈs			\checkmark	\checkmark
0,	\checkmark		\checkmark	\checkmark
Smoke shade	\checkmark	\checkmark		\checkmark
Temperature			\checkmark	\checkmark
Wind speed		\checkmark	\checkmark	\checkmark
Wind direction			\checkmark	\checkmark
RH			\checkmark	\checkmark
Solar radiation			\checkmark	\checkmark

period. Detection limits were taken to be 3 times the standard deviation (σ) of field blank filters.

Sampling Locations

Continuous PM_{2.5} data were collected in three urban areas (Newark, Elizabeth, and Camden) and one suburban area (New Brunswick) of New Jersey. Figure 1 shows the location of the sampling sites. The station at Newark is located in an area of dense industrial and commercial activity at Saint Charles and Berlin Streets near downtown Newark, which is the largest city in New Jersey with a population of 270,000. The site is ~0.5 mi west of the New Jersey Turnpike, a major tollway. The Elizabeth site is in an industrial area that includes petrochemical plants and refinery facilities; it is located within 0.3 mi of the New Jersey Turnpike. Elizabeth has a population of ~110,000.¹¹ The Newark and Elizabeth stations are ~11 and 8.5 mi south and southwest of New York City, respectively.

The Camden site is located ~2 mi southeast of Philadelphia, PA. The area surrounding the sampling site is mostly residential, with very few commercial activities and no industrial operations.¹² This station is expected to be influenced by the transport of $PM_{2.5}$ from greater Philadelphia. Camden has a population of ~85,000. New Brunswick is a small city (population 45,000) in central New Jersey with few industrial sources and surrounded by suburban towns. The New Brunswick site is located in the Rutgers University Gardens. All of these stations are part of the $PM_{2.5}$ monitoring network maintained by NJDEP.

RESULTS AND DISCUSSION Filter-Based and Continuous PM_{2.5} Measurements

New Brunswick gravimetric measurements and TEOM measurements collected through the same inlet and integrated over identical 24-hr time periods are shown in Figure 2. The detection limit for gravimetric measurements was ~11.4 µg (1.0 µg/m³), expressed as 3 times the standard deviation of the field blank. The coefficient of determination (\mathbb{R}^2) between the gravimetric and TEOM measurements was 95% (n = 104). The highest correlation ($\mathbb{R}^2 = 98\%$) was observed during the warmer months (May–October), as shown in Figure 2b (n = 40). During the cooler months (November–April), a somewhat lower correlation ($\mathbb{R}^2 = 87\%$) was observed. TEOM and gravimetric mass concentrations were not significantly different, with 95% confidence intervals for either period.

The use of a common sampling inlet and reasonably low NO_3 concentrations are likely explanations for the good agreement between the 24-hr integrated gravimetric and TEOM mass concentrations. Allen et al.⁹ also



Figure 2. New Brunswick gravimetric and TEOM $PM_{2.5}$ mass concentrations (μ g/m³) and 1:1 line. Data are 24-hr averages: (a) all year (n = 104), (b) May–October (n = 40), and (c) November–April (n = 40).

reported good agreement ($R^2 = 87-97\%$) between TEOM and manual methods for summertime $PM_{2.5}$ measurements in other eastern cities: Philadelphia, PA; Uniontown, PA; and Washington, DC. Larger differences were observed in Boston ($R^2 = 64\%$; annual), where the mean $PM_{2.5}$ concentrations measured by the TEOM during the winter months were 33% less than those measured gravimetrically. The good agreement between the TEOM and filter-based measurements in New Jersey suggests that the continuous $PM_{2.5}$ data reported here can be reliably used to examine issues relevant to the management of $PM_{2.5}$ concentrations in the state.

Daily and Annual Average PM_{2.5} Concentrations

Table 2 shows monthly averages of 1-hr daily maximum and 24-hr average $PM_{2.5}$ concentrations measured with a TEOM at each sampling site. Days with fewer than 21 hr of complete data were reported as missing data. Percent completeness for 24-hr averages was 98, 90, 82, and 97% at Newark, Elizabeth, New Brunswick, and Camden, respectively. Daily 1-hr maximum and 24-hr average $PM_{2.5}$ concentrations suggest the existence of a high $PM_{2.5}$ season (May–October) and a low $PM_{2.5}$ season (November– April).

Figure 3 presents 24-hr average concentrations as a time series, and they are presented as distributions for Newark and New Brunswick in Figure 4. The highest concentrations occurred during the summer months at all sites. Daily PM₂₅ concentrations exceeded the 24-hr standard (65 μ g/m³) at all sites, but only during the July 12–17, 1997, air pollution episode. The three urban sites exceed the PM_{2.5} annual standard of 15 µg/m³. Compliance, however, will be determined from a 3-year average of annual concentrations collected with FRM samplers. Newark had the highest annual average of 17.3 µg/m³, followed by 16.4, 15.3, and 14.1 μ g/m³ at Elizabeth, Camden, and New Brunswick, respectively. Analysis of hourly wind direction and PM25 concentrations over the year suggests that high PM_{2.5} concentrations occurred most frequently when winds were from the west and southwest.

Impact of Local Sources

Elevated $PM_{2.5}$ mass concentrations in Newark and Elizabeth relative to New Brunswick probably reflect the influence of local sources in these highly industrialized and populated areas. The difference is about 1–3 µg/m³ as an annual average (see Table 2). PM_{2.5} concentrations averaged on an hourly basis for each season are shown in Figure 5. Seasonally averaged autumn concentrations exhibit a pronounced peak between 0600 and 0930 hr EST at all sites. This pattern was also noticeable in spring

Table 2. Monthly averages of 1-hr daily maximum and 24-hr average $PM_{2.5}$ concentrations (μ g/m³) measured with a TEOM at each sampling site. The "high $PM_{2.5}$ season" is delineated with bold text.

Month	Newark		Elizabeth		New Brunswick		Camden	
	1-hr Max	24-hr Avg	1-hr Max	24-hr Avg	1-hr Max	24-hr Avg	1-hr Max	24-hr Avg
July 1997	42	24	30	19	36	23	36	22
August 1997	35	20	31	19	26	16	28	18
September 1997	34	19	35	20	25	15	31	19
October 1997	31	17	39	21	23	13	25	15
November 1997	26	13	28	13	16	10	22	12
December 1997	28	14	23	13	18	11	25	14
January 1998	28	15	23	14	NA	NA	20	12
February 1998	23	13	21	12	18	10	20	11
March 1998	29	16	24	15	20	12	20	13
April 1998	29	15	24	14	20	11	21	12
May 1998	36	21	31	18	26	16	29	18
June 1998	36	21	31	19	27	18	29	18
Annual Average	31.4	17.3	28.3	16.4	23.2	14.1	25.5	15.3



Figure 3. Twenty-four hour average PM_{2.5} concentrations (µg/m³) at each site: (a) summer 1997, (b) autumn 1997, (c) winter 1998, and (d) spring 1998.





Figure 4. Distributions of 24-hr PM_{2.5} concentrations at an urban site—Newark, and at a photochemical transport site—New Brunswick (July 1997–June 1998).

and winter. This morning peak illustrates the influence of local traffic. A small broad evening peak is suggested. However, it is much less pronounced, probably due to increasing mixing heights during the day.

Regional Transport

 $PM_{2.5}$ is highly correlated even between distant sites, as demonstrated in Table 3 and Figure 3. Inter-site correlations are highest during June and July for all sites ($R^2 =$ 84–98%). The lowest inter-site correlation was 50% R^2 for Elizabeth and Camden in December. The strong correlations between distant sites suggest the importance of regional transport of $PM_{2.5}$ and $PM_{2.5}$ precursors, particularly during the summer months when $PM_{2.5}$ is highest. In addition, strong correlations between sampling sites suggest that a relatively small number of sampling sites may be sufficient to quantify the distribution of $PM_{2.5}$ across the state.

From July 12 to July 17, 1997, 8-hr O_3 and 24-hr $PM_{2.5}$ standards were exceeded in an air pollution episode that yielded 1-hr maximum O_3 and $PM_{2.5}$ concentrations of 145 ppb and 112 µg/m³, respectively. The 24-hr average $PM_{2.5}$ concentration exceeded the daily standard (65 µg/m³)

at all sites. Elizabeth, Camden, and New Brunswick exceeded the standard for 1 day, and Newark exceeded for 2 days. The 1-hr average $PM_{2.5}$ concentration increased from ~15 µg/m³ to 80–112 µg/m³ at the peak of the episode. Concentrations of O₃ were quite similar across the state, as shown in Figure 6.

In contrast to the strong diurnal variations observed for O_3 , hourly $PM_{2.5}$ concentrations gradually and continuously increased, tracking the daily peak O_3 concentration as the episode developed, and decreased again as daily peak O_3 concentrations subsided, as shown in Figure 7. This pattern is consistent with the atmospheric formation of particulate matter through photochemical processes. Figure 8 shows wind back trajectories during the episode. Winds were primarily from the west and southwest during this episode. At the end of the episode, winds shifted to the north and east. An analysis of hourly winds from the New Brunswick site confirmed this pattern.

Characteristics of 1-hr Maximum PM_{2.5} Concentrations

Below, we compare 24-hr average and daily 1-hr maxima to better understand the degree to which a 24-hr standard could be expected to protect against acute exposures. Daily 1-hr maximum $PM_{2.5}$ concentration distributions are shown for Newark and New Brunswick in Figure 9. The 1-hr maximum exceeded 65 µg/m³ on only 4 days at the suburban New Brunswick site, compared with 10 days at the urban Newark site. Good correlations were seen between 1-hr maximum $PM_{2.5}$ concentrations measured at pairs of distant sites during the high PM season (May– October), as shown in Table 4. In contrast, poor correlations were observed during the low PM season (November–April).

Figure 10 shows daily 1-hr maximum PM_{2.5} concentrations in Newark and New Brunswick (July 1997-June 1998). High correlations were observed between 24-hr averages and 1-hr maxima at both locations ($R^2 = 76$ -85%). However, a reasonably large number of high 1-hr maximum days at Newark occurred when the 24-hr average concentration was not high. On 4 days, the 1-hr maximum $PM_{2.5}$ concentration was greater than 70 µg/m³, while the 24-hr average concentration was less than $30 \,\mu\text{g/m}^3$. High 1-hr maxima on low 24-hr average PM₂₅ days were not observed in New Brunswick. Closer examination of the Newark data (Figure 11) shows that days with high 1-hr maximum and low 24-hr average PM₂₅ concentrations occur during the November–April "low PM" season. During this season, a 24-hr standard cannot be expected to protect against short-term exposure to ambient PM2 5 in urban areas like Newark. It should be noted that a 24-hr average PM_{2.5} concentration of



Figure 5. One-hour average PM_{2.5} concentrations (µg/m³) in (a) summer 1997, (b) autumn 1997, (c) winter 1998, and (d) spring 1998. Data were averaged on an hourly basis for each season.



Figure 6. Concentrations of O₃ at Newark, New Brunswick, and Camden during the July 11–18, 1997, pollution episode.



Figure 7. Concentrations of O_3 (ppb) and $PM_{2.5}$ (µg/m³) during the July 11–18, 1997, pollution episode: (a) Newark—NK, (b) Camden—CD, and (c) New Brunswick—NB.

 Table 3.
 R² and number of samples (*n*) describing correlations between 24-hr

 PM_{2.5} TEOM mass concentrations at two pairs of distant sites: Newark–New Brunswick and Camden–Newark.

Month	Newark-New	Brunswick	Camden-Newark		
	R² (%)	п	R² (%)	п	
July 1997	94	24	84	31	
August 1997	96	21	83	29	
September 1997	87	28	73	30	
October 1997	78	28	78	28	
November 1997	85	26	73	23	
December 1997	65	17	71	31	
January 1998	NA	NA	58	31	
February 1998	89	26	69	28	
March 1998	76	30	75	30	
April 1998	87	29	77	30	
May 19998	80	29	73	31	
June 1998	95	29	98	30	

 $65 \ \mu g/m^3$ corresponds to a 1-hr maximum concentration of 103 and 125 $\mu g/m^3$ for New Brunswick and Newark, respectively, based on a linear least-squares fit of July 1997–June 1998 data.

CONCLUSIONS

This research describes temporal, spatial, and seasonal features of daily and 1-hr maximum PM25 data that provide insights into transport and transformation issues related to the development of an effective PM25 SIP and an understanding of the effect of averaging time on data features. Local aerosol inputs were observed in two ways. The influence of local traffic is evidenced in the 0600-0930 hr EST peak in the diurnal PM_{2.5} profiles. This feature is particularly pronounced in Elizabeth and during autumn. Additionally, annual average PM_{2.5} concentrations were $1-3 \mu g/m^3$ greater in the urban locations than in New Brunswick, a location considered descriptive of regional photochemical transport. Intersite covariance and comparisons with other pollutants were also found to be useful exploratory tools, in this case suggesting the importance of long distance transport and secondary formation to summertime PM_{2.5} concentrations. This is particularly important since PM25 concentrations were highest during the photochemical smog season, that is, May-October.

The TEOM measurements presented here also provide insights into compliance issues in advance of the FRM compliance data. The high covariance between sampling sites suggests that a relatively small number of FRM samplers are needed to characterize ambient $PM_{2.5}$ in New Jersey, and annual average $PM_{2.5}$ concentrations suggest that some areas of New Jersey will not meet the annual standard of 15 μ g/m³. Large, frequently upwind, source regions, high summertime $PM_{2.5}$ concentrations, and highly correlated $PM_{2.5}$ concentrations across the state in the summer underscore the importance of regional transport issues in the development of an effective SIP for New Jersey.

Finally, the comparison of 1-hr maxima and 24-hr average concentrations illustrates (i.e., Newark, winter)

 Table 4. R² and number of samples (n) describing correlations between 1-hr maximum PM_{2.5} TEOM mass concentrations at two pairs of distant sites: Newark–New Brunswick and Camden–Newark.

Period	Newark-New	Brunswick	Camden-Newark		
	R² (%)	п	R² (%)	п	
All year	74	315	73	363	
High PM season	82	179	80	184	
Low PM season	58	135	56	179	



Figure 8. Wind back trajectories (1000 mb) for New Brunswick, July 11–18, 1997.



Figure 9. Distributions of 1-hr maximum PM_{2.5} concentrations at an urban site—Newark, and at a photochemical transport site—New Brunswick (July 1997–June 1998).



Figure 10. Daily average and 1-hr maximum concentrations for (a) Newark—NK (n = 360; R² = 6%) and (b) New Brunswick—NB (n = 308; R² = 85%) from July 1997 to June 1998.



Figure 11. Daily average and 1-hr maximum concentrations for Newark (NK) during the (a) "high PM" season (May–October; n = 183; $R^2 = 85\%$) and (b) "low PM" season (November-April; n = 177; $R^2 =$ 62%).

that control strategies designed to attain compliance with a 24-hr standard will not necessarily protect against elevated short-duration exposures that might be associated with acute health effects.

ACKNOWLEDGMENTS

This research was supported in part by the New Jersey Department of Environmental Protection, the New Jersey Agricultural Experiment Station, and a graduate fellowship from the Royal Thai Government. The assistance of Greg Cooper and Andy Mikula with data retrieval, Terry Juchnowski with monitoring equipment, Dr. Brian Mapes with trajectory analyses, and Robert Porcja with weighing-room maintenance is gratefully acknowledged.

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