POLLUTANT TRANSFORMATIONS OVER LAKE MICHIGAN

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An aircraft, a chartered boat, and a constant altitude balloon were used to study pollutant transformations over Lake Michigan in a Lagrangian frame of reference. The experiments were conducted during the summer under strong atmospheric stability where diffusion and dry deposition of pollutants can be neglected.

Since 1976, Pacific Northwest Laboratory (PNL) has been conducting field experiments over Lake Michigan to study the rates and mechanisms of secondary pollutant formations in the Chicago and Milwaukee plumes. Emphasis is placed on the conversion of $\rm SO_2$ to sulfate. The ultimate goal of the study is to provide parameterized input for use by the Multi-State Atmospheric Power Production Pollutant Studies (MAP3S) modeling community.

During the daylight hours of summer, the surface water temperature of Lake Michigan is usually well below the ambient air temperature. The lake's cooling effect along with the drastic reduction in surface roughness stabilizes off-shore flow, inhibits turbulent mixing and produces nearly laminar flow. Under such conditions, dry deposition and diffusion become very small and can be neglected. Therefore, the changes in the concentrations of reactive species in the main body of the mixing layer should be primarily due to chemical reactions, making it far simpler to determine the rates of these reactions.

During the summer of 1978, several Lagrangian experiments were conducted using the PNL DC-3 aircraft, a chartered boat and a constant level balloon (tetroon). Measurements were made on days during which high-pressure circulation determined the transport winds. A subsidence inversion was usually associated with the synoptic high, and surface flow was generally light with a southwesterly component. Particular emphasis was given to days on which air stagnation occurred.

During the mornings, the boat was used to transport an inflated tetroon to a position downwind of either Milwaukee or

Chicago. The release was on the basis of observed and predicted winds in the mixed layer. The DC-3 later took off and rendezvoused with the boat at the selected location. When visual contact between the boat and the aircraft was achieved, a tetroon, balanced to rise to about 500 ft agl, was released from the boat. The boat then followed behind the balloon while collecting samples for SO_2 , nitrate, ammonium, and ammonia analysis. In addition, O_3 , SO_2 , NO/NO_x, light scattering, particle concentration, and aircraft location (longitude, latitude) were measured in real time and recorded on magnetic tape. The aircraft sampling route consisted of 10-mile transects, perpendicular to the tetroon trajectory and at about tetroon altitude. The DC-3 passed over the boat at the middle of each transect.

Figure 1.1 shows a typical tetroon trajectory, which was made on September 6, 1978. Each cross represents the tetroon location at the moment the aircraft crossed its path. Superimposed on the figure are the surface wind speeds and directions reported by several stations around the lake.

Data recorded on magnetic tape and samples collected on filters are being analyzed and results will be reported. The data-processing capabilities at the PNL Muskegon facility have been substantially improved over the past year. An asynchronous hardwire communications link was constructed to enable transfer of data from the aircraft system (seven-track magnetic tape) to the laboratory computer. Enhancements in both hardware and software for the laboratory minicomputer have enabled much of the necessary data reduction and analysis to be performed onsite.

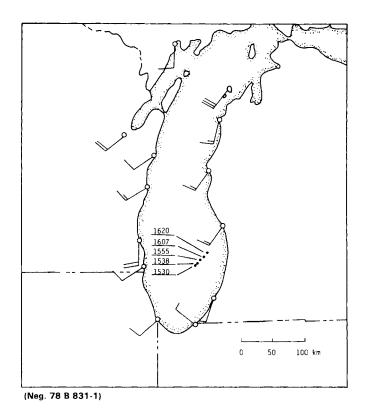


FIGURE 1.1. Lake Shore Winds and Tetroon Trajectory for September 6, 1978, 1400 EDT.

AEROSOL FORMATION IN URBAN PLUMES OVER LAKE MICHIGAN

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To determine the oxidation rates of SO_2 to sulfate in urban plumes, data from three field experiments, conducted over Lake Michigan, were analyzed and interpreted, using a photochemical smog model. A maximum rate of 4 to 6%/hr was found, and it appeared to occur around noon. Using conservative estimates for the rates of the SO_2 reactions with free radicals, the model predicted maximum rates of 4 to 5%/hr. According to the modeling results, OH and RO_2 radicals were each responsible for about 40% of the oxidation during midday, while HO_2 contributed about 20%.

During the past three years, several field experiments have been conducted over Lake Michigan. In these experiments, the oxidation rates of SO_2 to sulfate in urban plumes were studied, using instrumented aircraft and boats. A sampling program was designed to determine changes

with time in the concentrations of sulfate and several reactive species, such as SO_2 , NO_X , O_3 and non-methane hydrocarbons (NMHC). The transformation of SO_2 to sulfate has been interpreted using a chemical kinetics code, which was devised to simulate the photochemical reactions

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