TRANSPORT AND DEPOSITION OF POLLUTANTS
DOWNWIND OF ST. LOUIS

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I. Introduction

The St. Louis metropolitan area generates large numbers of pollutant materials, some of them toxic, into the atmosphere. These contaminants are carried large distances by the prevailing winds and may affect regions considerably downwind of St. Louis. The downwind decreases in the atmospheric concentrations of these contaminants with time and distance depend upon atmospheric mixing processes, wet and dry deposition on the earth's surface, and in some cases, chemical conversion processes. The relative importance of mixing processes versus removal processes in decreasing the contaminant concentrations downwind of St. Louis may be determined by comparing the concentrations of non-conservative contaminants with the concentrations of conservative contaminants. The Freons released into the atmosphere by the St. Louis metropolitan area are essentially conservative, having residence times in the atmosphere of several years. Freon-11, CCl_3F , is released into the atmosphere primarily by aerosol spray cans, while Freon-12, CCl_2F_2 , is released primarily by air conditioners. Carbon monoxide is often also considered to be essentially conservative, although it is known to react at appreciable rates with soil microflora, especially at warmer temperatures.

II. Experimental

The concentrations of trace elements, sulfates, SO_2 , CO, and Freons are being measured in air samples collected upwind, in, and downwind of St. Louis during July of 1973 and 1974 to determine the atmospheric concentrations of these contaminants produced by St. Louis and to determine the rates at which

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the concentrations are decreased downwind by atmospheric mixing processes, dry deposition on the earth's surface, and, in the case of SO_2 , chemical reactions.

In 1973 the aerosol samples for X-ray fluorescence and neutron activation analysis for trace elements were collected using HiVol pumps to draw air through five cm. diameter IPC filter papers at flow rates of about 1.7 cubic meters per minute. In 1974, eleven cm. diameter Whatman-41 filter paper was employed at a flow rate of about 0.6 cubic meters per minute. Samples were collected for SO_2 analysis by bubbling air through sodium tetrochloromercurate solutions at rates of one liter per minute. Evacuated 460 cm^3 capacity gas cylinders were used to collect whole air samples for analysis for CO and Freons in 1973. However, the results of the Freon and CO analyses were fairly erratic, probably because of the very short sampling period, so in 1974 a sampler was used which collects a whole air sample in one or two hours by slowly drawing a piston up a cylinder.

There were six days on which air samples were collected on three arcs downwind of St. Louis in 1973. The three sampling arcs were set up 20, 40, and 60 miles downwind of St. Louis on four of the six sampling days; 20, 40, and 80 miles downwind on one day; and 20, 50, and 80 miles downwind on the other day. The sampling arcs were set up by five people equipped with completely mobile sampling apparatus. On each day the first arc consisted of five sampling locations arranged more or less equidistant apart on a 30 to 50 mile line perpendicular to the predicted wind 20 miles downwind of the St. Louis arch. The length of the line was varied according to the uncertainty in the expected wind. The sampling sites were located on side roads away from towns and highways. At each of the sites an aerosol sample and an SO_2 sample were collected over a period of about one hour, and a

whole air sample was collected at the beginning of the sampling period. The samples were collected, as much as possible, at approximately the same time at each location. After collecting one set of samples at the first site, the five people moved usually 20 miles downwind where they set up a second site where they again collected a complete set of samples over a period of usually about 1.5 hours. They then again moved usually 20 miles farther downwind and collected a third set of samples over a period of usually two hours.

The spacing of the sampling arcs was chosen so that the sampling sites were moved downwind at about the same speed as the predicted wind. The elapsed time between the start of sampling at the 20-mile arc and the start of sampling at the 60-mile arc varied from five to six hours on the days the wind speed was predicted to be 5 to 10 miles. On July 27 the predicted wind was 10 miles per hour so the third arc was set up 80 miles downwind.

On each of the sampling days, at least one set of air samples was collected upwind of St. Louis to determine the contaminant concentrations in the air entering St. Louis. Sequential samples were also collected on the roof of the ten-story Federal Building in downtown St. Louis to determine the concentrations inside the city. On two days sequential samples were also collected just east of the Mississippi near Granite City. On three of the days vertical profiles were taken using the Battelle Cessna-411. Contaminant concentrations as functions of particle size are being determined from Lundgren impactor samples collected upwind of St. Louis, on the roof of the Federal Building, and at two other sites in the St. Louis area.

In 1974 the sampling procedure was changed somewhat. On each sampling day at least one set of air samples was collected upwind of St. Louis,

sequential samples were collected on the roof of the Federal Building, and two to four sequential samples were collected at stationary sites on arcs 20 to 100 miles downwind of St. Louis.

III. Results

The analysis of the 1973 samples is essentially complete, while the 1974 samples are being analyzed at the present time. This paper will discuss primarily the results of the analysis of the 1973 samples for lead, bromine, SO₂, sulfate, and Freon-11.

Lead and bromine result from automobile exhaust, while SO₂ is also produced in large quantities by fossil fuel-fired power plants. The concentrations of lead, bromine, SO₂, and sulfate were reasonably uniform along each sampling arc, and also in sequential samples collected at a given location. In 1973, however, the Freon-11, Freon-12, and CO concentrations varied widely, probably because of the very short two or three second sample collection period. Preliminary results indicate the concentrations of the Freons in the whole air samples collected over either one or two hours in 1974 were much more consistent, varying approximately as much as the other contaminants measured.

A. Lead and Bromine

The concentrations of lead and bromine averaged 20 times higher in downtown St. Louis than upwind of St. Louis. The lead and bromine concentrations at the 20-mile arc averaged only 20% and 13% of the downtown concentrations. This large decrease in concentration was probably due primarily to dilution resulting from vertical mixing through the mixing layer, since particle size analysis indicates that only a relatively small fraction would be expected to settle out in this distance, and the decrease in concentration with distance farther downwind of St. Louis was much smaller. The lead and

bromine concentrations at the outer arc (either 60 or 80 miles downwind of the arch) averaged 68% and 74%, respectively, of the concentrations at the 20-mile arc.

B. Freon-11

The Freon-11 concentrations at the 20-mile arc averaged 60% of the downtown concentrations. Freon-11 is probably released primarily in the suburbs, while lead and bromine should be produced in large amounts by downtown traffic. This was probably the primary reason why the decrease in Freon-11 concentration between downtown St. Louis and the 20-mile arc was considerably less than the decrease in the lead and bromine concentrations, although deposition of lead and bromine on the earth's surface by gravitational settling and turbulent diffusion certainly contributed to the decreases in the lead and bromine concentrations. The Freon-11 concentrations at the outer arc (60 or 80 miles) averaged 90% of concentrations at the 20-mile arc. Comparison of the decreases in lead, bromine, and Freon-11 concentrations between the 20-mile arc and the outer arc indicate that dilution due to atmospheric mixing was responsible for approximately a 10% concentration decrease, while deposition on the earth's surface caused approximately a 20% decrease in the lead and bromine concentrations.

C. Sulfur Dioxide

The SO_2 concentrations in downtown St. Louis averaged 20 times the upwind concentrations. On days with westerly winds, the SO_2 concentrations at the 20-mile arc averaged 2.5 times higher than the downtown concentrations, presumably because of the presence of large SO_2 sources along the Mississippi east of St. Louis. On days with easterly winds the SO_2 concentrations at the 20-mile arc west of the city averaged only 65% of the downtown concentrations. The SO_2 concentrations decreased with distance beyond 20 miles much more

rapidly than did the concentrations of lead, bromine, and Freon-11. The SO_2 concentration at the outer arc averaged 27% of the 20-mile arc concentration. This relatively large decrease in SO_2 concentration was probably due to the conversion of SO_2 to sulfates, and to the reaction of SO_2 with vegetation. The half-residence time for SO_2 in the atmosphere downwind of St. Louis before being deposited on the earth's surface or converted to sulfate was calculated to be 4.5 hours.

D. Sulfate

The sulfate concentrations in downtown St. Louis averaged only a few percent higher than the upwind concentrations, probably because it takes an appreciable period of time for the SO_2 produced by St. Louis to be converted to sulfate. The sulfate concentrations at the 20-mile arc averaged 185% of the downtown concentrations, and the outer arc concentrations averaged 125% of the 20-mile arc concentrations, indicating a continual conversion of St. Louis-produced SO_2 into sulfate. However, the increase in the sulfate concentration between the 20-mile arc and the outer arc was only about 10% of that necessary to account for the decrease in the SO_2 concentration, even if it is assumed that both the SO_2 and the sulfate concentrations were decreased by 30% by mixing and deposition as was the case for lead and bromine. These results indicate that SO_2 and/or sulfate were being deposited on the earth's surface at a significantly faster rate than either lead or bromine. Probably this decrease was due primarily to the reaction of SO_2 with vegetation.