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Aircraft measurements of CO_2, O_3, water vapor, aerosol fluxes and, turbulence over Lake Michigan

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RESUMEN

Con un avión dotado de instrumentos hicimos un vuelo a altura constante sobre el Lago Michigan cerca de la línea de costa y 50 km, aproximadamente, viento abajo de ella. El vuelo fue hecho en junio 18 de 1992 entre las 1:30 y las 3:30 p. m. El viento era del suroeste 12 ms^{-1} en promedio. Empleando el método de correlación torbellinaria calculamos los flujos de CO_2 y O_3, vapor de agua y aerosoles en una gama de diámetros de 0.1 a 3.0 μm. Se encontró que los flujos cerca de la costa eran significativamente más altos que en medio del lago.

Los flujos del O_3 y aerosoles cerca de la costa estaban dirigidos hacia la superficie y correspondían a las velocidades de transporte (depositación) de 0.15 cm s^{-1} y 0.85 cm s^{-1} respectivamente. En cuanto al CO_2, el vapor de agua, los flujos estaban dirigidos hacia arriba y tenían unas velocidades de transporte de 0.040 cm s^{-1} y 0.54 cm s^{-1}, respectivamente.

En medio del lago los flujos del O_3 y el vapor de agua eran hacia arriba y correspondían a una velocidad de transporte de 0.045 cm s^{-1} y 0.003 cm s^{-1}. Para el CO_2 y los aerosoles los flujos estaban dirigidos hacia abajo y correspondían a velocidades de transporte (depositación) de 0.006 cm s^{-1} y 0.226 cm s^{-1}

ABSTRACT

Using an instrumented aircraft we made a constant altitude flight over lake Michigan near the Chicago shoreline and about 50 km downwind of it. The flight was made on June 18, 1992 between about 1:30 and 3:30 pm. The wind was southwesterly averaged about 12 m s^{-1}. Employing the eddy correlation method we calculated the fluxes of CO_2, O_3, water vapor and aerosols in the diameter range of 0.1 to 3.0 μm. The fluxes near the shoreline were found to be significantly higher than those in the middle of the lake.

The fluxes near the shoreline for O_3 and aerosols were directed toward the surface and corresponding to transfer (deposition) velocities of 0.15 cm s^{-1} and 0.86 cm s^{-1}, respectively. For CO_2, and water vapor, the fluxes were directed upward and corresponding to transfer velocities of 0.04 cm s^{-1} and 0.54 cm s^{-1}, respectively.

At mid-lake the fluxes of O_3 and water vapor were directed upward and corresponding to transfer velocity of 0.045 cm s^{-1} and 0.003 cm s^{-1}. For CO_2 and aerosols the fluxes were directed downward and corresponding to transfer (deposition) velocities of 0.006 cm s^{-1} and 0.226 cm s^{-1}.

1. Introduction

From spring to the end of summer the temperature of the Great Lakes can be as much as 20°C cooler than the surrounding land (Eichenlaub, 1979). The temperature difference between the lake's surface and the air above is in the range of 5.0 to 10°C. Air advected over the cooler lake becomes very stable and dry deposition and diffusion will be insignificant. Miller *et al.* (1978) showed that a plume emitted from Oak Creek, south of Milwaukee, WI remained intact after it travelled 120 km across Lake Michigan. However, turbulent air advected over the lake will have to travel some distance from the shoreline before it becomes stable. The length of the travel depends upon prevailing meteorological condition such as air water temperature difference, and wind speed. Alkezweeny *et al.* (1977) measured vertical profiles of O_3 downwind of Milwaukee over the lake in August and found the advected air did not become stable until it travelled 20 to 50 km from the shoreline. This means that atmospheric input of pollution to the lake surface can be substantial even during spring and summer seasons.

2. Description of the measurements

The measurements were made using the a Cessna Citation Aircraft. The aircraft was instrumented to measure meteorological parameters, concentrations of aerosols and several trace gases. The aerosol concentrations were measured using the Particle Measuring System (PMS) passive cavity aerosol spectrometer probe (model PCASP-100X). The sampling time of this instrument is 0.25 s, and it covers the size range from 0.1 to 3.0 μm in diameter. The probe was mounted under the wings. The ozone concentrations were measured using the eosin-Y ozone analyzer manufactured by Scintrex. This instrument relies on a highly specific chemiluminescent reaction between the eosin-Y and ozone and has a response time of less than 1.0 s. The CO_2 and water vapor concentrations were measured using a nondispersive infrared instrument manufactured by Li-Cor. It has a response time of less than 0.1 s.

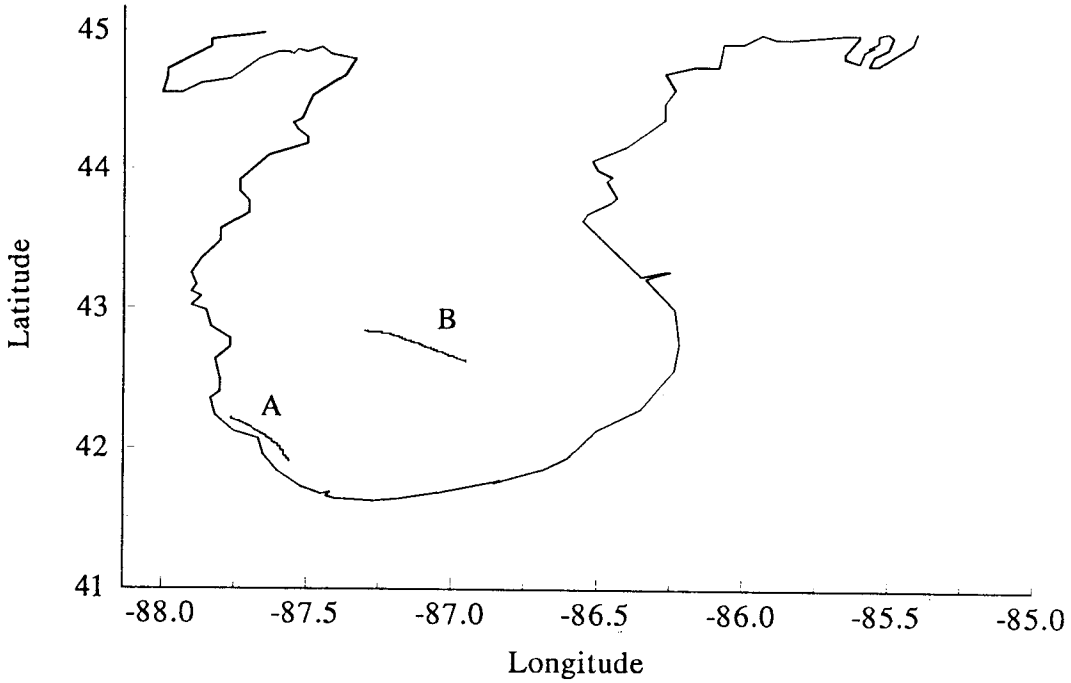

Fig. 1. The aircraft sampling route over Lake Michigan.

The horizontal and vertical winds were calculated from the data collected from a Rosemount Gust probe Model 858AJ, A Rosemount Model 102 temperature probe, and an LTN-76 Inertial Navigation System (INS). The calculation was made using the method described by Lenschow (1972).

The data were sampled at 4 Hz for the PMS probes and 24 Hz for the other data. The aircraft speed was approximately 100 m s^{-1}. The sampling altitude was maintained at about 300 m above the lake. Figure 1 shows two segments of the flight during which data were collected and analyzed. The first was downwind of Chicago about 7 to 8 km from the shoreline between 42°13'/87°46'w and 41°54'/87°34'w (pass A), the second segment was in the middle of the lake between 42°38'/86°58'w and 42°50'/87°18'w (pass B). Both passes were made across the wind. Measurement of the wind speed and direction by the instrumentation on-board the aircraft at the sampling altitudes indicated that average wind speed was about 12 m s^{-1} and occasionally gusting to 18 m s^{-1}, from the southwest.

3. Results and discussion

Figure 2a and 2b show the concentrations of O_3 and aerosols along pass A and pass B. Although the average O_3 concentration have increased from less than 39 ppb to about 53 ppb during transport, the average aerosol concentration actually decreased from 943 to 746 # cm^{-3} (Table 1). The increase in the average O_3 concentration was the result of photochemical reaction during transport. Alkezweeny *et al.* (1977) and Alkezweeny (1980) used an instrumented aircraft and boats to study the formation and transport of ozone and sulfate aerosols over Lake Michigan.

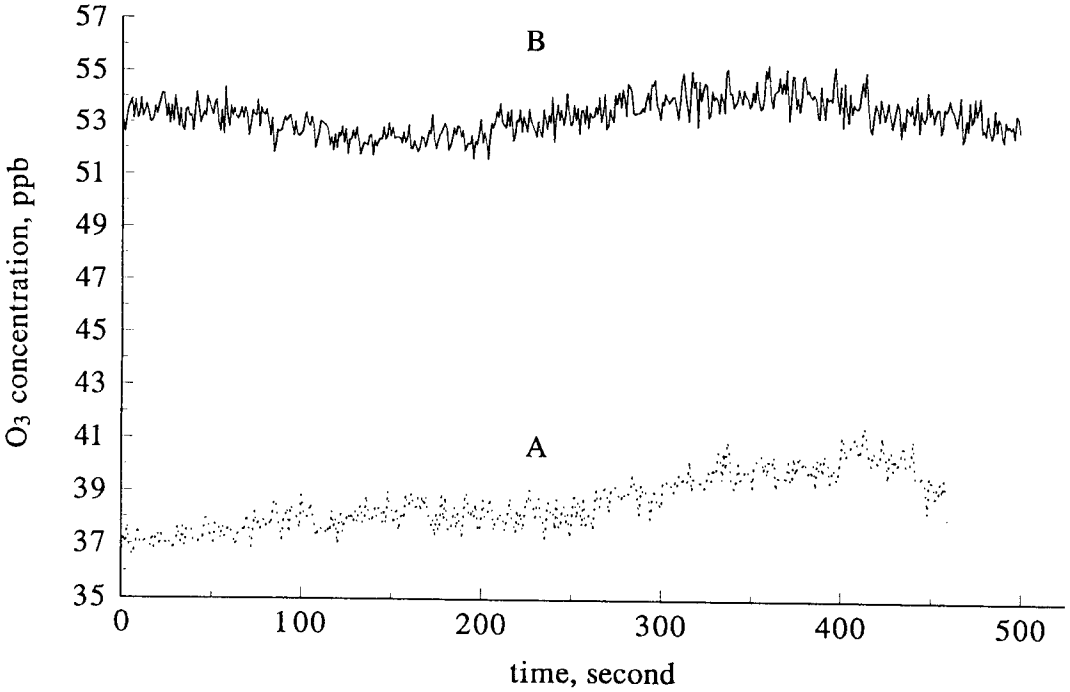

Fig. 2a. Ozone concentrations near the western Lake Michigan shore line, the dotted line, and at mid-lake, solid line. The letters refer to the sampling routes in Fig. 1.

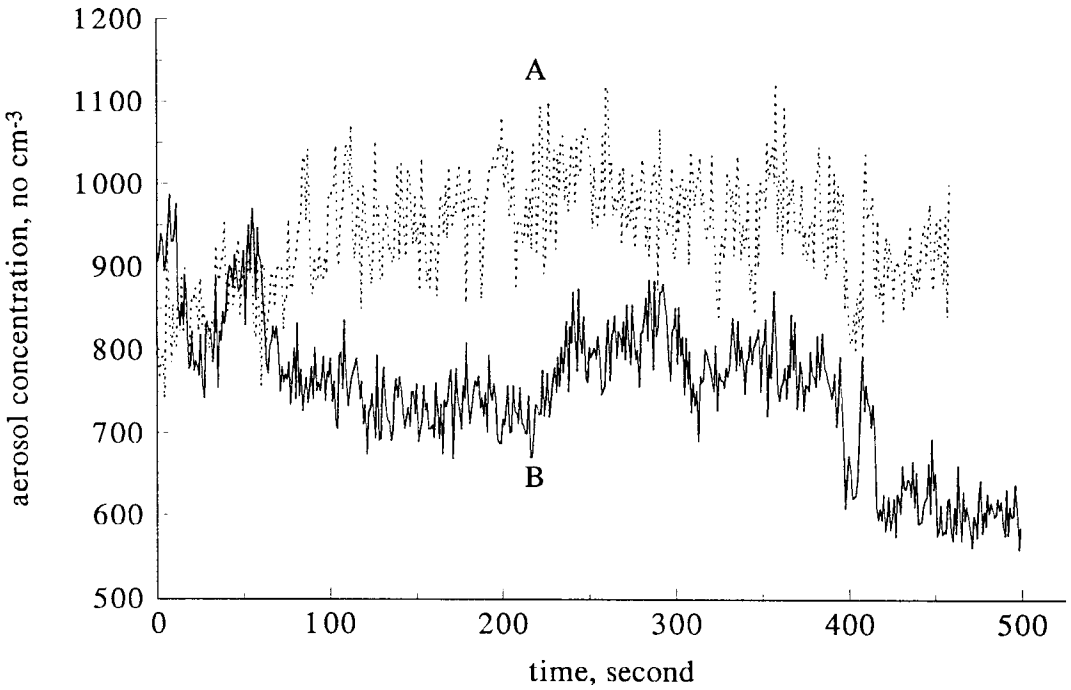

Fig. 2b. The same as Fig. 2a except for aerosol concentrations.

Table 1. Averages of parameter levels measured over lake Michigan

Parameter	Near Chicago	Mid-lake
CO_2 (ppm)	351±1	352±1
O_3(PPb)	39±1	53±1
Aerosols (# cm^{-3})	943±69	746±81
D (mean, μm)	0.21±0.01	0.23±0.01
Water vapor (gkg^{-1})	10.7±0.2	11.2±0.2
CN (# cm^{-3})	2195± 3104	643±198
Turbulence ($\epsilon^{1/3}$)	2.92±0.75	1.34±0.39
Temperature (°C)	22.4±0.2	20.3±0.1
Wind speed (ms^{-1})	12±1	13±1
Wind direction (°)	258±6	262±3

Although ozone formation was detected in all their daytime flights, no sulfate aerosol formation was found when the initial ozone concentrations were below 30 ppb. Furthermore, in that study the correlation coefficients between O_3 and aerosol concentrations for the data collected near the shoreline and mid-lake were 0.14 and 0.15 respectively. However, we have found a correlation coefficient of 0.755 for measurements taken on June 13, 1992 downwind of Chicago where the O_3 concentrations were between 65 and 100 ppb.

Since aerosol formation in urban plumes is dominated by HO reactions with SO_2 and NO_2 and ozone formation is dominated by HO_2 reaction with NO, the ratios of HO/HO_2 should differ from days with different upwind levels of reactive species and meteorological conditions. When O_3 concentrations are high HO's sinks are suppressed and thus more HO molecules become available for the oxidation of SO_2 and NO_2.

The fluxes of aerosols, water vapor, O_3, and CO_2 were calculated using the eddy correlation method. This method allows us to determine the eddy fluxes (turbulent fluxes) as described below. The eddy flux is the rate of transport of fluid properties by means of eddy in a turbulent motion; the rate of turbulent exchange. The vertical wind, W, and the pollutant concentrations, S can be represented by the following equations:

$$W = \overline{W} + W' \tag{1}$$

$$S = \overline{S} + S' \tag{2}$$

the total vertical flux is:

$$\overline{SW} = \overline{W} * \overline{S} + \overline{W'S'} \tag{3}$$

The bar denotes averages over a period of about 8 minutes and the primes denote deviations from the means. The first term in equation (3) represents the flux due to the mean flow and the second term, the covariances between fluctuations of the vertical wind velocity and S, represents the vertical turbulence flux. Figure 3 shows the variation of the covariance along pass A and pass B for O_3, CO_2, water vapor, and aerosols. It can be seen that the covariance near the shoreline is more than an order of magnitude larger than in the middle of the lake for all the gases and aerosols.

The transfer velocity, V is defined by the following equation:

$$V = \frac{\overline{W'S'}}{\overline{S}} \tag{4}$$

where the bar denotes time averages over about 8.0 minutes. A positive V values indicate emission, negative values deposition, and they are called deposition velocities.

The transfer velocities of O_3, water vapor, CO_2, and aerosols during pass A, 7.5 km from the shoreline are -0.15, 0.54, 0.038, and -0.856 cm s^{-1} respectively, and in mid-lake during pass B, are 0.045, 0.003, -0.006, and -0.226 cm s^{-1} (Table 2).

Table 2. Calculated deposition velocities (cm s^{-1})

Parameter	Near Chicago	Mid-lake
CO_2	0.038	-0.006
O_3	-0.15	0.045
Aerosols	-0.856	-0.226
Water vapor	0.54	0.003

There are not many aircraft measurements of the transfer velocities of the above species over water. Lenschow *et al.* (1982) used the eddy correlation method from aircraft, to measure the vertical fluxes of ozone over the North Pacific Ocean. They reported deposition velocities between 0.050 to 0.063 cm s^{-1}. These values are much lower than the 0.15 cm s^{-1} we measured.

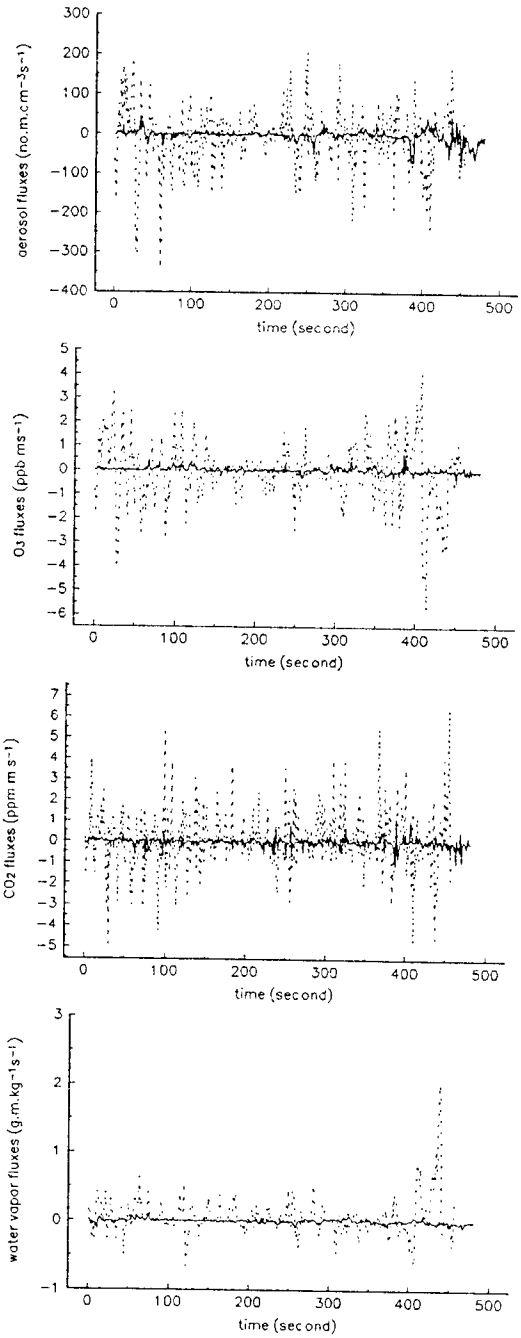

Fig. 3. Covariance between vertical velocity and O_3, aerosols, CO_2. and water vapor near the shoreline, pass A, dotted line, and at mid-lake, pass B, solid line.

Earlier aircraft measurements of Lenschow *et al.* (1981) 100 km northeast of Denver CO showed a deposition velocity of 0.47 cm s^{-1}. Wesely *et al.* (1981) measured the ozone deposition velocity over Lake Michigan using eddy correlation from a tower about 25 km southeast of Chicago. They reported values between 0.008 to 0.045 cm s^{-1}. Their values are still lower than ours. It should be noted that their measurements were made during northeasterly winds with speeds between 3.0 to 6.5 ms^{-1}. These value are much lower than the wind speeds during our measurements which were in excess of 10 ms^{-1} with a maximum of 18 ms^{-1}. Furthermore, under their northeasterly wind conditions the air traveled long distances over the cooler water and may have become more stable.

Wesely *et al.* (1982) also measured the transfer velocities of CO_2 at the Lake Michigan site they used for the ozone flux measurements. They reported a deposition velocity of (negative transfer velocity) 0.00102 cm s^{-1}. This is in contrast to our finding in which we measured an upward flux near the shoreline. Their measurements were made at wind speeds of 3 to 10 m s^{-1} which were much lower than ours. Although this value is about six times what we measured at mid-lake, at least it has the same sign. The positive value of our V is supported by results of a vertical profile of CO_2 we made on June 7, 1992 near the eastern shore of Lake Michigan across from Muskegon, MI. During the measurement a temperature inversion was present at about 700 m msl. A definite change in the CO_2 concentrations was observed across the inversion; the concentrations below were 2 to 4% higher than above. Upstill-Goddard *et al.* (1990) measured the transfer velocity of CO_2 from two small lakes in upland SW England from a boat using an SF_6 tracer. The measurement was carried out at wind speeds between about 2 to 13 ms^{-1}. Their upward transfer velocities are in the range of 0.003 to 0.007 cm s^{-1}, the higher values were associated with higher wind speeds. Our shoreline value of 0.006 cm s^{-1} obtained during the mid-lake flight is within their range. Our upward transfer velocity of 0.038 cm s^{-1} is in agreement with the results of the Heidelberg circular wind tunnel study (Jahne *et al.*, 1979).

Our aerosol deposition velocities are 0.86 cm s^{-1} near the shoreline and 0.23 cm s^{-1} at mid-lake. Wesely *et al.* (1982) used a nephelometer to measure aerosol loading and to determine the aerosol deposition velocity. This instrument measures the light scattering from aerosols in the accumulation mode, which is the same size range as our instrument. They reported a deposition velocity of 0.5 cm s^{-1} which is between our two values. Our upward transfer velocity for water vapor, 0.54 cm s^{-1} near the shoreline is in a very good agreement with wind tunnel study of Liss (1983).

We are confident that our transfer velocities during pass A, near the shoreline are representative of those near the lake's surface because of the strong turbulence we experienced during the flight. MacCready (1964) proposed turbulence scales of $1.5 < \epsilon^{1/3} < 3.5$ and $3.5 < \epsilon^{1/3} < 8.2$ $cm^{2/3}$ s^{-1} for moderate and heavy turbulence, respectively. We have recorded an average value for $\epsilon^{1/3}$ of 3.92 ± 0.88 $cm^{2/3}$ s^{-1}. This value is also about what we found during other flight we made over land at mid-day in the presence of convective activity. During the over land flight the boundary layer was very well mixed vertically as indicated by measurement of the vertical profiles of O_3 and aerosols.

Two factors influence the transfer velocity during transport, changes in the surface properties and/or turbulence intensities. Since both aircraft sampling tracks were made over water, the lake surface properties should not change. At mid-lake the averaged value of $\epsilon^{1/3}$ was 0.80 ± 0.23 $cm^{2/3}$ s^{-1}; such a value is generally found above the mixed layer. This value is also in the $\epsilon^{1/3}$ range values of 0.0 to 1.5 $cm^{2/3}$ s^{-1} which is considered as representative of light

turbulence (MacCready, 1964). If the air had stabilized the surface layer might have decoupled from the layers above the surface. In this case our computed transfer velocities would not be representative of the lake-surface values. However, the values of $\epsilon^{1/3}$ we measured on this day are still much larger than what we measured at the same location one week earlier under northwesterly wind. On that day the air had spent a much longer time over the cool water and had enough time to stabilize. Therefore, our result may still represent transfer velocities for this particular wind speed. The positive transfer velocity of ozone might have been caused by higher ozone concentration near the surface of the lake. Possible reason is the enhanced the UV light intensity because of reflection from the water surface. Furthermore, lake temperatures near the shoreline may had been much warmer than mid-lake. The cooler temperature may have influenced the transfer velocities and possibly caused the CO_2 transfer velocity to be negative (deposition).

The data in Table 1 also agree qualitatively with the transfer velocity calculations. For instance, the average mixing ratios of CO_2 and water vapor are higher at mid-lake than near the shoreline; which is consistent with the measurements of positive transfer velocities near the shoreline. The increase in the CO_2 mixing ratio only about 0.28% compared with to about 4.67% for water vapor. Our calculations show that near the shoreline the transfer velocity of water vapor is about 14.21 times that of CO_2, which is similar to the 16.68 value for the ratio of the percentage increase in the water vapor mixing ratio to the percentage increase of CO_2 between the shoreline and mid-lake. The reduction in the aerosol concentrations also indicated deposition as our calculation showed negative transfer (deposition) velocity. As expected the temperature of the air decreased by about 2.1 °C. The wind speed also increased as the air moved over smother surface and it was deflected to the right as a result of the Coriolis force.

4. Conclusion

We have used the eddy correlation method to determine the transfer velocity of gases and aerosols over lake Michigan downwind of Chicago. Our results show downward transfer velocities (deposition) of 0.15 and 0.86 cm s^{-1} for O_3 and aerosols in the size range of 0.1 to 3.0 μm in diameter and upward transfer velocities of 0.04 and 0.54 cm s^{-1} for CO_2 and water vapor about 7.5 km from the shoreline. At mid-lake much lower transfer velocities were measured. The turbulence intensity, in the subrange, was found to decrease as the air traveled over the cooler water.

Even during summertime where the air is generally stable over the lake, deposition close to the upwind side of the lake cannot be ignored. The deposition will be enhanced and may be extended deeper into the lake surface under high wind conditions.

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