Scavenging of Urban Pollutants by Thunderstorm Rainfall: Numerical Experimentation

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ABSTRACT

Rainfall collected for the squall line case of 21 August 1972 in the St. Louis area has been analyzed for certain pollutants known to be present in urban air. To aid in the interpretation of these measurements a two-dimensional time-dependent squall line model is utilized to approximate the internal air flow and rainwater distribution within the storm. At a particular time when a typical mature circulation is present, a steady state is assumed and a pollutant plume with dimensions and concentrations characteristic of the St. Louis urban plume is allowed to interact with the model thunderstorm circulation. The study focuses upon the effect of internal storm motion upon precipitation scavenging and treats microphysical processes in a relatively simple manner.

Two extreme situations are considered: 1) the pollutant does not interact with the water substance in the storm, but is merely redistributed by the storm circulation; and 2) all pollutant enters into cloudwater immediately upon entering the cloud boundary. In the second case, scavenging of pollutants by precipitation is calculated along with the deposition of pollutant on the ground in rainfall. The deposited amounts are compared with the limited number of measurements from the same storm. The trend and order of magnitude of deposition arrived at in the model compare favorably with the observations. Areas where additional observations are greatly needed are specified and desirable directions for model development are discussed.

1. Introduction

It is well known that high concentrations of pollutants are at times found in the lower troposphere in the immediate vicinity of large urban areas. Less well known is the idea that rainfall can remove pollutants from the urban atmosphere in a highly efficient manner. It is reasonable to suppose that the highest removal efficiencies are achieved by well-organized thunderstorms, whose structure is such that low-level pollutant bearing air is quickly delivered to the region of the storm where water vapor is condensing, and subsequently travels to the rain formation region. These convective storms may be contrasted in their scavenging efficiency with rain from clouds produced by large-scale forced lifting (i.e., mid-latitude cyclonic storms, often termed frontal storms) where the pollutant is either removed by falling rain below the clouds (a much less efficient process) or by rising very slowly over large horizontal distances to reach regions of condensation. Each of the latter processes would result in much smaller fluxes of pollutant in rain to the ground than in the case of squall lines, thunderstorm clusters or large isolated thunderstorms. The research described here seeks to estimate how much and where pollutant is deposited on the ground as a result of thunderstorms passing through the St. Louis urban pollutant plume on a particular day.

Field experimentation in the area of precipitation scavenging of *tracer materials* in convective storms has been carried out extensively since the first experiment in the U. S. S. R. in 1964. A review is available on this subject by Gatz (1977). A unique series of experiments was carried out in 1972 and 1973 in the St. Louis area as reported by Dana *et al.* (1974), involving the measurement of urban pollutants scavenged by individual convective storms as the storms passed through the urban plume. This type of experiment is advantageous in that it involves the very pollutants with which the concern lies and avoids the possibility of unrealistic (compared with actual plume distributions) size distributions and concentrations which may be inherent in aircraft, rocket or ground-based releases of tracer materials. The tracer releases, on the other hand, involve a known amount of material and may more easily be followed in time in order to ascertain scavenging rates and material trajectories.

Research in scavenging by convective storms utilizing dynamic cloud models is a relatively new area. Molenkamp (1974) utilized the one-dimensional time-dependent cumulus cloud model of Wisner *et al.* (1972) as a framework within which to do scavenging calculations. In that case, the mechanism considered for incorpora-

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tion of pollutant suspended in air into cloud droplets was condensation nucleation. Conversion of cloud droplets into raindrops and collection of cloud droplets by raindrops accounted for incorporation of pollutant into falling rain. Dingle and Lee (1974) attempted to explain the "direct" and "inverse" relationships often found between rainfall rate and pollutant-in-rain concentrations. They utilized a drop trajectory method (including drop breakup and evaporation) in prescribed fields of motion, rainwater and contaminate concentration in cloudwater in a cumulonimbus. One conclusion resulting from this work was that the various relationships of rainfall rate to contaminant concentration depend heavily upon the structure of the convective storm and its interactions with the environment. Molenkamp (1975) modified Murray's (1970) two-dimensional axisymmetrical cloud model to include the scavenging of radioactivity from a free air burst. This cylindrical model, though limited to cases of no ambient vertical wind shear, apparently simulates some isolated convective cells more realistically than two-dimensional models with bilateral symmetry. A conclusion derived from this work is that particles initially suspended at low levels and of favorable composition to serve as nuclei for condensation will be rapidly and efficiently scavenged.

The work to be described here is in the main directed toward elucidation of the effects of cloud internal air motions upon precipitation scavenging by well-organized squall line thunderstorms occurring on a particular day. The microphysics of the scavenging process are treated rather simply, but with sufficient realism it is hoped to study first order effects of cloud internal motions upon deposition magnitudes and locations relative to an urban plume. Scavenging calculations are carried out within a steady-state storm circulation resulting from the running of the two-dimensional time-dependent thunderstorm model of Hane (1973) in an actual past storm situation.

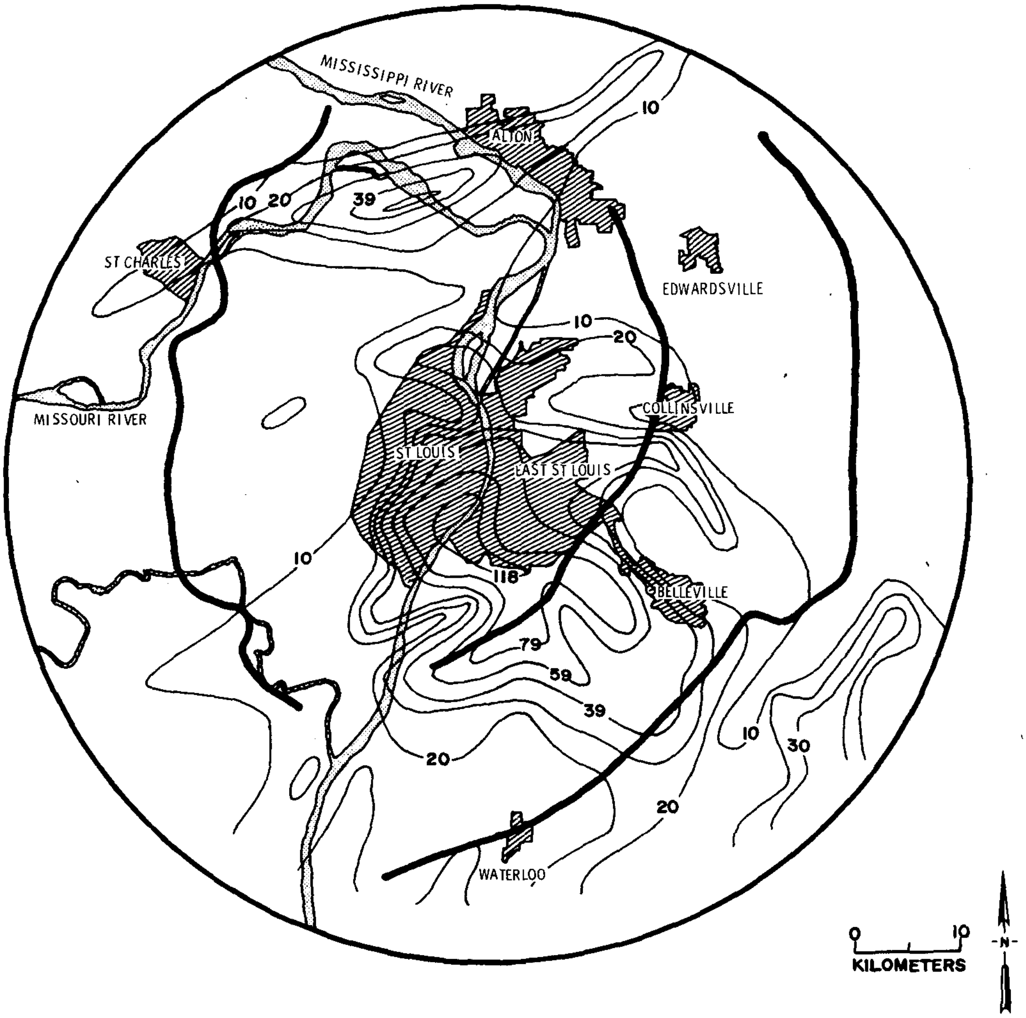

Fig. 1. Areal distribution of rainfall (mm) in the St. Louis area resulting from thunderstorm activity on 21 August 1972.

2. The St. Louis Storm of 21 August 1972

As a part of Project METROMEX (see METROMEX Participants, 1974), field experiments in precipitation scavenging of urban pollutants by convective storms were conducted in the St. Louis area during the summers of 1972 and 1973. The results of these experiments have been reported by Dana *et al.* (1974), including one particular storm on 21 August 1972 whose rainfall pattern is illustrated in Fig. 1. In these experiments, precipitation collectors were set out prior to the onset of approaching storms and retrieved immediately following the cessation of rainfall. Rainfall samples were frozen as they were collected and were later analyzed for concentrations of trace inorganics (H^+, $SO_3^=$, $SO_4^=$, NH_4^+, NO_2^- and NO_3^-). The collectors were located at 1–2 km intervals near the city along three arcs depicted by the heavy lines in Fig. 1. Some of the results of these experiments will be referred to later in this article.

The storm which occurred on 21 August possessed several squall line characteristics. Heavy rain (amounts in excess of 100 mm) along with some hail, fell in southeast portions of St. Louis and across the Mississippi River in Illinois. Individual cells moved approximately from west to east, but the storm complex or line segment of interest propagated toward the southeast (at ~6 m s^{-1}), indicating a quasi-continuous regeneration to the right of mean cell motion. A rawinsonde ascent from Lambert Field shortly before the storm occurrence indicated a moderately deep adiabatic layer near the ground with winds veering from southeasterly at 6–8 kt near the surface to westerly at 15–20 kt at about 700 mb. The temperature, moisture, and pressure data from this sounding (interpolated to 400 m height increments for model initialization) are shown in Table 1.

Table 1. Variables of state and water vapor mixing ratio as a function of height used as environmental conditions for model calculations. Based upon observations taken at Lambert Field, St. Louis, on 21 August 1972 at 2017 GMT.

Height (km)	P (mb)	T (°C)	q_v (g kg^{-1})	ρ (kg m^{-3})
0.0	990	35.2	15.542	1.1082
0.4	947	30.4	14.526	1.0775
0.8	905	26.0	13.305	1.0458
1.2	865	21.6	12.193	1.0147
1.6	826	17.5	11.503	0.9828
2.0	788	14.0	11.216	0.9492
2.4	751	11.8	8.691	0.9134
2.8	716	8.7	6.264	0.8812
3.2	682	5.7	4.429	0.8492
3.6	649	4.2	4.238	0.8130
4.0	618	2.7	3.825	0.7782
4.4	588	1.1	3.317	0.7450
4.8	559	−1.1	2.707	0.7146
5.2	532	−3.6	2.182	0.6861
5.6	505	−6.9	1.844	0.6600
6.0	480	−9.8	1.551	0.6339
6.4	455	−12.3	1.187	0.6075
6.8	432	−14.6	0.896	0.5816
7.2	409	−16.7	0.664	0.5559
7.6	388	−18.7	0.448	0.5312
8.0	368	−21.1	0.377	0.5080
8.4	348	−23.5	0.307	0.4857
8.8	329	−25.9	0.251	0.4641
9.2	312	−28.3	0.207	0.4433
9.6	295	−30.7	0.163	0.4233
10.0	278	−33.5	0.131	0.4045
10.4	263	−36.6	0.104	0.3865
10.8	248	−39.1	0.077	0.3690
11.2	234	−42.2	0.057	0.3525
11.6	220	−45.4	0.045	0.3368
12.0	207	−48.7	0.033	0.3216
12.4	195	−52.0	0.021	0.3069
12.8	183	−54.2	0.015	0.2914

3. Numerical model calculations

a. *Justification for scavenging calculations in a steady motion field*

The precipitation scavenging measurements, available for the storm described above, were found difficult to interpret with only PPI radar data to describe storm structure. Multiple Doppler radar data would have provided valuable information on internal air motions and liquid water distribution, had it been available. The next best estimate of air motions and storm structure might be provided by a numerical model which would simulate the basic structure of this storm which formed over northwest St. Louis, showed certain squall line characteristics, and which persisted with considerable intensity as it passed over and to the east of the St. Louis industrial area. In the present application, a squall line model is utilized in a time-dependent calculation, after which a steady state is assumed to approximate the internal structure of the storm.

The steady-state assumption, necessary for computational reasons, is believed well-justified on a physical basis. Computationally, inclusion of pollutant variables in the time-dependent calculations would increase computer storage requirements by approximately 50% and computer time by a like amount for a program which was using virtually all readily available space on the computer with no pollutant variables included. Physically, if the computations were to be done for a hypothetical cloud or storm, time-dependent equations for pollutant variables should be included. However, in this particular case the actual storm was not short-lived convection (such as is often simulated in numerical cloud models), but was a developed squall line which left 0.6–7.6 cm of rain (some hail) over a considerable area. The time evolution of the model storm (shown later) exhibited oscillatory behavior in terms of storm intensity, believed to be due in part to physical causes and in part to the two-dimensional restriction (which even in a squall line leads to inaccuracies in some portions of the storm). There is no justification for assuming that the details of the model *time evolution* are a good approximation to the actual storm evolution over a 1–2 h period in this case or, indeed, in the case of any numerical cloud model which might be applied in this situation, especially where virtually no data are available for specification of time varying boundary conditions. It is far more likely that the basic structure of the system would be approximated at a given time during the model time evolution. An inaccurate time evolution might well decrease the reliability of the results as compared to the steady state calculations. The difficult matter is choosing some model time when the structure is "typical" of that of the actual storm; the avenue which is taken is to choose that structure (shown later) to which the model results repeatedly return.

b. *Model application*

The squall line model of Hane (1973) is utilized to produce a storm circulation to correspond to the environmental conditions summarized at the end of Section 2. The sounding shown in Table 1 is utilized to produce a horizontally uniform temperature and moisture field for the storm environment. The wind field contains a perturbation initially produced by using slightly different horizontal wind profiles along the left and right portions of the domain. These profiles, time invariant along the boundaries, result in low-level convergence, upper level divergence and upward motion with maximum value of about 2 m s^{-1} in the lower troposphere and constrained initially to be within an 8 km wide region. These two profiles are prescribed here since only one sounding is available in this case rather than two or more soundings which might provide an estimate of the mesoscale divergence (e.g., see Hane, 1975).

The model variables are the two components of air motion in the vertical cross section along the direction of storm propagation, the temperature and the mixing ratios of water vapor, cloudwater and rainwater. The microphysical treatment of cloud and rain is based upon the Kessler (1969) parameterization formulation and excludes the ice phase. Integrations are carried out over a rectangular grid with total dimensions of 25.6 km in the horizontal and 12.8 km in the vertical with 400 m grid intervals in both the horizontal and the vertical. Important boundary conditions include no vertical motion through the upper or lower boundary and time invariant vertical distributions of horizontal motion along the boundaries. The calculations are done in a relative horizontal wind field; i.e., the motion of the storm is subtracted from the total motion field.

The model is run for approximately 1 h model time with the result that a series of developments evolve from the initial perturbation, as has been the case in past applications of the same model (see Hane, 1973, 1975). Each development is characterized by maxima in upward motion, rainwater mixing ratio and downward motion as depicted in Fig. 2, and a maximum in cloud-top height. Each development appears to contain "the seeds of its own destruction" but leaves behind conditions favorable for subsequent development. This quasi-periodic evolution as opposed to a steady-state solution is believed to result from a combination of the period of the condensation-hydrometeor production cycle and the two-dimensional bilaterally symmetric condition imposed.

4. Precipitation scavenging calculations

As noted in Section 3a, a time is chosen (at about 55 min) when a typical storm circulation is occurring. At this time the motion field, rainwater mixing ratio distribution and cloudwater distribution are assumed to be fixed in time for purposes of experimentation with pollutant inclusion into the storm. It should be noted that the steady-state assumption does not mean that

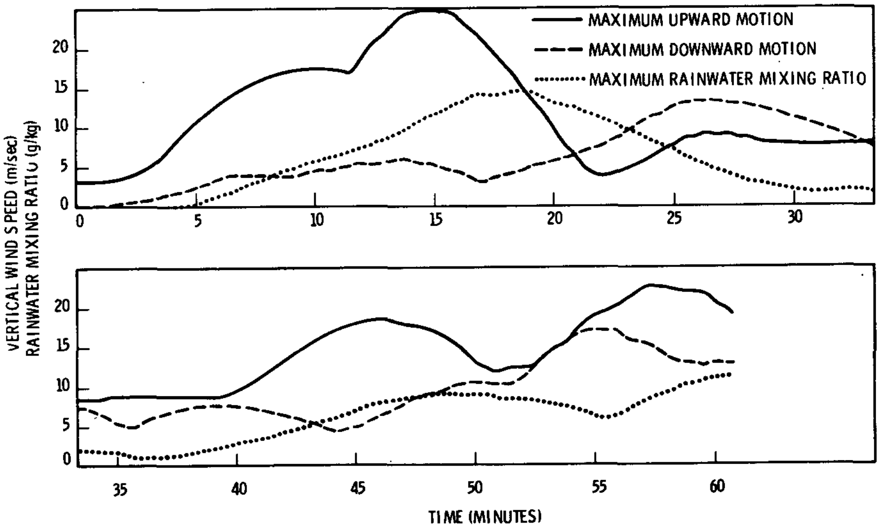

FIG. 2. The variation with time of various parameters derived from the results of model calculations for the 21 August 1972 storm.

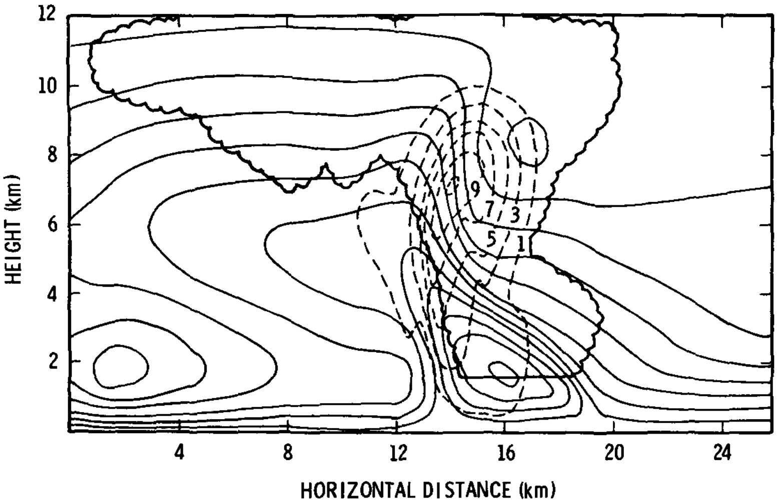

Fig. 3. The steady motion field (as depicted by solid streamfunction contours), rainwater mixing ratio (dashed, g kg^{-1}) distribution and cloud outline (scalloped) which were utilized in the precipitation scavenging calculations. Basic relative flow in low levels is from right to left.

the accretion and autoconversion processes for the water drops have been halted. It simply means that processes which tend to reduce the water concentration at a particular location are exactly balanced by processes which tend to increase the concentration at that location. The steady-state (assumed) distributions of these variables at the selected time are depicted in Fig. 3.

Two basic types of experiments are run, each for a period of 90–120 min, corresponding to two extremely different types of aerosol particles believed to be subjected to precipitation scavenging in this convective storm. In the first type, which will include cases referred to as 1, 2, 3 and 4, the assumption is made that the pollutant has no interaction with cloud or rain, but is merely redistributed by the storm air motions; these cases may be considered control experiments for the cases which follow. The type of aerosol most likely to react in this manner is insoluble and in the size range 0.1–1 μm. The other type of experiment, which will include cases referred to as A, B, C and D, includes the assumption that the pollutant immediately becomes involved in cloudwater when it crosses the cloud boundary at or near cloud base. This assumption is consistent with the existence of large soluble aerosols such as ammonium sulfate, which is apparently the major particulate pollutant in the atmosphere around St. Louis. Since this assumption is of critical importance to the outcome of Cases A–D, it is appropriate here to provide additional justification for it.

As has been pointed out by Molenkamp and Rosenkilde (1975), nucleation occurs on a single particle and since the number of particles is at least an order of magnitude larger than cloud droplet numbers, nucleation can remove only a small fraction of the *number* of particles. However, the large soluble particles are the favored aerosols for nucleation sites, and because of their size account for a large fraction of the total *mass* of that particular soluble pollutant. Furthermore, it is well known that soluble particles can increase in size by nearly an order of magnitude in subsaturated air, say in going from an environment with relative humidty of 85% to one with 100%. Fitzgerald (1975) has calculated the variation with relative humidity of the equilibrium size of ammonium sulfate in aqueous solution droplets. It would seem appropriate to perform additional calculations (not done here) to ascertain the time required to reach this equilibrium in relation to the time available for a parcel of polluted air to ascend in updraft air (well-organized storm) from near the ground to cloud base.

The nucleation assumption made here is consistent with the same assumption made by Slinn (1975) for his theoretical calculations from which he concluded that the efficiency of a convective storm in removing particles which enter the updraft is equal to the storm's efficiency to remove water. His theoretical curve compares favorably with the experimental curves of Burtsev *et al.* (1970), which show "washout" rate as a function of rainfall rate.

In cases 1–4 and A–D, a Lagrangian scheme is used to accomplish redistribution of the pollutant plume by the storm circulation. This involves 1) a first approximation to the displacement of an air parcel ending at each grid point during the previous time step by using the wind components at that grid point, 2) a recalculation of the wind components at the midpoint of that trajectory, 3) the use of these components to define a new displacement from the grid point of the location from which the parcel moved during the time step, and

TABLE 2. Concentration profiles used in model experiments.

Height (m)	Mixing ratio (10^{-8} g g^{-1}) Case 1	Case 2	Case 3	Case 4
0	12	30	0	17
400	18	27	12	17
800	27	18	14	17
1200	30	14	18	17
1600	14	12	27	17
2000	1	1	30	17
2400	0	0	0	0

4) bilinear interpolation at that location to obtain the value of the variable in question to be transported during the time step to each grid point.

5. Results of calculations

Cases 1–4 differ only by the initial vertical profile of pollutant which is located in the lower 2.4 km of the domain in a region such that the airflow will carry it to the updraft region of the storm. The plume initially occupies a 6 km horizontal extent; it is allowed to enter the boundary for a period of 45 min during the model runs. Since the grid domain is translating over the earth at the storm speed (~6 m s^{-1}) this corresponds to a plume with total horizontal extent of approximately 22 km. The initial vertical pollutant distributions in the various cases are shown in Table 2. The results of Case 1 (mid-layer maximum) are shown in Fig. 4 at four times following initiation. The results of other cases bear great similarity to Case 1 in the distribution of pollutant at these times. The most noticeable differences occur near the ground in the downdraft region. Calculations of total pollutant in the downdraft region at a particular time indicate that almost twice as much pollutant is located in this region in Case 2 (low-level maximum) as compared to Case 3 (high-level maximum). This result is simply a consequence of the fact that air near the ground entering the storm travels along a trajectory which passes through both updraft and downdraft, while air entering the storm but originating at a higher level (say, 2 km) travels along a trajectory which passes through the updraft and into the upper portion of the storm. In these cases where no precipitation scavenging is included, there is, of course, no deposition of pollutant on the ground. One might, however, speculate that in the cases where a scavenging mechanism is included, pollutant which travels through both the updraft and downdraft regions will be scavenged and deposited most effectively. This initial profile-dependence is one outcome of these first cases; the other result (which should not be surprising) is the fact that in the absence of any scavenging but with net upward motion within the grid domain, the storm redistributes the pollutant in the vertical. With the particular net vertical motion imposed in these cases, the maximum accumulation of pollutant occurs near an altitude of 8 km.

In Cases A–D, the pollutant is assumed to enter cloud droplets immediately upon entering the cloud boundary. The cloud droplets are then accreted by rain, i.e., the rate of incorporation of pollutant into rain in a given volume of air is determined by the volume swept out by the assumed distribution of raindrop sizes and the concentration of pollutant in cloud droplets in that volume. Since the accretion of cloud droplets by rain is quite efficient (as compared with the inertial capture of pollutant particles directly by rain, for instance), a collection efficiency of unity is assumed in most cases.

With a collection efficiency of unity the accretion term resulting from the assumed Marshall-Palmer distribution of raindrop sizes may be written

$$\frac{dq_r}{dt}=c\rho Q/\lambda^{3.5}, \tag{1}$$

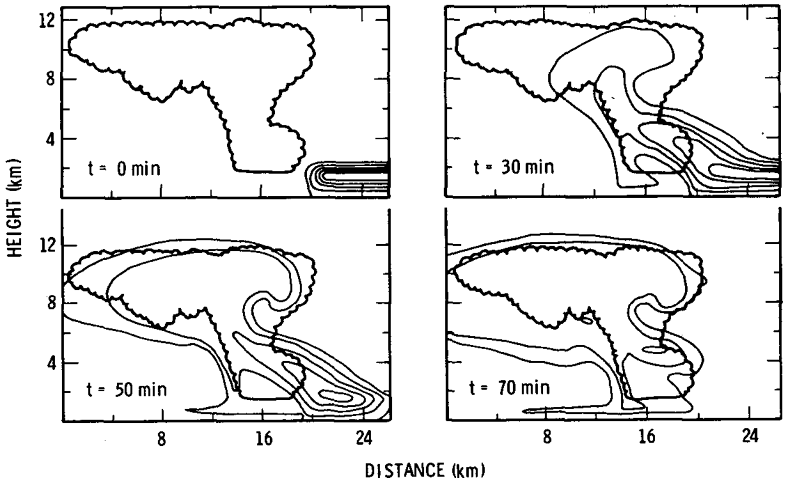

FIG. 4. The pollutant-in-air mixing ratio and cloud outline (steady state) at four times for Case 1. Contour interval is 5×10^{-8} kg kg^{-1}.

where $c=308$ is a dimensioned constant and

$$\lambda^4=251/q. \tag{2}$$

The variable q is the rainwater mixing ratio, Q the pollutant in cloudwater mixing ratio, ρ air density and q_r the pollutant in rainwater mixing ratio. The above term is calculated at the midpoint of the trajectories of pollutant-bearing rain and pollutant-bearing cloudwater in the Lagrangian scheme which is used in the time integrations for pollutant variables. This process adds pollutant to the rain at a given point and subtracts it from the air, but by slightly differing amounts, since in general the air and rain at a point arrive along different trajectories. The evaporation of rain and release of pollutant to the air is accounted for in this model whenever the trajectory of pollutant-in-rain carries it from the steady-state field of rainwater.

Table 3 summarizes Cases A–D, noting the parameters which are varied in each case. It should be noted here that Cases A and B utilize the initial pollutant profile of Case 2, Case C utilizes that of Case 3, and Case D, that of Case 4. Fig. 5 shows the results of Case D (constant initial pollutant profile) in a vertical cross section at four different times. At ten minutes the pollutant-in-air (cloudwater) has entered the updraft region and some pollutant has already entered the rainwater via the accretion of cloudwater process. The pollutant can be seen to be reaching the ground in rainfall at 30 min. The pollutant-in-rain is also spreading upward in the cloud. There are two reasons for this upward spread: 1) the pollutant is contained within drops whose terminal fallspeed is less than the upward air speed, and 2) the pollutant is entering rain at higher levels (zero contour for pollutant quantities is not drawn). The distributions at 50 and 70 min are quite similar, and one might suspect that a quasi-steady state would be achieved were it not for the cessation of pollutant influx at 45 min.

Evaporation of rain that contains pollutant results in the pollutant-in-air maximum found in low levels to the left of the rain area. Because a time is chosen when very little rain is reaching the ground, this maximum is thought to be exaggerated in these experiments. The

Table 3. Summary of cases run noting varied parameters.

Case	Collection efficiency	Type of pollutant profile
A	1.0	Low-level maximum (surface)
B	0.1	Low-level maximum
C	1.0	High-level maximum (2.0 km)
D	1.0	Constant with height

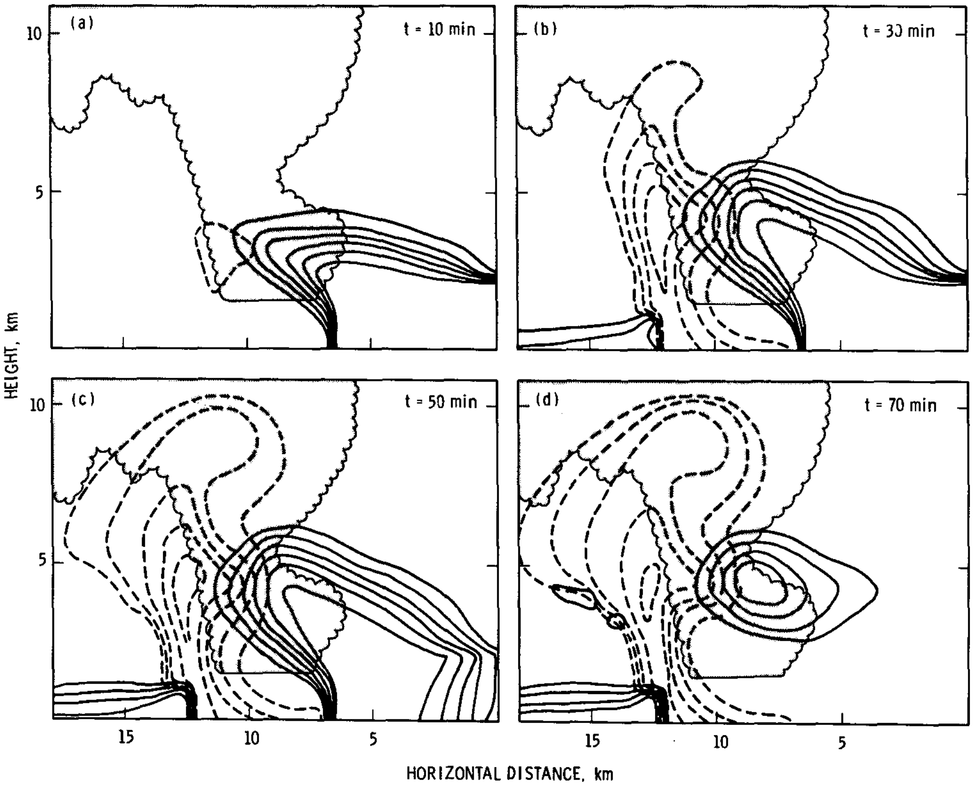

Fig. 5. Pollutant-in-air or cloud mixing ratio (solid) and pollutant-in-rain mixing ratio (dashed) distributions and cloud outline for Case D at four different times. Contour interval for both pollutant quantities is 2.5×10^{-8} kg kg^{-1} (kg of pollutant per kg of air).

pollutant-in-rain, as it approaches the ground, decelerates and travels horizontally (to the left) more than it would if there were more rainwater near the ground. This leads to a slight underestimation of the deposition of pollutant on the ground. Since evaporation is instantaneous whenever a trajectory proceeds to a region where there is no rainwater, some distortion is inevitable. However, this occurs significantly in only this one location in these runs and in this location the pollutant is advected away from the cloud.

In each of the four cases, a pollutant budget is computed during the time of calculation accounting for the following: total pollutant-in-rain (P_r) in the two-dimensional slab per meter slab thickness, total pollutant-in-air or cloud (P_a), cumulative influx minus efflux of pollutant through the lateral boundaries (P_b), and cumulative deposition on the ground of pollutant in rainfall (P_d). If P_i represents the amount of pollutant present within the domain initially, the following balance is required:

$$P_i+P_b=P_r+P_a+P_d. \tag{3}$$

An error is also computed which at each time is defined as the right-hand side of Eq. (3), minus the left-hand side. The result of these budget calculations is shown for Case C in Fig. 6. The cutoff in inflow at 45 min is seen to strongly affect the pollutant-in-air, which decreases from 45 to 75 min during which time the pollutant-in-rain reaches a maximum. The cumulative deposition of pollutant on the ground increases steadily from 15 to 75 min after which the accumulation is slower, due to a lesser amount of pollutant in the rain. Though the Lagrangian scheme used is not mass-conservative the error is thought to be numerical and due to inaccuracies in computing the flux through the lateral boundary near the ground. Compared to the total pollutant amount, it is small and certainly does not obscure the important processes. The budget calculations for the other cases show similar results.

Fig. 7 shows the deposition of pollutant-in-rain for Cases A–D as a function of downstorm distance. It is clear that the largest deposition occurs when the pollutant is concentrated near the ground, rather than concentrated at higher levels in the environmental air. The major reason for this result is simply that the trajectories through the storm followed by pollutants depend upon the location of entry into the storm from the near environment, and that certain trajectories favor scavenging more than others. A secondary reason is that the total amount of pollutant included in the model varies by a maximum of about 7% due to the fact that mixing ratios of pollutant were used as input rather than mass per unit volume, and air density decreases with height. This results in a slight increase in deposition in case where the pollutant-in-air mixing ratio maximum is near the ground, but is much less significant than the primary effect mentioned above. Case B in Fig. 7 represents the result of decreasing the collection efficiency of cloud droplets by rain, by a factor of 10 as compared with Case A. A collection efficiency (ϵ) of unity is quite realistic for large cloud droplets; however, the $\epsilon=0.1$ case was run to gain an estimate of the magnitude of change if the cloud were composed of much smaller droplets. The deposition values are reduced in Case B, but by no means by a factor of 10. The reason for the significant deposition, even in this case, is the fact that cloud droplets are accreted at a slower rate and that, there-

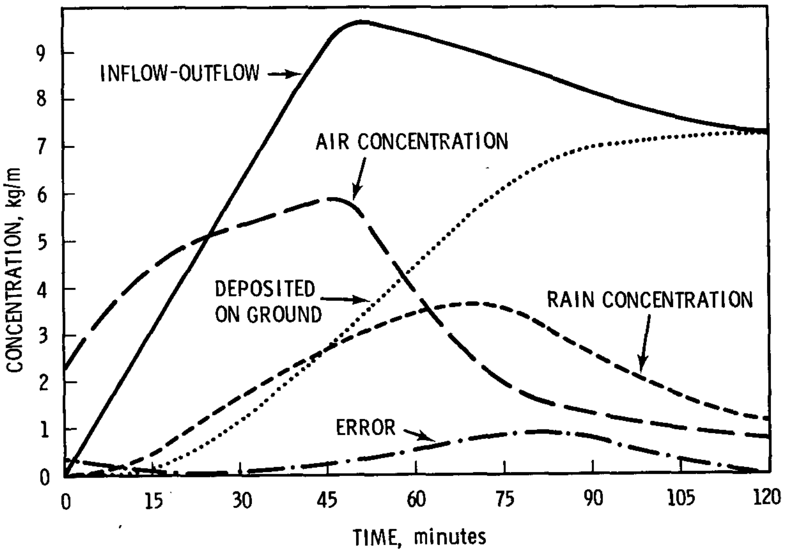

FIG. 6. Variation with time of grid-domain integrated budget parameters. The inflow-outflow curve and deposition on ground curve represent cumulative amounts, whereas other curves represent instantaneous (space-integrated) amounts.

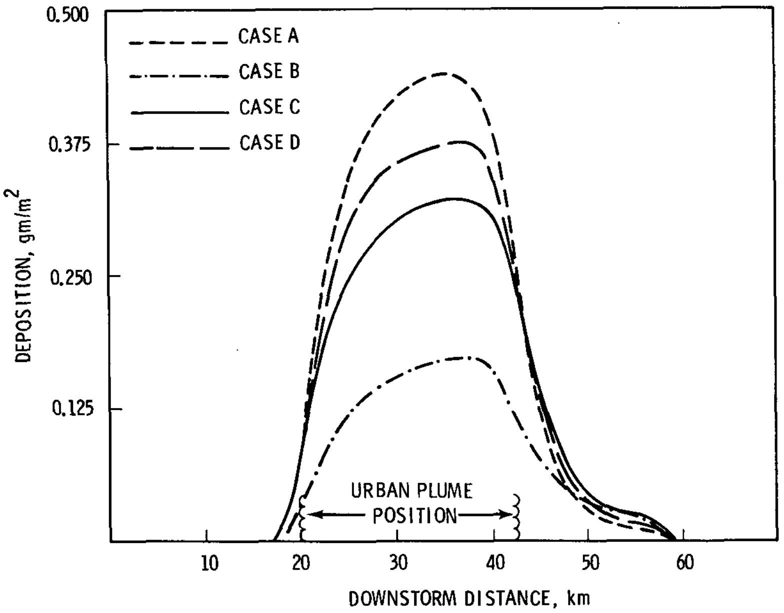

Fig. 7. Deposition of pollutant in rainfall for Cases A–D as a function of downstorm distance. The zero point on the distance scale does not refer to any particular geographical location, but corresponds to the left-hand edge of the grid domain at the initial time in relation to the urban plume position.

fore, more are available to be accreted at a later time. Indeed, this is reflected in the slight time delay (downstorm displacement of 3–4 km) of maximum deposition which may be detected in Fig. 7, in comparing Cases A and B. Fig. 7 also illustrates the location of the various deposition patterns in relation to the pre-storm location of the urban plume. The deposition pattern is skewed in the downstorm direction with a maximum 5–10 km from the center of the plume, and is spread over approximately twice the horizontal distance across the plume. This spreading results from the different horizontal air motions at different levels within the storm. If c is the storm speed and if u the mean horizontal wind speed over the pollutant trajectory within the storm, but directed oppositely to c, the $c>u$ defines the case where the deposition will occur to the right (downstorm) of the point where it entered and pollutant will have been "carried" by the storm. If $c<u$, then the pollutant will be deposited to the left (upstorm) of the point where it entered. For example, the deposition in Fig. 7 to the left of the urban plume position most probably represents pollutant from the extreme left-hand edge of the urban plume which was first to enter the condensation region as the storm approached and traveled with a speed within the storm in a direction opposite to and slightly greater than the storm speed. The slight shift of the location of maximum deposition in the downstorm direction with increasing height of the maximum environmental pollutant concentration (from A to D to C) is very likely due to this effect, since the in-storm wind shear would tend to be in the same sense as the environmental wind shear. In general, it might be said that in storms where $c>u$ at all levels, all deposition will occur within and downstorm of the urban plume location, and that the extent to which c exceeds u, determines the magnitude of this downstorm relocation. Also, in storms where the vertical wind shear is very large, the alongstorm horizontal extent of the deposition pattern might be expected to be correspondingly large.

6. Comparison with observations

An attempt is made to compare the deposition results of this model with deposition amounts obtained by Dana *et al.* (1974), in the field experiment on the day of the storm. This comparison may only be judged as an order of magnitude type comparison for several reasons: 1) only two downstorm distances (see arcs in Fig. 1) were included in the precipitation collection and chemical analysis during these field experiments in 1972, 2) no data are available for air concentration of pollutants on this day for input to the model, and 3) the model does not account for conversion of chemical species which might occur in the cloudy urban atmosphere (such as the sulfur dioxide to sulfate conversion). Because of 1) above, only two points on the calculated curve can be compared with observations. In order to deal with problem 2) above, air concentrations used for model input are based upon average values for East St. Louis, Ill., as measured by the U. S. Environmental Protection Agency (1972). The assumption is also made that the urban plume was centered over the Mississippi River on this day.

TABLE 4. Deposition (g m^{-2}) from 21 August 1972 storm.

Site	Distance from Lambert Field (km)	Flux for storm			Adjusted due to observed air concentration		
		$SO_4^=$	NO_3^-	NH_4^+	$SO_4^=$	NO_3^-	NH_4^+
1	27	0.43	0.16	0.023	0.068	0.20	0.37
2	45	0.044	0.040	0.0064	0.0070	0.050	0.10

Another obvious question concerns the extent to which differences between the model storm and actual storm will affect this comparison. Very limited observations are available for this storm in order to help answer this question. A favorable comparison of model output with detailed air motion observations for any storm would lend credence to the model results, but no such observations are as yet available. The speed of the storm is calculated from radar observations over a 90 min period to be approximately 6 m s^{-1}. The storm speed resulting from model computations also is computed to be approximately 6 m s^{-1}. The rainfall resulting from the model storm is considerably less than the maximum observed, perhaps due to the fact that the two-dimensional model best represents average conditions along a squall line, rather than the conditions contained within the more intense individual elements which compose the line. Whether this rainfall intensity factor is of critical importance would seem to depend upon whether the given model storm is capable of scavenging nearly all of the pollutant available or not. If so, an increase in intensity could not result in an increase in scavenging. Since the model results indicate that nearly all available pollutant is scavenged (under the assumptions used in Cases A–D), then perhaps the intensity question is not so critical in this particular type of application.

Deposition from the actual storm at two downstorm distances is listed in Table 4. The "points" are actually the average of values at 7 and 6 collection sites along cross-storm arcs located approximately 27 and 45 km, respectively, from the National Weather Service WSR-57 radar at Lambert Field. The model results are obtained for a prescribed initial pollutant-in-air concentration which averages 0.17×10^{-6} g g^{-1} for the 2.2 km layer near the surface. The cumulative deposition, as it turns out, is directly proportional to this initial air concentration in the model, for a given initial distribution of pollutant. Accordingly, the numbers in Table 4 are adjusted for comparison with the calculated curve by multiplying the values by the ratio of model initial air concentration to "observed" air concentration. These adjusted values are also shown in Table 4. As was mentioned earlier, the observed plume position relative to the arcs is unknown on this day also. The plume center is assumed to be along the Mississippi River or approximately 17.5 km southeast of Lambert Field. The sampling sites as a result of this assumption, are located 9.5 and 27.5 km from the plume center as shown in Fig. 8. This figure compares the deposited amounts at these two distances with the calculated amounts. The agreement is quite encouraging at Site 1, except for the sulfate, which is calculated to be higher than observed by a factor of 4. At Site 2,

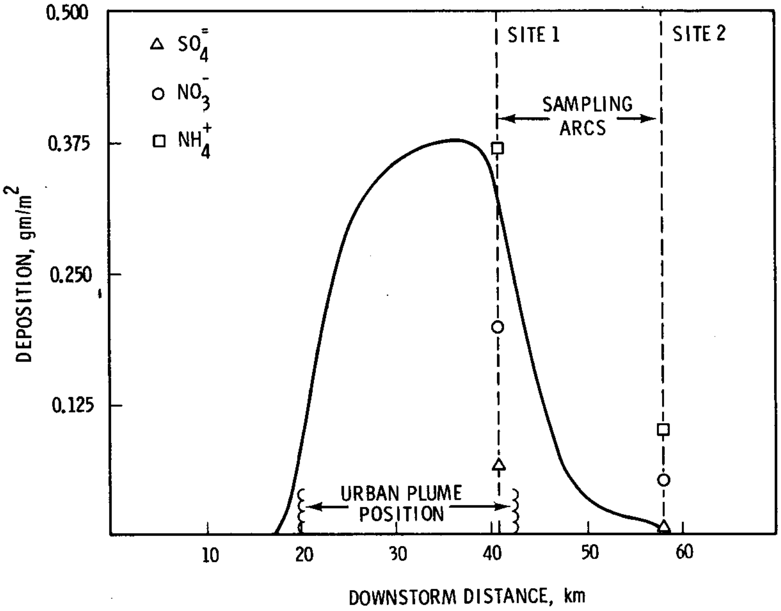

FIG 8. Comparison of deposition amounts for cross-storm averaged measurements along arcs at two downstorm distances and for model output deposition versus distance. The observed values were adjusted (see text) in order to allow for a comparison.

all observed values are slightly higher than calculated. It is quite possible that these observations at Site 2 represent deposition of "background" (regional) pollution rather than local urban-industrial pollution, and inclusion of this background pollution in the model would have resulted in better agreement. Perhaps more detailed comparisons are not justified due to the overwhelming need for additional measurements of pollutant in both rain and air; however, it might be noted that the trend and order of magnitude in calculated and observed values are the same.

7. Conclusions and recommendations

Internal air motions estimated from numerical model calculations for the 21 August 1972 storm in the St. Louis area indicated that basic storm structure allowed very efficient incorporation of urban pollutant. Given that pollutant becomes incorporated in cloudwater upon entering the condensed cloud, the model calculations indicate that a significant fraction of the pollutant is scavenged. On the average (for Cases A, C and D) approximately 80% of the pollutant entering the storm is deposited on the ground. Comparison of calculated pollutant deposition with limited observations on this day indicates that the model is predicting the amounts and downstorm locations of scavenged pollutant with reasonable accuracy and therefore that the observations may be interpreted with the aid of the air motions within and in the surroundings of the model storm. Pollutant deposition in rain is influenced by the vertical profile of pollutant in the pre-storm environment. The varying of these profiles from one case to another results in cumulative deposition variations of approximately 10–20%.

There exists an overwhelming need in connection with future precipitation scavenging field experiments to measure pollutant concentrations in the pre-storm environment. Individual pollutants measured in the rainfall should be measured in the air also. More downstorm precipitation sampling locations are needed, along with sites within the city where much of the deposition is predicted to occur. The modeling effort should be extended to include additional scavenging mechanisms applicable to all particle sizes and solubilities, and to include possible feedback into time variations in the storm dynamics. Where the object is not to simulate a particular situation, but to vary parameters and learn the effect of a particular variation upon the final result, the scavenging calculations should be done within the framework of a fully time-dependent numerical model. However, in application to particular clouds or storms, great care must be taken in order that the time evolution compare well with the actual event, lest an inappropriate time evolution lead to an erroneous interpretation of results. In some cases, a steady-state circulation might be assumed where observations indicate reasonable justification.

After conducting improved field experiments, extending model capabilities and verification of the model, a much simpler model could and should be formulated to provide 1) the areal distribution of long-term deposition of pollutants surrounding an urban-industrial area and 2) maximum short-term doses of pollutant in rainfall. This information would be of great significance to problems in areas of agriculture, water pollution, city planning and regional and global air pollution.

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