An Examination of Regional Pollutant Structure in the Lower Troposphere—Some Results of the Diagnostic Atmospheric Cross-Section Experiment (DACSE-I)[1]

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ABSTRACT

The diurnal variation of the structure of pollutant transport and diffusion in the Ohio River Valley is examined by combining meteorological cross sections with vertical profiles of several air pollutants. An increased frequency of rawinsonde releases from NWS stations at Salem, Dayton and Pittsburgh during the period 1–10 August 1977 was supplemented on 5 and 6 August by vertical profiles taken along the cross-section line by an aircraft. The data on 5 August, a day without convective activity to complicate analyses, merit special study. Analyses of the b_{scat} structure superimposd on the meteorological cross sections lend support to the theory of horizontal transport of polluted layers above the nocturnal inversion or developing mixed layer without significant dilution. The effect of the daily breathing of the planetary boundary layer on the vertical structure of pollutants can be seen in the sequence of cross sections.

1. Introduction

According to boundary-layer theory, pollutants emitted into the mixed layer during the daytime are diffused more or less uniformly. As surface cooling begins, stable layers form and vertical diffusion of pollutants within these layers essentially ceases. More importantly, the layers are released from the constraints imposed by the underlying surface, and all surface-layer turbulent exchange processes, including dry deposition, are either cut off or greatly reduced (Wesely and Hicks, 1977). In this "decoupled" state, pollutants aloft are transported downstream without appreciable dilution. If sufficient surface heating occurs on the following day, the top of the boundary layer will rise and reincorporate yesterday's pollutants into today's mixed layer. If surface heating is weaker, the maximum boundary-layer height may be below some of the polluted layers from the previous day and these layers can remain essentially intact.

An opportunity to investigate the diurnal variation of pollutant transport and diffusion arose in August 1977. In a study termed the Diagnostic Atmospheric Cross-Section Experiment (DACSE-I), a subprogram of the Multistate Atmospheric Power Production Pollution Study (MAP3S), air-quality profiles obtained by a research aircraft were combined with rawinsonde soundings. This paper discusses some of the results of DACSE-I.

2. Data acquisition and analysis

By special arrangement with the National Weather Service (NWS), rawinsonde stations at Salem, Illinois, Dayton, Ohio, and Pittsburgh, Pennsylvania, which are located along a WSW-ENE line lying north of the Ohio River (Fig. 1), each made five soundings daily from 1 to 10 August 1977; the soundings were taken at 0300, 0700, 1000, 1500 and 1900 EST. The vertical distribution of temperature, humidity, wind and pressure along the line defined by the stations was thus sampled 50 times during DACSE-I. For the special soundings at 0300, 1000 and 1500 EST, NWS rawinsonde stations provided temperature and relative humidity data at ~25 levels in the first 3.0 km. Regular soundings, for the most part, were analyzed for temperature and humidity at significant levels only (defined according to standard NWS procedures). Wind data consisted of 1 min averages, corresponding to layers approximately 300 m thick.

Additional information on the evolving structure of the atmosphere was obtained from sodar located near Manilla, Indiana. (In the following discussion, the location of Manilla is referred to by the acronym AMBIENS, which stands for the Atmospheric Mass

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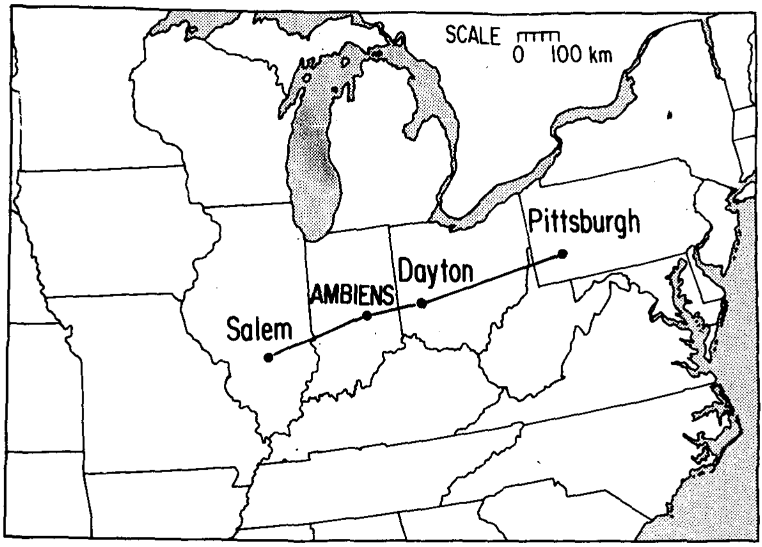


FIG. 1. Surface projection of the vertical cross-section defined by the soundings at Salem, Dayton, and Pittsburgh.

Balance of Industrially Emitted and Natural Sulfur, a field study of MAP3S that was centered near Manilla during October 1977.) Although the sodar could not determine actual temperatures, it did produce records of stable layers in the lowest kilometer of the atmosphere that were useful in determining the heights of concentrated isentropic surfaces. Specific details of the data analysis as well as the entire set of 50 atmospheric cross sections are presented elsewhere (Sisterson and Shannon, 1979).

Profiles of SO_2, O_3, b_{scat} (an indirect measure of aerosol concentration through light scattering by particles of size comparable to the wavelength of visible light), water vapor mixing ratio w and potential temperature θ were measured with the Battelle Pacific Northwest Laboratory DC-3 research aircraft along the cross-section line on 5 and 6 August 1977. Small-scale fluctuations in SO_2, O_3, and b_{scat} values have been smoothed by averaging the data over 30 s intervals, typically corresponding to a vertical interval of 8–10 mb, or about the minimum resolution of the rawinsonde soundings. Such averaging does not significantly change the heights of maxima and minima in the air-quality profiles when compared, for example, to profiles derived from data averaged over 6 s (2 mb), but the maxima and minima are somewhat reduced in amplitude. The effects of smoothing of extremes are much greater for SO_2 and O_3 concentrations than for b_{scat} values, which tend to vary more gradually. Temperature and mixing ratio profiles made by the DC-3 show little change if the averaging interval is increased from 6 to 30 s. The detection threshold and resolution of the air-quality instrumentation on board the aircraft are shown in Table 1.

TABLE 1. Detection threshold and resolution of the air-quality instrumentation on board the PNL DC-3 aircraft.

Instrument	Detection threshold	Resolution
MRI Integrating Nephelometer Model 1562	0	$\pm .05 \times 10^{-4}\ \mathrm{m}^{-1}$
Bendix Model 8002 Ozone Monitor	2 ppb	±1 ppb
Teco 14D NO_x Analyzer	10 ppb	±4 ppb
Meloy SA 285 Sulfur Analyzer	2 ppb	±2 ppb (with measured baseline or on ground) ±5 ppb (when baseline not measured as a function of altitude)

The times of the aircraft soundings often differed from the nearest times of rawinsonde releases by 1–2 h, and the DC-3 could not always descend far enough to be clearly within the rising mixed layer. The ascents and descents were also made enroute, rather than by spiral, for obvious logistical reasons. These factors complicate the determination of the mixed-layer top. Since the air above inversions and mixed layers tends to be drier than below, discontinuities in mixing ratio profiles are taken to be good indications of the tops of inversions and mixed layers. Another clue is provided by the O_3 profile; since nocturnal inversions restrict vertical mixing and since O_3 is destroyed both at the surface and in gas-phase reactions with some anthropogenic pollutants, concentrations of O_3 are often greatest at the top of a nocturnal inversion (Samson and Ragland, 1977). These qualitative indications have been used to supplement temperature measurements in determining the tops of nocturnal inversions and mixed layers.

While air chemistry is not the primary topic of discussion here, reference is made to the general behavior of NO, O_3, SO_2 and aerosol particles (as implied by b_{scat}) in an industrial plume, since such plumes are evident in the analyses. Generally, an SO_2 peak associated with a reduction of O_3 characterizes a plume near its source. SO_2 decreases downwind from the source while b_{scat} values and O_3 increase (c.f. Wilson *et al.*, 1973; Davis *et al.*, 1974; Waggoner *et al.*, 1976; White *et al.*, 1976; Wolff *et al.*, 1977; Miller *et al.*, 1978).

3. General synoptic conditions

During 1–10 August, flow around a stationary low-pressure system centered over Hudson Bay periodically brought cool Canadian air into the upper Great Lakes area. Between frontal passages an east-west ridge of high pressure, extending from the Bermuda high throughout much of the southeastern United States, brought maritime tropical air into the Ohio River Valley. When the high-pressure system was intense, surface flow along the cross section was moderate to strong from the southwest. During much of the period a quasi-stationary frontal zone lay along the southern Great Lakes. The passage of short waves aloft led to frequent convective activity along the southern edge of the frontal zone, particularly during the latter days of DACSE-I.

4. Data discussion

The data collected on 5 August are discussed below. The complete set of air quality profiles on 5 August and on 6 August, a day somewhat complicated by convective showers, appear elsewhere (Sisterson and Shannon, 1979). Maximum aircraft-measured concentrations of O_3 and SO_2 recorded on 5 August were 80 and 45 ppb, respectively, with maximum b_{scat} values of 2.5×10^{-4} m^{-1}.

A difficulty worthy of note in the following cross-section analyses is the reconciliation of the different times at which data were taken. There is some variation in the time of rawinsonde launch, with the balloons typically being released 30–45 min before the official time of the cross section and requiring about 10 min to rise above the 700 mb level, but the rawinsonde launches can be considered simultaneous. The cross-section analyses of potential temperature are thus relatively routine. Since there is only one aircraft, however, the air quality profiles are made over a range of time, during which the mixed layer may be undergoing significant development.

It is felt that transport above the mixed layer follows isentropic surfaces, as suggested in some regional dispersion models (Davis and Wendell, 1976). However, the spatial sampling limitations imposed by the single aircraft and the relatively flat isentropic surfaces make these data insufficient for conclusively demonstrating that transport in this case is isentropic rather than simply horizontal.

The earliest cross section on 5 August was at 0300 EST. Since aircraft operations began at first light, there are no coincident air quality profiles. The major features in the cross section (Fig. 2a) are the stable nocturnal boundary layer and the flat isentropic surfaces.

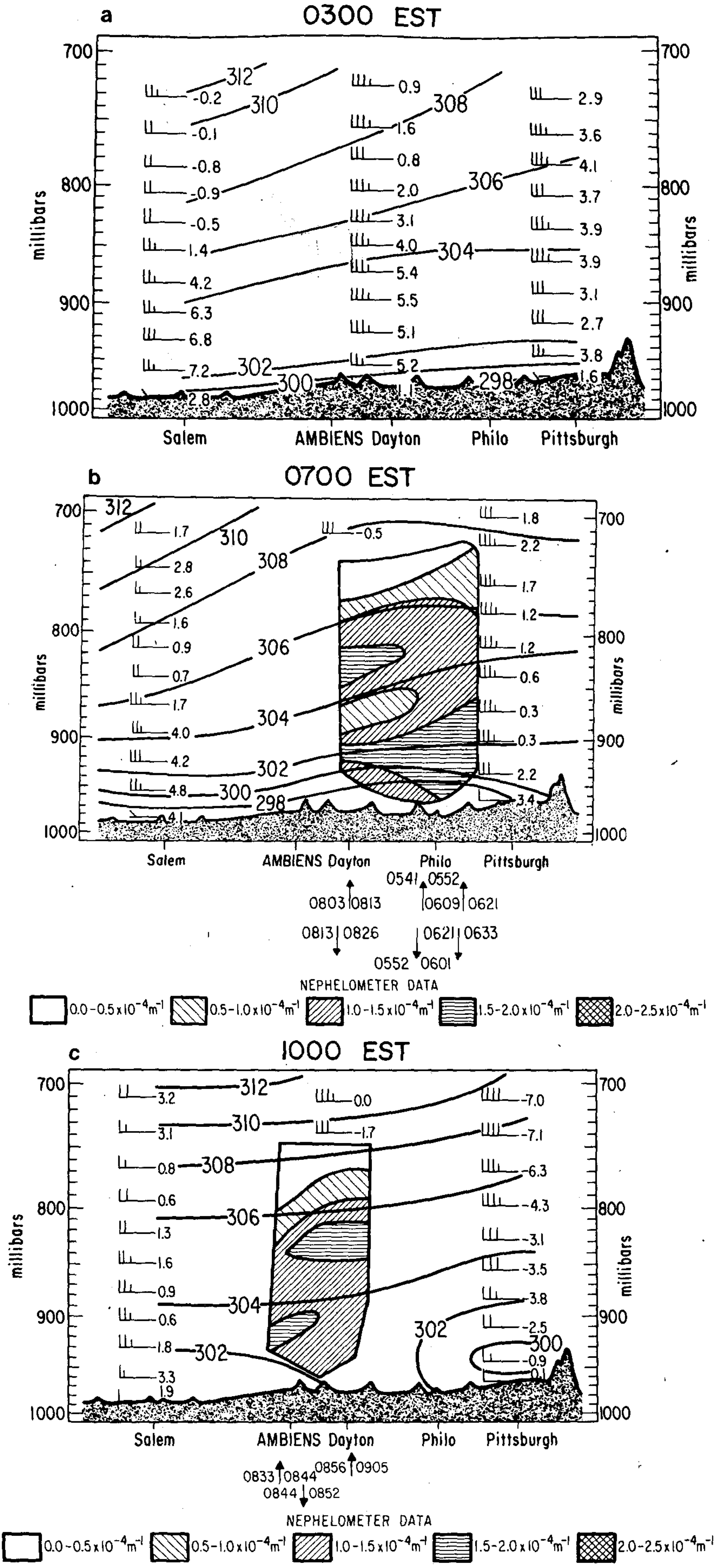
The cross-section analysis for 0700 EST 5 August is shown in Fig. 2b. The diverging feature of the lowest isentropes in the eastern half of the cross section results from the earlier onset of heating there. There are two distinct layers of high b_{scat} values. The lower layer spreads from a relatively shallow 930–910 mb over Dayton, to a thick layer extending from below the sounding base up to 860 mb west of Pittsburgh. The upper layer of particles implied by the higher b_{scat} values, between 850–810 mb near Dayton, and centered about the 305 K isentrope, may be a remnant of the mixed layer upstream on the previous day.

Two distinct layers of elevated b_{scat} values are also evident in the analysis for 1000 EST (Fig. 2c). The lower layer, between 930 and 900 mb over AMBIENS, occurs near the top of the surface inversion (935 mb). The lower layer of higher b_{scat} values has disappeared over Dayton. The upper layer of particles inferred from b_{scat}, as was observed in the previous cross section, is centered around the 305 K isentrope.

The b_{scat} analysis on the cross section for 1500 EST (Fig. 2d) is based on quite limited data (one partial ascent and one partial descent), showing monotonically decreasing vaues of b_{scat} between 860 and 780 mb. Although the analysis does not extend below 860 mb, it can be expected that the layers of particles implied by b_{scat} observations at 0700 and 1000 EST cross sections would not be present since by this time the 305 K isentrope has been incorporated into the mixed layer.

In the cross section for 1900 EST (Fig. 2e), a core of high b_{scat} values is shown between 940 and 860 mb over Dayton. Air-quality profiles for this cross section were measured over or downwind of Dayton and mark the urban pall. No distinct elevated layers of light-scattering aerosols are found, as the mixed layer probably grew to at least the height of the mixed layer of the previous day.

Ozone cross sections (not shown) did not provide evidence of regional transport. In a fresh industrial plume, NO quickly destroys O_3 and therefore oxidant concentrations may rapidly fall below off-plume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NEPHELOMETER DATA
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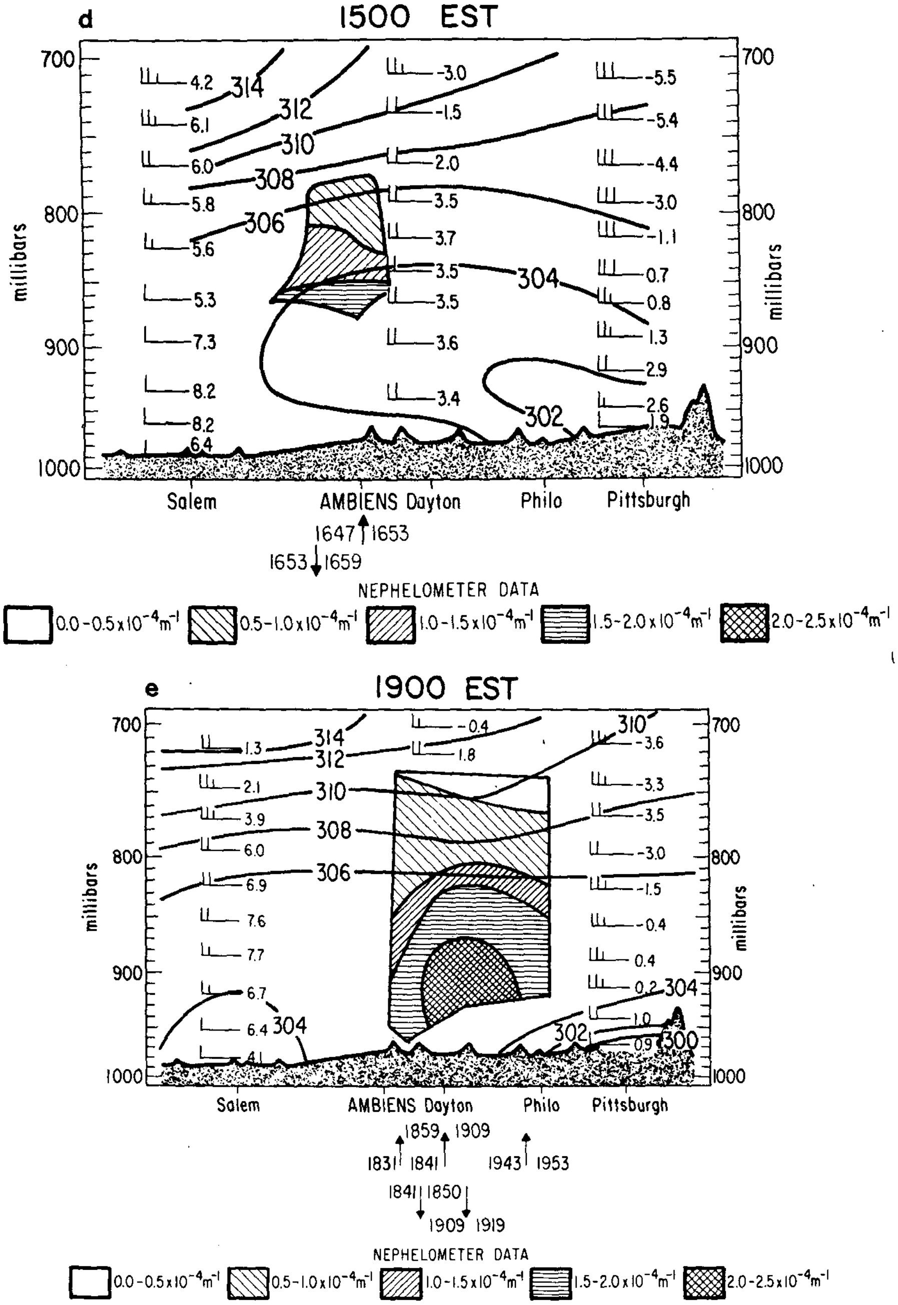

FIG. 2. Cross-section analyses of 0300–1900 EST [(a)–(e)] 5 August 1977. The horizontal wind component along the cross section line is represented by wind arrows with each full barb representing 5 m s^{-1}, each half barb representing 2.5 m s^{-1}. The horizontal wind component normal to the cross-section line is represented numerically (into the page is positive). The solid lines are potential temperature (θ). Also shown are the location of aircraft profiles along the cross section, with (↑) representing ascents, (↓) representing descents and the beginning and end times (EST) shown for each profile. The shaded terrain includes major elevations within 50 km of the cross section line.

concentrations in a thin layer (e.g., see Fig. 3). While the SO_2 and O_3 profiles are in agreement for a fresh plume, the particles represented by the b_{scat} values would be expected to increase, not decrease as shown. In a cross-section analysis based upon relatively few air-quality profiles, such local effects may mask any evidence of long-range transport of O_3. Individual pairs of air-quality profiles, however, are not inconsistent with regional O_3 transport. The profiles in Figs. 4a and 4b, taken one hour apart

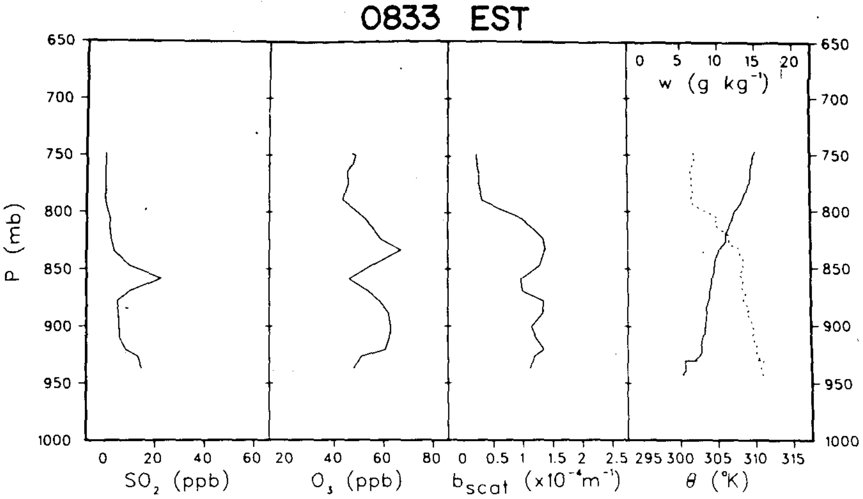

FIG. 3. Profiles of sulfur dioxide, ozone, $b_{\rm scat}$, potential temperature (solid) and mixing ratio (dashed) west of AMBIENS.

approximately in the same location, clearly show O_3 maxima at the same 305 K isentrope.

An important feature of individual profiles is the similarity of patterns of O_3 and $b_{\rm scat}$. This can be interpreted as evidence that the O_3 is not the result of stratospheric intrusion, since $b_{\rm scat}$ values are small above the tropopause (Hofmann *et al.*, 1975).

The O_3 produced during the day by photochemical reactions of pollutants can become decoupled from the surface and near-surface chemical sinks at night by the formation of a nocturnal inversion. This O_3 layer can then be transported downstream, and remain aloft until sufficient vertical mixing is reestablished to incorporate the O_3 into a new mixed layer. Relatively large O_3 concentrations above the nocturnal inversion have been observed overnight by Gloria *et al.* (1974) and for much longer periods by Samson and Ragland (1977). In the latter study, a region of reduced visibility and high O_3 concentrations was associated with a high-pressure system. Trajectory analysis inferred a long period of pollutant loading into a relatively small volume of air and movement of the air mass, essentially intact, through the Midwest. These episodes are similar to those found in the Midwest by Husar *et al.* (1976) and Wolff *et al.* (1976). The explanation of O_3 transport, particularly at night, by Samson and Ragland is similar to that outlined in this report; i.e., the thermal decoupling and increased wind speed aloft at night (Sisterson and Frenzen, 1978) could be expected to produce an efficient isentropic transport mechanism. While the frequency of large-scale episodes of decreased visibility and high O_3 concentrations has not been determined, this study suggests that long-range transport of O_3 may not be uncommon. This could explain observations of elevated O_3 concentrations in and above rural mixed layers, hundreds of kilometers downwind of urban area sources of industrially produced precursors of O_3.

Profiles of SO_2 appeared to be dominated by local sources, and thus no regional cross sections are shown. However, industrially emitted SO_2 can form sulfate aerosols. In fact, many of the light-scattering aerosols measured by nephelometry in urban atmospheres have been found to be comprised of sulfate particles (cf. Rodhe *et al.*, 1972; Charlson *et al.*, 1974; Waggoner *et al.*, 1976). Hence, while it is difficult to demonstrate SO_2 transport directly, secondary effects of SO_2, i.e., sulfate particles, may be the primary constituent of the aerosols (represented by $b_{\rm scat}$) that exhibit long-range transport.

5. Conclusion

A case study of aircraft-measured profiles of $b_{\rm scat}$ combined with sequential atmospheric cross sections in the Ohio River Valley is consistent with regional-scale transport of particles in the lower troposphere. Layers of higher $b_{\rm scat}$ values above the developing planetary boundary layer occur repeatedly, often near the same isentropic surface. Although cross-section analyses of SO_2 and O_3 are apparently dominated by local sources, individual O_3 and SO_2 profiles during the morning show stratification above the mixed layer and give indications of possible isentropic transport. Secondary products of SO_2, i.e., sulfate particles, may be a significant portion of the aerosols represented by the $b_{\rm scat}$ analysis.

Generally, the cross-section analyses demonstrate, at least in part, the "respiration" or breathing phenomenon of the diurnal evolution of the atmospheric mixed layer; i.e., pollutants are incorporated into a growing boundary layer whose turbulent structure subsides as sunset approaches. Pol-

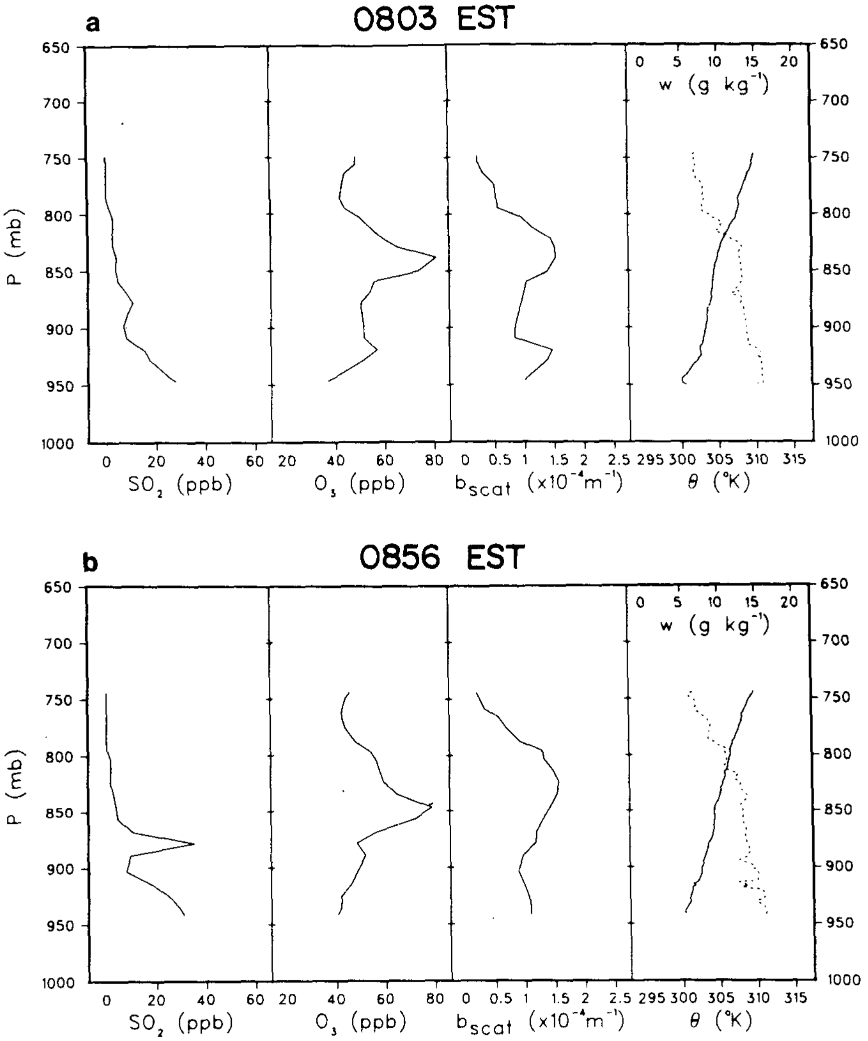

FIG. 4. Profiles of sulfur dioxide, ozone, b_{scat}, potential temperature (solid) and mixing ratio (dashed) near Dayton at 0803 (a) and 0856 (b).

lutants remaining aloft are then transported in the isentropic flow (frequently accelerated due to frictional decoupling) along stably stratified layers until the next day's "breath" establishes a new mixed layer from below.

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