

## On the Sources of Summertime Haze in the Eastern United States

**Abstract.** *The summertime haze transported from the Gulf Coast northward in maritime tropical air masses is partially formed from emissions in the midwestern and northeastern United States. Several cases are documented in which sulfate particulates, formed from emissions in the Midwest and Northeast, traveled to the Gulf of Mexico and, in some cases, returned to their source regions.*

Recent studies in the eastern United States have demonstrated the long-range transport of sulfate haze (1) and ozone (2) during the warmer half of the year. Although the Midwest and Northeast have been identified as the primary source areas of both sulfate and ozone precursors, high concentrations of sulfate and ozone have been observed upwind of these areas in maritime tropical air masses traveling northward from the Gulf of Mexico (3, 4). We studied three haze episodes associated with maritime tropical air and found that they all originated in the Northeast and Midwest, traveled in a large circular path to the Gulf of Mexico, and then returned northward. The findings are based on (i) air quality data collected from 5 August to 11 September 1979 by our mobile atmospheric research laboratory 15 km north of the Gulf Coast and 10 km south of Abbeville, Louisiana, (ii) extensive meteorological data, and (iii) satellite photographs of the eastern United States. The sampling procedures employed in this study were described previously (5).

The three episodes were easily identified by temporal traces of ozone and by the extinction of light due to scattering by dry particulates. Comparative statistics for haze episodes and nonepisodic

periods are summarized in Table 1. The first episode is not included because it occurred before the equipment was completely calibrated. The most striking features of the episodes were the approximate doubling of the daily ozone maxima and the very high concentrations of total suspended particulates (TSP). Most of the increase in TSP was attributable to increases in fine particulates, especially sulfate and ammonium. The molar ratio of 1.46 to 1.76 during the episodes suggests that the predominant sulfate species were  $\text{NH}_4\text{HSO}_4$  and  $(\text{NH}_4)_2\text{SO}_4$ , which indicate an aged aerosol (6). The threefold increase in light scattering was also primarily due to the sulfates, since the concentrations of the other species (nitrate and carbon, which occur principally in the fine particulate mass) increased only slightly during the episodes. In addition, nearly all the light extinction during the episodes was due to scattering rather than absorption by particulates. During the second episode, absorption caused only 6.4 percent of the extinction attributable to dry particles. The dominance of hygroscopic sulfate particulates in the presence of the high relative humidities (80 to 90 percent during the second episode) would further increase the importance of scattering relative to total extinction. Using published

relationships (7), we estimate that most of the scattering was caused by sulfates (7).

The shaded areas in Fig. 1, which are based on data from 230 stations, are areas that had visibilities  $\leq 10$  km (8) from 14 to 18 August 1979 (the second episode). Visibility was measured at 1300 hours Eastern Standard Time (EST) in the absence of precipitation or fog. Charts showing the 850-mbar streamlines at 0700 hours EST were used to reconstruct the movement of the haze. Before discussing Fig. 1 in detail, we would like to emphasize several points. First, the sulfate is primarily in the accumulation mode (diameter  $\leq 1 \mu\text{m}$ ) (1) and has a lifetime in the atmosphere of many days in the absence of precipitation (9). Second, since the area of the haze on 14 August was about  $4 \times 10^5 \text{ km}^2$ , dilution was negligible. In addition, the visibility in areas 200 to 300 km from the haze was generally only slightly improved. Consequently, if unreacted particulate precursors were present or if additional emissions occurred in this fringe area, the haze would have grown in area. In light of these points, it is reasonable to expect that the haze, after forming in the Northeast, would remain distinct and would expand for several days as it traveled over other regions.

On 14 August the haze was over the southern Midwest, extending into Pennsylvania ahead of the approaching cold front. It stayed ahead of the cold front during the next 24 hours, following the 850-mbar streamlines eastward and then southward. The haze arrived at our Louisiana site at 0900 hours (EST) on 16 Au-

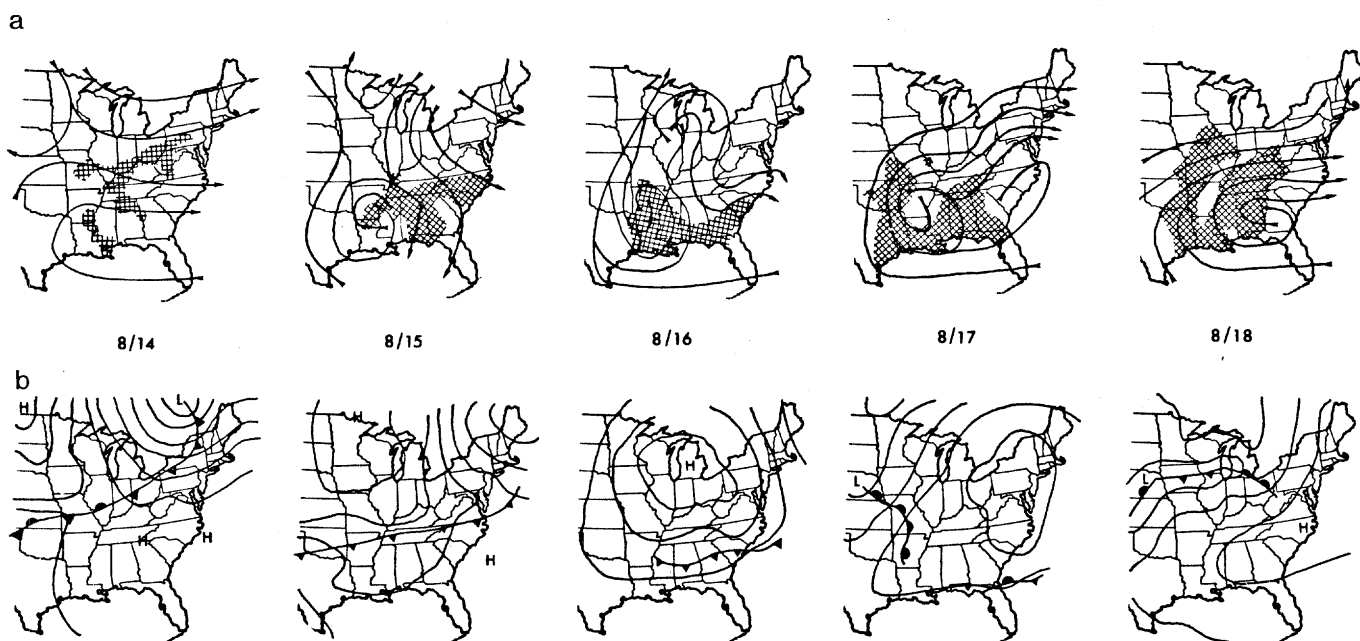


Fig. 1. (a) Progress of the second haze episode. The arrows indicate 850-mbar wind direction. (b) Surface weather charts for the same days.

gust, as evidenced by increases in extinction due to scattering (from  $0.72 \times 10^{-4}$  per meter to  $2.17 \times 10^{-4}$  per meter),  $\text{SO}_4^{2-}$  (from 5.2 to  $16.1 \mu\text{g}/\text{m}^3$ ), TSP (from 20 to  $67 \mu\text{g}/\text{m}^3$ ), and maximum hourly  $\text{O}_3$  (from 49 to 93 parts per billion). The pattern became more complicated by 16 August, but the movement of the haze still followed the 850-mbar streamlines because its top extended above the 850-mbar level and because surface flow was generally weak. Daily aircraft measurements from 16 to 18 August show that the top of the haze layer was located near 1800 m, while the mean 850-mbar level was 1570 m. The good relation between the movement of the haze and the 850-mbar wind direction was observed throughout the study. The few exceptions occurred in the Northeast behind shallow "back door" cold fronts.

By 17 August the haze was moving back toward the southern Midwest on the return flow of the surface high-pressure system, which extended above the 850-mbar level. By 18 August the haze occupied most of the southern Midwest. This flow pattern continued for the next 2 days.

Peak concentrations of particulates at our site were observed from midafternoon on 17 August through the next day. During this time the  $\text{SO}_4^{2-}$  averaged  $32.6 \mu\text{g}/\text{m}^3$ , with a 4-hour maximum of  $40.4 \mu\text{g}/\text{m}^3$ , while extinction due to scattering averaged  $3.27 \times 10^{-4}$  per meter, with a 4-hour maximum of  $3.69 \times 10^{-4}$  per meter. Three samples collected by aircraft

Table 1. Average pollutant levels near Abbeville, Louisiana, during episodic and nonepisodic conditions.

Parameter	Non-episodic	Second episode	Third episode
TSP* ( $\mu\text{g}/\text{m}^3$ )	43.1	94.7	92.6
FPM† ( $\mu\text{g}/\text{m}^3$ )	12.9	60.0	
FPM (%)	29.9	63.4	
$\text{SO}_4^{2-}$ ( $\mu\text{g}/\text{m}^3$ )	5.2	25.8	28.3
$\text{NO}_3^-$ ( $\mu\text{g}/\text{m}^3$ )	0.9	1.0	1.3
$\text{NH}_4^+$ ( $\mu\text{g}/\text{m}^3$ )	1.9	8.6	7.7
Extinction by dry particulates			
Scattering ( $10^{-4}$ per meter)	0.90	2.99	3.04
Absorption ( $10^{-4}$ per meter)	0.13	0.19	
Maximum daily $\text{O}_3$ (ppb)	38	88	71
$\text{NH}_4^+/\text{SO}_4^{2-}\ddagger$	2.00	1.76	1.46

\*Total suspended particulates were measured with a General Motors low-volume sampler. †Fine particulate mass (diameter  $\leq 3.5 \mu\text{m}$ ), measured with a virtual impactor. ‡Molar ratio.

610 m above the site indicated  $\text{SO}_4^{2-}$  concentrations from 19.8 to  $32.9 \mu\text{g}/\text{m}^3$ .

The first haze episode (5 to 9 August) followed a transport pattern nearly identical to that of the second episode. It originated in the Midwest and Northeast and moved clockwise to the Gulf Coast. The third episode (4 to 11 September), however, traveled from the Midwest and Northeast to the Gulf Coast in a counterclockwise circulation induced by Hurricane David, which was moving up the East Coast. Once this haze reached the

Gulf Coast, it was advected clockwise around a high-pressure system that moved down from Canada into the eastern United States. This circulation transported the haze back to the Midwest.

As mentioned previously, satellite photographs were also used to determine the movements of the haze. The photographs were taken one to three times a day during daylight hours by the Synchronous Meteorological Satellite/Geostationary Operational Environmental Satellite. In many cases the haze was readily visible; nevertheless, all the images taken at 1600 hours (EST) were photographically enhanced (10) to better delineate the haze from clouds (Fig. 2). The streamlines on 4 September gave no indication that the haze would move southwestward around Hurricane David. The satellite photograph, however, shows that this did occur and removes any doubt that the Ohio Valley haze of 4 September was the same that reached northern Louisiana 1 day later.

It may reasonably be concluded that the summertime haze associated with maritime tropical air intruding into the northern United States from the Gulf of Mexico was at least partially formed in an air mass that traveled across source regions in the Midwest and Northeast. After the air mass traveled southward to the Gulf Coast, the subtropical environment caused it to acquire maritime tropical characteristics (increased temperature and humidity). Since all three episodes followed this pattern, we feel that this pattern may be responsible for the high concentrations of visibility-reducing particulates observed upwind of source regions in the Midwest and Northeast. Of the two transport routes, the clockwise route is probably much more prevalent because it is associated with weather patterns that occur more frequently (11).

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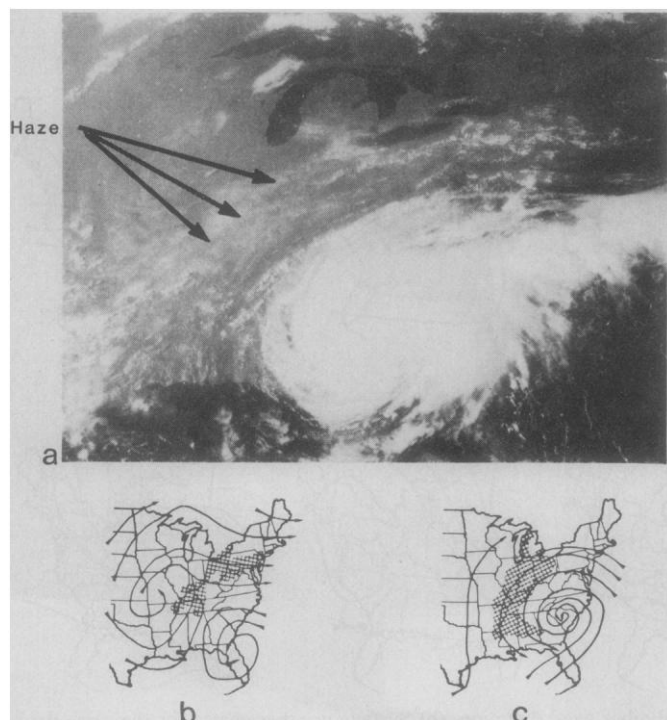


Fig. 2. (a) Enhanced photograph taken by satellite at 1600 hours (EST) on 4 September. (b) Haze at 1300 hours (EST) on 4 September. The map also shows the 850-mbar streamlines at 0700 hours on the same day. (c) Same as (b), except 1 day later.

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12. We thank G. Morris, M. Ruthkosky, W. Scruggs, and J. Zemla for conducting sampling and chemical analyses; D. Stroup for sampling, chemical analysis, and enhancing satellite photographs; D. Pierson for data management; M. Schauk of Baylor University for conducting aircraft sampling; and W. Lyons and his MESOMET staff for providing some of our synoptic weather data and for providing satellite images.

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## Hormesis: A Response to Low Environmental Concentrations of Petroleum Hydrocarbons

**Abstract.** *Crab zoeae (Rhithropanopeus harrisi) were exposed to water-soluble fractions of jet fuel (JP5) for the first 5 days or for the duration of zoeal development (11 to 14 days). Short-term exposure or continuous exposure to low concentrations of petroleum hydrocarbons caused no increase in mortality or changes in development rate, and increased megalopal weight was characteristic of such groups. This phenomenon, termed "hormesis," is probably a generalized aspect of environmental stress etiology but has seldom been reported as such.*

Most experiments dealing with the environmental effects of oil pollution have examined short-term determinations of acute toxicity (1, 2) or long-term effects on growth, development rate, or reproduction (3). The results of these experiments have often been used to assess the damage resulting from episodic events such as spectacular oil-tanker wrecks or offshore oil-well blowouts. Although these incidents receive a great deal of public scrutiny, they make relatively small contributions to the total petroleum hydrocarbon burden entering the marine environment each year (4). In many oil spill incidents, hydrocarbon concentrations return to base-line levels after the surface slick dissipates, usually several days to weeks after the event (5). Therefore, the question often arises of whether these short-term exposures to pollutants cause lasting harm to affected individuals after the exposure ends (6).

To investigate the recovery process after exposure to petroleum hydrocarbons, we exposed zoeae larvae of the mud crab, *Rhithropanopeus harrisi*, to water-soluble fractions (WSF) of jet fuel (JP5) for either the first 5 days or for the duration of zoeal development (11 to 14 days). There are four zoeal stages followed by metamorphosis to the megalops stage. The first zoeal stage, lasting 3 to 4 days, is the most sensitive to petroleum hydrocarbon exposure (7). The response in two salinities, 5 and 15 per mil (8), was determined for a control and a range of WSF concentrations from 10 to 100 percent of the original solution (9). Zoeae were reared in 8-cm (diameter)

finger bowls containing 50 ml of JP5 WSF with ten zoeae per bowl; there were three bowls per hatch to give 30 larvae from each female. Each day the zoeae were censused for living and dead animals. They were moved to clean bowls containing freshly prepared WSF

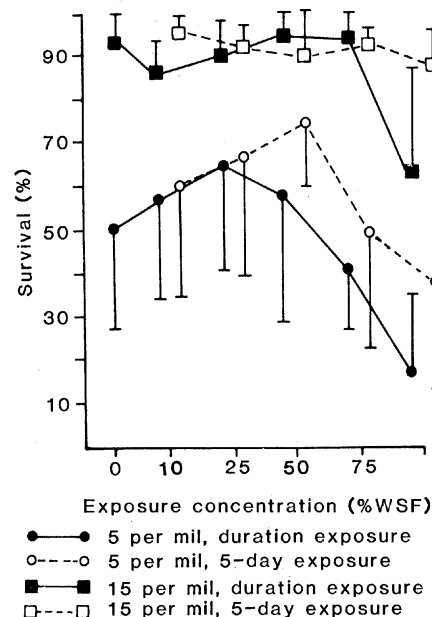


Fig. 1. Survival of zoeal mud crabs, *Rhithropanopeus harrisi*, exposed to JP5 WSF. Survival in 15 per mil salinity was much higher than in 5 per mil salinity. Lowest survival occurred in high, continuous WSF exposure in low salinities. For the purpose of clarity in this and subsequent figures, points for 5-day and duration exposure groups are slightly displaced relative to each other over the WSF designations. Vertical lines are 1 standard deviation of the mean.

or clean artificial seawater, as appropriate, and given freshly hatched *Artemia* nauplii as food. The indices of sublethal stress that we used were development rate and the weight of the megalops (10).

All the factors tested, salinity, WSF concentration, and the length of exposure, influenced the survival of the zoeae (Fig. 1). Zoeae in a salinity of 5 per mil showed markedly decreased survival compared to those in 15 per mil. Exposure to JP5 WSF caused much greater toxicity for zoeae in 5 per mil than in 15 per mil salinity, with maximum toxicity within each salinity occurring in groups exposed for the duration of zoeal development to 100 percent WSF. Under most 5-day exposure regimes, there was no effect of JP5 WSF on survival; however, there appeared to be enhanced survival in midrange and a decrease at maximum JP5 WSF concentrations in 5 per mil salinity. An analysis of variance showed that the effects of salinity and WSF concentration, but not the length of exposure, were significant ( $P < .01$ ) (11). By comparison, the JP5 WSF are much less toxic to developing crustaceans than those of other refined oils tested (7, 12) and would not be classified as acutely toxic ( $\geq 90$  percent mortality) as defined by Epifanio (13).

Sublethal effects of JP5 WSF exposure were evident in terms of changes in both development rate and the weight of the megalops. At each salinity, development rates were approximately the same, with a slight tendency for those in 15 per mil salinity to require longer at higher WSF doses to complete development (Fig. 2). Controls required 11.5 to 12 days to reach metamorphosis. In general, there was a dose-dependent increase in development time which was markedly greater in the group in 15 per mil salinity. Notably, at low JP5 WSF levels and low salinity the exposure duration exerted the least effect. Analysis of variance showed that all the factors tested contributed significantly to the variance ( $P < .01$ ) (10).

As shown here, megalopal dry weight is a sensitive indicator of sublethal stress, the degree of growth inhibition being proportional to toxicity. Larvae grown in low salinities showed the poorest performance overall (Fig. 3). The difference between the "duration" and 5-day groups is noteworthy. At each salinity, the line connecting the duration and 5-day groups tends to diverge with increasing WSF concentration. Differences between the two groups in each salinity are statistically significant ( $P < .001$ ). In every case, mean megalopal weights for 5-day groups were equal to or in