A Survey for Atmospheric Contamination following Forest Spraying with Fenitrothion

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Concern over environmental contamination with pesticides has increased in recent years, and has resulted in the curtailment of use of several persistent chlorinated insecticides, and their replacement with more selective and short-lived chemicals. In Canadian forest protection practices, for example, fenitrothion (0,0-dimethyl 0-(4-nitro-m-tolyl) phosphorothioate) has been used since 1968 in place of DDT for control of most lepidopterous defoliators. The first large-scale forest use of fenitrothion was in the 1969 Spruce budworm (Choristoneura fumiferana, Clem.) control programme in New Brunswick, when approximately 700,000 pounds of fenitrothion were applied by aircraft to approximately 3 million acres of pulpwood forest within a 4-week period (Fig. 1). Results obtained from monitoring the atmosphere during this spray operation are reported here, and comprise part of an ecological study of the fate of pesticides in the forest environment (1,2).

Contamination of the environment with pesticides has been studied less thoroughly in the atmosphere than in soil and water (3). It is known from relatively small-scale studies that much of the dosage applied by aircraft to land targets can be lost to the atmosphere and will drift locally (4,5). Also, atmospheric transport may account for pesticides being found in air, rain and dust remote from significant application sites (3,6).

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A preliminary report on the drifting of DDT from large forest-spraying operations was made by Yule and Cole (5) while developing air-sampling methods for pesticides, and Bamesberger and Adams (7) have reported herbicide concentrations occurring in the air during general seasonal and regional use. However, the present project was designed to give direct measurements of fenitrothion in the atmosphere associated with a specfic large-scale pesticide application.

Methods and Materials

Air Sampling

Survey design. The atmospheric survey was designed to give measurements of the phosphorus (P) component of fenitrothion in air, whose significance might be interpreted ecologically by reference to a quantitative indicator criterion such as human health standards. Five sets of air-sampling apparatus were operated near the main centres of population in New Brunswick and around the spray operation (Fig. 1). The air intake of the apparatus was set at a height of 5 feet above ground level, the sampling flow rate was regulated to an average human breathing rate of 12 litres per minute, and samples were changed and air meters read daily at noon. Meteorological data were collected for each sampler district, and operational spray data were obtained for each day (site, time and volume of application).

Sampling apparatus. The apparatus developed by Yule and Cole (5) for collecting particulate and vapour forms of pesticides from air was used for this survey. The apparatus comprised a florisil filter column linked in series with a bubbler containing glass-distilled dimethylformamide (DMF), and operated according to the human-exposure parameters given above. Airflows, solvent levels etc. were inspected every two hours, except at night, when inspection was 4-hourly.

Chemical Analysis

The daily florisil and DMF samples were shipped weekly to Ottawa for analysis by a modified total P method (8). This non-specific method of analysis was used to ensure detection of possible breakdown products of fenitrothion, and to accommodate the proposed use of some phosphamidon insecticide on the forest. Some specific analyses for fenitrothion using gas chromatography (flame-photometric detector) were performed on DMF samples collected in the apparatus of Bamesberger and Adams (7). These were taken at the Chatham and Fredericton sites, and served to confirm the presence of the parent compound in the air that was being sampled in the survey.

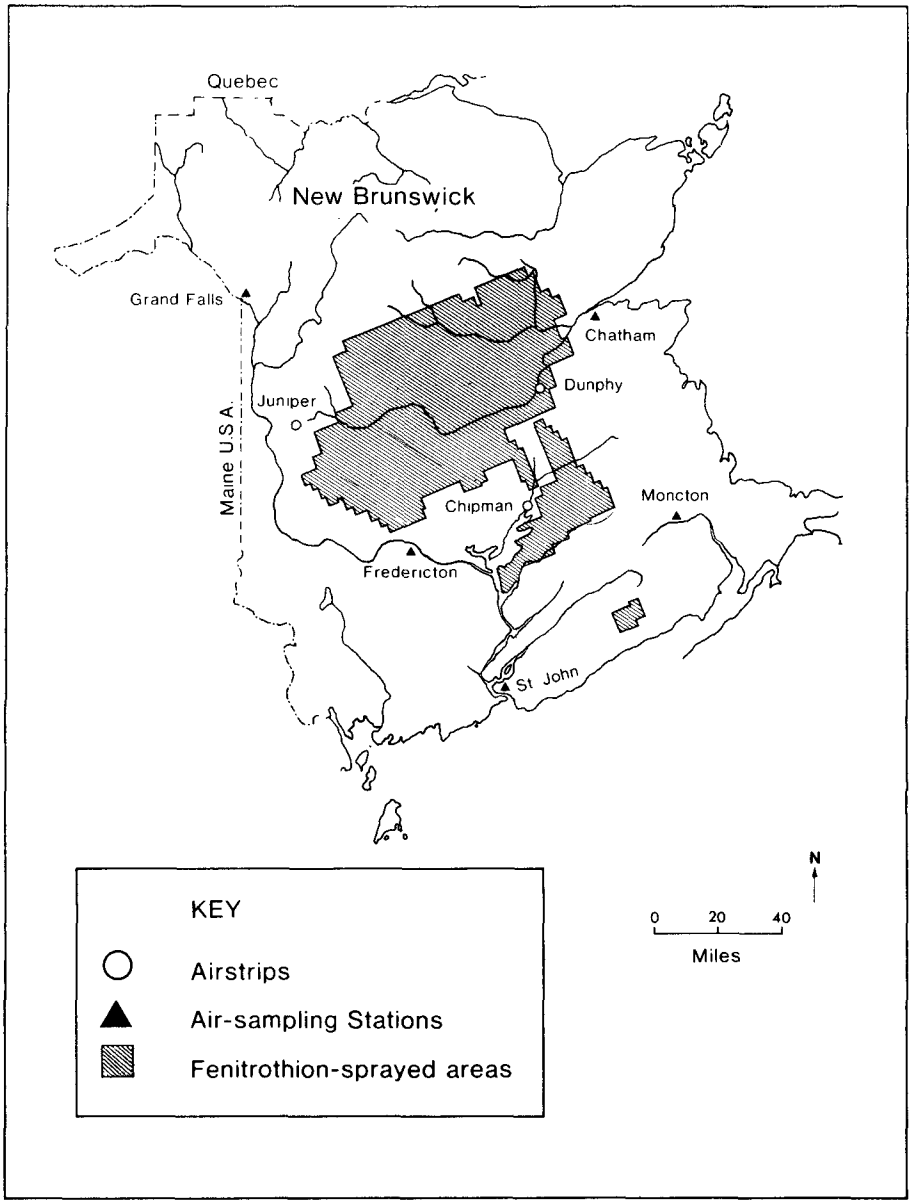


Figure 1. Map of New Brunswick showing the forest areas sprayed with fenitrothion in 1969.

Procedure. Florisil samples (20g.) were transferred and compacted in an acid-cleaned chromatographic column and eluted 4 times with 25.0 ml. dichloromethane, followed by 50.0 ml. water to displace the organic solvent into an acid-washed collecting beaker. The DMF samples (100 ml.) were transferred to an acid-washed beaker, and all volumes were reduced to 3 ml. prior to digestion, using low heat. Redistilled HNO_3 (10.0 ml.) was added to each sample beaker and the volume reduced to 3 ml. by heating. The solution was then cooled and $HClO_4$ (3.0 ml.) added and heated to dense white fumes. After further cooling, the beaker was rinsed with distilled water and reheated to dense white fumes.

The digested sample was made up to 10.0 ml. with distilled water, $HClO_4$ (1.0 ml.), ammonium molybdate solution (1.0 ml.) and freshly prepared amino-naphthol-sulphonic acid solution (0.5 ml.) were added, mixed and transferred to a clean cuvette. The colour reaction was measured exactly 10 minutes after mixing, using a Bausch and Lomb "Spectronic 20" with a red filter. Readings were taken at 760 mμ and the P content of samples estimated by reference to a standard P solution (K_2HPO_4) absorbance curve.

Results and Discussion

The total daily P collected in the twin air samplers at the 5 air-sampling stations are given in Figs. 2 and 3, together with daily fenitrothion spray outputs from the three airstrips, and summarised meteorological data for Fredericton. All daily data are from noon.

If the average pre-spray P background level is subtracted from the during-spray readings (Fig. 2), it is apparent that the population of New Brunswick was exposed to only very small amounts of fenitrothion for short periods during the local forest spray operation (Fig. 2). Brief increases in P concentration in air were found early and late in the application programme (Fig. 2). These peaks comprised somewhat more of the vapour form than the particulate form (Fig. 3), but this situation could not be correlated with meteorological records (Fig. 2), (cf. 7).

The human health significance of the survey results has been interpreted with the help of Dr. P.E. Braid, Occupational Health Division, Department of National Health and Welfare, Ottawa (personal communication).

It is known that the mammalian toxicity of fenitrothion has an intermediate value in the thiophosphate group of insecticides (eg. the acute oral LD_{50}'s for rats of parathion, fenitrothion, and malathion are 13, 250 and 2800 mg./kg.

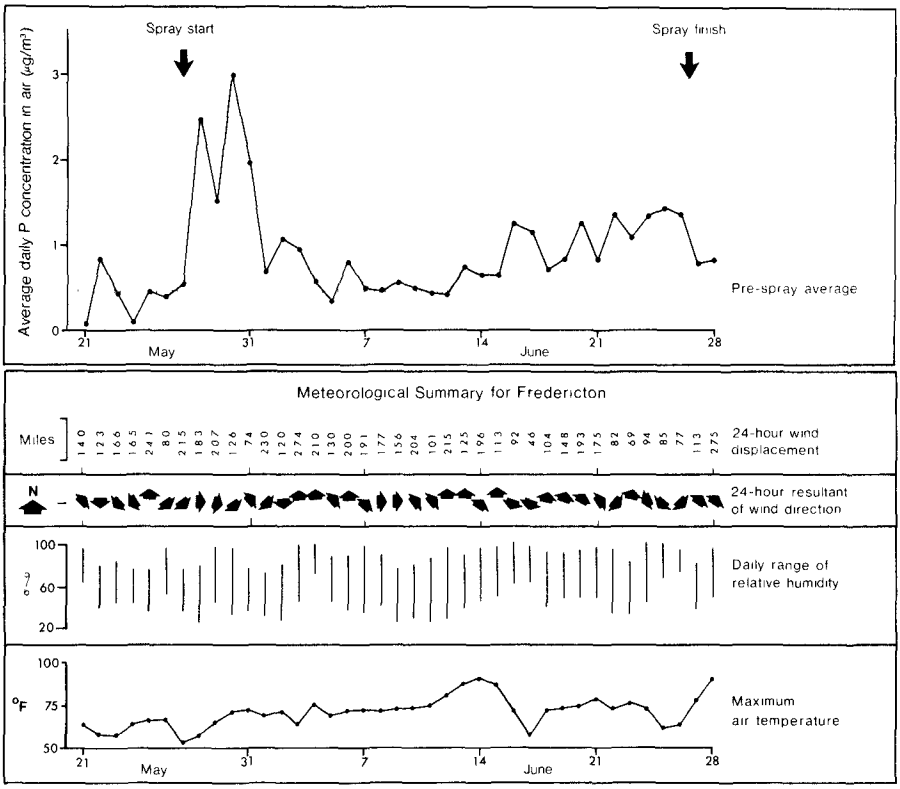
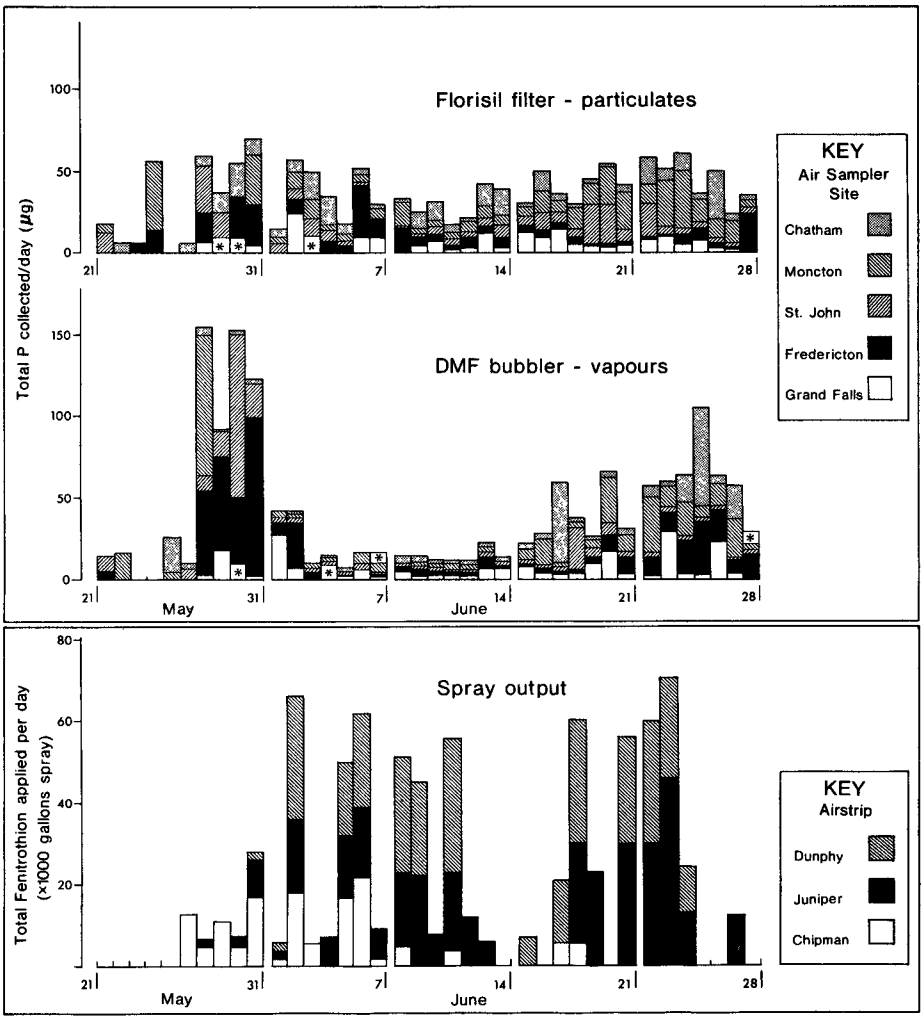


Figure 2. Average daily P concentrations (from 5 sampler stations) in the air of New Brunswick before and during the 1969 forest spray operation, with a summary of meteorological conditions at Fredericton during that period.

respectively) (9). In the absence of direct data relating to the toxicity of fenitrothion by the respiratory route, however, the most useful available value is its intravenous toxicity (LD_{50} rat, 33 mg./kg.) (10). The amounts of phosphorus collected daily in this atmospheric survey, if considered to be derived entirely from fenitrothion, all represent less that 0.01 mg./kg. body weight that might be inspired per human per day. Even assuming total absorption of inspired fenitrothion, a very much larger safety factor than the 100-fold normally used in toxicological studies between rat and man is available for concluding that the 1969 spray operation in New Brunswick's forests did not produce a health hazard to the local



* Sample lost

Figure 3. Daily output of fenitrothion by airstrip, together with total P collected as particulates and as vapours by air sampler station.

human population through atmospheric contamination. However, the increase in P content of the local atmosphere that was measured when the spray programme began (Figs. 2 & 3) does appear to demonstrate the occurrence of off-target environmental contamination under operational conditions.

Precise spray block locations were obtained for daily sorties but analysis of the data became too complex for the general purpose of this survey. If it is taken that airstrips served local forest districts (Fig. 1), a South to North shift in applications occurred with advancing date, and a similar trend was reflected in sampler-site contributions to the total P collected daily (Fig. 3). It may be concluded in this case that atmospheric contamination with insecticide could be measurable only locally, but the situation is complicated by meteorological effects.

The meterological data given in Fig. 2 is for Fredericton, a representative central location (Fig. 1), less subject to local coastal effects that most of the other sampler sites. Although the detailed local meteorological data could not be correlated with P collection and location, the general prevailing wind at this time was Westerly (Fig. 2), and the relatively large contributions of P from the samplers at Chatham and Moncton to the total daily P collected during the general spray program indicate a down-wind drift effect (Fig. 3) (5).

In conclusion, it was found by this survey that a large forest spray operation contributed relatively small amounts of insecticide to atmospheric contamination, and to other parts of the environment outside the target area. The average daily intake of fenitrothion over the duration of the spray operation is considered not to constitute a health hazard to the local human population, the criterion used as an indicator of ecological significance. Atmospheric contamination appeared to incorporate local application and down-wind drift component factors, but the cause-and-effect situation in such a large operation is very complicated and apparently not amenable to precise analysis under local maritime weather conditions.

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