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ETEX: A EUROPEAN TRACER EXPERIMENT; OBSERVATIONS, DISPERSION MODELLING AND EMERGENCY RESPONSE

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Abstract—In Autumn, 1994, two releases of perfluorocarbon tracers from north-east France were tracked across northern Europe using a network of 168 ground stations with limited airborne sampling support. Simulating an emergency response situation, modellers from 20 countries reported their long-range dispersion predictions, initially within a few hours of the release and then over the coming days in line with the evolving meteorological data. Subsequent comparison of the predictions with the environmental results showed model performance varying from good, for the conditions of the first release in the majority of cases, to unsatisfactory for the second release in all cases. The experimental database now established represents a unique tool for investigating the effectiveness of future model developments. The papers in this special issue reflect the major scientific results © 1998 Published by Elsevier Science Ltd. All rights reserved

INTRODUCTION

By the mid-1980s development of atmospheric dispersion models for point source releases already stretched back over many years but in 1986 the Chernobyl accident served as a rude reminder that relatively little effort had been devoted to atmospheric long-range transport models. Deficiencies covered both our ability to understand and represent dispersion processes on the spatial and temporal scales involved and our capacity to acquire and treat the necessarily large amounts of data. Appreciable progress has been achieved on both fronts especially in data communications and computing power, but a further barrier exists in the form of the relatively sparse experimental data against which model results can be compared. Thus validation work post-Chernobyl has frequently relied on data resulting from that

accident. Such data, however, were heterogeneous (Raes *et al.*, 1990) often lacking in quality control and, being scattered over many institutes, usually only selectively available to individual modellers (Apsimon *et al.*, 1987; Wheeler, 1988; Pudykiewicz, 1988; Albergel *et al.*, 1988; Kimura and Yoshikawa, 1988; Pudykiewicz, 1989; Piedelièvre *et al.*, 1990; Hass *et al.*, 1990; Maryon *et al.*, 1991; Ishikawa, 1995). Moreover, no common protocols existed for evaluation of the model results. Accordingly, in 1987 the European Commission (EC) initiated the Atmospheric Model Evaluation Study (ATMES) co-sponsored by the International Atomic Energy Agency (IAEA) and the World Meteorological Organisation (WMO). ATMES assembled an extensive quality-controlled database of environmental measurements from many institutes and arranged access to archived meteorological data at the European Centre for Medium-term Weather Forecasting (ECMWF). A best estimate of the source term was established in co-operation with the Russian authorities and computerized procedures for statistical evaluation of the results from different

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models were set up. The results (Klug *et al.*, 1992) were encouraging but ATMES itself was subject to certain limitations—irresolvable uncertainties in the source term, the intrinsic heterogeneity of the environmental data, the fact that the models were not run entirely blind against the test data due to various prior publications and, with an eye to any further accident, had not been tested under emergency response conditions.

The European Tracer Experiment (ETEX) was initiated in 1992, again cosponsored by the EC, the IAEA and WMO, for the purpose of addressing these deficiencies. It thus involved controlled tracer releases, systematic environmental monitoring at up to 2000 km, the collection of model predictions in simulated emergency response conditions and the evaluation of these predictions based on the software developed for ATMES purposes.

This introductory paper contains a brief summary of the preparations for the field campaign and the experiment and contains the main findings of the dispersion modelling results and emergency response analysis.

The articles in this special volume are all devoted to ETEX. They address a broad range of aspects of the experiment and the dispersion modelling, including (i) a description of the field campaign, (ii) the role of the meteorological conditions, (iii) model validation, data assimilation and basic dispersion studies, (iv) statistical aspects of comparison between model output and observation, and (v) emergency response modelling.

PREPARATORY WORK

The preparations for ETEX started in 1992, well in advance of the experiment, in order to select a suitable tracer and to co-ordinate simultaneously and on short notice the release activities, the start of sampling at the stations and the aircraft operations as well as communications. All these procedures had to be tested out and checked in the period well in advance of the experiment. Perfluorocarbon compounds are suitable tracer substances for experiments on transport over long distances (Dietz, 1986; Begley *et al.*, 1988). They are non-toxic, non-depositing, non-water-soluble, inert and environmentally safe. At ambient pressure and temperature they are odourless and clear liquids which are released into the atmosphere by spraying the liquid into a hot air stream, causing it to evaporate.

A total of 168 stations, all part of the synoptic network of national meteorological services in 17 countries were equipped with sequential air samplers. The air samples were collected in metal tubes filled with absorbing material. They were distributed prior to the experiment and sent back to Ispra afterwards for chemical analysis by thermally desorbing the metal tubes and using gas chromatography with electron capture detection.

Two studies were carried out to determine the ambient levels of the PFC tracers in Europe. The first took place in early 1994 and the second in October 1994 just before the experiment (Piringer *et al.*, 1997). At all ETEX stations adsorption tubes were exposed for 14 days and their contents analysed at Brookhaven National Laboratory (BNL), USA to determine PFC levels. For quality control reasons, at a couple of measuring sites duplicate sampling was performed. A high number of field and laboratory blanks were distributed. Also the PFC standards in use at BNL and Ispra were compared (Nodop *et al.*, 1998).

THE EXPERIMENT

To conduct a tracer experiment in Europe where meteorological conditions with westerly-south-westerly air flow prevail, a release location in the western part of France was selected. During the first release on 23 October 1994, starting at 16:00 UTC, perfluoromethylcyclohexane (PMCH) was used. In the second release on 14 November 1994 at 15:00 UTC perfluoromethylcyclopentane (PMCP), was used to avoid cross contamination. Both releases lasted 12 h. A total of 340 kg PMCH and 490 kg PMCP was emitted, respectively, corresponding with average release rates of 7.95 and 11.56 g s⁻¹.

During the first release a rather strong west to south-westerly flow was advecting the tracer in the direction of the sampling network. The synoptic situation was characterized by a cold front over middle Europe. A deep low (975 hPa) east of Scotland was slowly moving north, maintaining a strong south-westerly flow over the release site and central Europe. There was no centre of high or low pressure and no extending ridges or troughs were foreseeable to pass, or be very close to the release site. A day later unstable flow was still observed over the release site and the advection area. However, because of the northerly movement of the low pressure area over the North Sea, the winds decreased after the end of the release.

During the second release a weak frontal system moved in from the west. The cold front passed at the end of the 12 h release period. The situation during the release was characterized by a deep low between Iceland and Norway, moving slowly eastward and filling. Strong south-westerly winds were blowing at the release site, but decreased and veered to west, after the cold front passed in the early morning hours (Gryning *et al.*, this volume).

During both releases dispersion characteristics of the atmospheric boundary layer near and at the release site were monitored with constant level balloons floating at different heights in the atmosphere (Koffi *et al.*, 1998; Stohl and Koffi, 1998). Balloons were tracked for up to 100 km from the release site. Their trajectories were stored together with other *in situ* data in a meteorological data base.

Each station took 24 samples with a sampling time of 3 h per sample, in total covering a period of 72 h. The sampling was initiated just before the expected arrival time of the tracer plume at each station. The last measurements were taken 96 h after the start of the release in the easternmost countries. During each release, over 5000 air samples were taken. The laborious chemical analysis and quality control took approximately two years (Nodop *et al.*, 1997a, b). During both releases aircraft have been monitoring on line the dispersing plume. During the first release tracer was measured at four different altitudes enabling in principle a three-dimensional analysis of the dispersing plume. Results of this analysis will be shortly available.

Table 1. ETEX participants

Country	Institute Acronym
Belgium	KMI
Bulgaria	BMI
Canada	CMC
Czech R.	CHI
Denmark	DMI
Finland	FMI
France	METEOFRACTANCE
France	IPSN
Germany	DWD
Israel	IIBR
Italy	ANPA
Japan	JAERI
Netherlands	RIVM
Netherlands	KNMI
Norway	NMI
Rumania	RMI
Russia	TYPHOON
Slovak R.	SHMU
Sweden	SMHI
Switzerland	SMI
United Kingdom	MetOffice
United States	NOAA
United States	ARAC
United States	SRS

DISPERSION MODELLING RESULTS

Twenty four institutions took part in the real-time forecasting of the cloud evolution, with 28 long-range dispersion models, using the meteorological forecast data from various sources and later from the ECMWF. (see Table 1). Surface concentration evolution at the locations where the tracer was sampled was simulated. The results of these calculations were compared for both experiments with the measurements. A short overview of the basic principles used in dispersion modelling is given below.

Mathematical concepts in dispersion modelling

The dispersion of passive (not interacting with the dynamics of the fluid in which it disperses) tracer is generally described by a (tracer) mass-conservation equation which can be formulated as

$$\frac{\partial \rho c}{\partial t} + \nabla \cdot (\rho u c) = 0$$

where ρ denotes the atmospheric density, u , the three dimensional velocity field and c the tracer mixing ratio field. Using the fluid mass continuity equation this can be transformed to

$$\frac{dc}{dt} = 0.$$

where the operator $d/dt \equiv \partial/\partial t + \mathbf{u} \cdot \nabla$, expressing that the mixing ratio is conserved along a fluid trajectory. Molecular diffusion is neglected. Decomposing the variables in mean and fluctuating quantities and aver-

aging the resulting equation is

$$\frac{\partial C}{\partial t} + \mathbf{U} \cdot \nabla C = \frac{1}{\rho_0} \nabla \cdot (\rho_0 \mathbf{K} \nabla C)$$

where upper case symbols denote mean quantities and ρ_0 expresses the atmospheric density which is assumed to be a function of height only. The term at the right-hand side is an approximation for the turbulent transport induced by the fluctuations. It contains the eddy-diffusivity tensor \mathbf{K} . It is a dominant term certainly in the vertical direction and also in low horizontal mean-wind conditions. In order to solve the last equation \mathbf{U} and \mathbf{K} are required as input data. These data are normally obtained from weather prediction models.

An alternative for the above, Eulerian description is the so-called Lagrangian description of dispersion where the position of individual fluid particles is evaluated from

$$\mathbf{X}(t) - \mathbf{X}(t_0) = \int_{t_0}^t \mathbf{U}(t') dt.$$

Here, \mathbf{X} denotes the fluid particle position and \mathbf{U} its velocity, which also may consist of a mean and a fluctuating component. Any initial concentration field at time t_0 can now be mapped onto a new field at arbitrary time t , through integration of the above equation which displaces fluid particles (with conserved mixing ratios). The Eulerian and Lagrangian description are mathematically equivalent, though the absence of numerical diffusion in the latter method offers some advantages certainly in the case of a single point source.

Since it was judged impractical to extend the scope of the project to include communications through the transfrontier governmental networks that will be used in the case of a real accident, an "emergency" network, specific to the purposes of ETEX was created, using fax and electronic mail for communication. Modellers were informed in advance that the two releases would occur within the time window 15 October to 15 December 1994, and that they would be called to react in real time. They were, however, notified of the exact time and location only after the start of the release.

The procedures adopted required the modellers to transmit their results initially by fax to the evaluation team in Ispra as soon as possible, and to send subsequently the same information in digital form for statistical processing.

The quantitative evaluation of the predicted concentrations led to the conclusion that there is a large number of models which give satisfactory results for the first release (Archer *et al.*, 1996). In Fig. 1 some statistical results of this evaluation are summarized. A more comprehensive review is given by Graziani *et al.* (1998). The fair agreement between model prediction and data for the first release does not seem to be strictly dependent on the type and complexity of the model used, although, not surprisingly, coarser

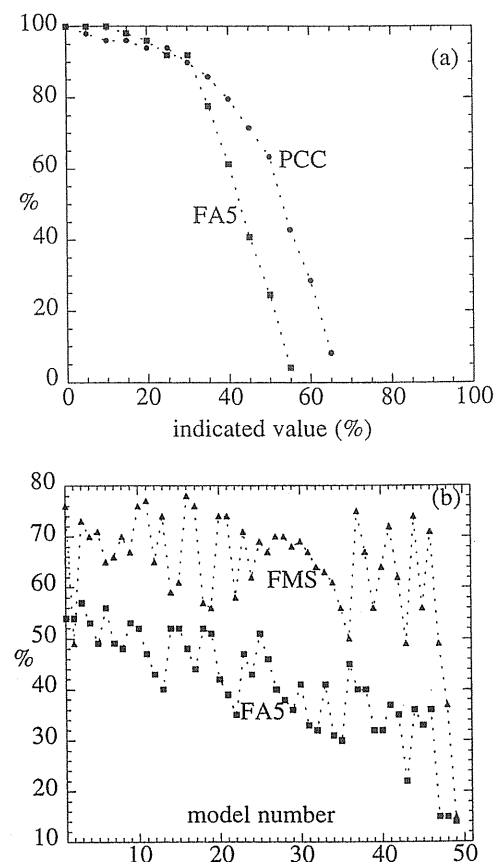


Fig. 1. Model performance as derived from a statistical comparison between 72.3 h observed and predicted concentrations at 168 stations for the first release. The data used here result from a "post factum" analysis, using observed meteorological fields and are generally superior to the real time results. In total 49 models were included in this analysis. (a) Along the vertical axis the percentage of models is shown for which the statistical parameter PCC and FA5 exceeds the indicated value along the horizontal axis. PCC denotes the (linear) correlation coefficient on the logarithmic values. FA5 denotes the percentage of predictions which are within a factor of 5 from the observed values. (b) Along the horizontal axis the models are ranked from high to low according to their value of the correlation coefficient (PCC). Along the vertical axis the statistical parameters FA5 and FMS are shown. The FMS is the figure of merit in space for the time integrated concentration. If A_m and A_o , respectively, denote the relative areas where the model and observed dose exceeds a given value, the dose FMS is defined as the ratio between intersection and union of these areas ($(A_m \cap A_o) / (A_m \cup A_o)$). The best models shown here have a PCC > 0.6, an FA5 > 0.5 and an FMS > 0.7.

meteorological input fields produce concentration values that are less accurate than those produced using finer scale meteorological models.

For the majority of models the differences between using the forecast data and analysed meteorological data are not significant. This may indicate that the current meteorological (mesoscale) models could be considered adequate for emergency response pur-

poses, at least in this selected simple meteorological situation.

The second release showed larger discrepancies between observations and model results where all models significantly overpredicted surface concentrations after 12 h from the start of the release. No clear explanation has yet been given for this result. Further, the evaluation of model performances was more difficult, since both the number of sites hit by the tracer and the concentration levels were reduced, due to the strong winds during the release period. The results obtained by the large majority of the models indicate consistently an initial north-easterly cloud displacement, bending more towards the east with time. After about 24 h the tracer cloud had left France. This evolution is not confirmed by the experimental results, which indicated that only a part of the release behaved as foreseen by the models, while a part of the material was still found within a few hundred kilometres east of the release site 24–48 h later. No final conclusions could be derived yet for the second release. Possible stratification of the released air-tracer mixture is being investigated, together with other local effects which might have altered the near-field evolution of the plume in the particular conditions of the second release.

MAIN FINDINGS

ETEX demonstrated the capability of conducting a continental scale tracer experiment across Europe using the perfluorocarbon tracer technique and assembled a unique experimental database including tracer concentrations and meteorological data. It successfully established a communication network through which meteorological services and other institutes disseminated on demand in real-time their forecasts of concentrations of material released into the atmosphere.

It further created widespread interest and resulted in considerable dispersion model improvement and development (see e.g. Pudykiewicz; Robertson and Langner, this volume), as well as in reinforcement of communication and collaboration between national institutes and international organizations. As a consequence, a large number of institutes can (and will in the case of a real accident) predict the long-range atmospheric dispersion of a pollutant cloud.

While it is encouraging that many modellers can produce predictions fairly rapidly, it is evident that a much better appreciation is needed of the quality of the various predictions, especially of their inherent limitations which may vary considerably with the complexity of the meteorological conditions encountered in practice; this is an essential input for effective decision support.

Even more important, however, will be to reconcile the disparate forecasts of different modelling groups or agencies, with a view to promote more coherent

and appropriately qualified input to decision makers in the event of a future accident. In any case a proliferation of independent judgements, which would be inevitably discordant, is obviously undesirable, even if this discordance may serve to remind the decision maker of our current limited capability in this area.

The rapidity of modellers in predicting the cloud evolution and transmitting the results to a central point (within 3–6 h), the precision in predicting arrival times of the cloud (± 3 –6 h) and maximum surface concentrations (within a factor of 3) at selected, close and remote locations should be viewed as the currently achievable limit of accuracy in real-time long-range dispersion modelling in relatively "simple" meteorological conditions. Presumably the quality of predictions would diminish for more complex conditions. In emergency response the time of arrival and the maximum concentrations are crucial answers for governments to the questions: (a) is my country in danger and (b) if so, how much danger.

In the two years following the experiment, considerable improvements in comparison with the real-time results were found which can be attributed to improvement of the dispersion model formulation, the use of more detailed meteorological data or to a tuning of model parameters. Nevertheless, a considerable disparity in the concentration values as presented by the different models persisted.

Major uncertainties in the position and intensity of a dispersing cloud will remain to exist also in the future since they will always include the basic uncertainty of a meteorological prediction. Deficiencies in the (diagnostic) description of a dispersing cloud using observed meteorological data should be added to this uncertainty.

The experiment contained a number of artificialities compared with what would be experienced in nuclear accident (e.g. no deposition, no monitoring data in real time as material disperses). The availability of such data would enhance the quality of the dispersion forecasts compared with the ETEX experiment. Few of the current models, however, have a capability to utilise monitoring data in real time to enhance predictions and this is an area for future developments (Robertson and Langner, this volume).

CONCLUSIONS

• Based on the experiments no preference was found for the mathematical description (Lagrangian vs Eulerian models) with regard to its performance. Some Eulerian model results improved by initiating the dispersion with a Lagrangian particle dispersion model in the area near the source.

• Increasing the resolution of meteorological data improves model performance (see e.g. Sørensen *et al.*, 1981; Nässtrom and Pace, 1998, this volume).

• Successful application of theories of atmospheric boundary layer structure and turbulence in an arbitrary environment in varying and complex conditions is still far from perfect: it appeared that simplified dispersion schemes, using e.g. constant boundary-layer height, still rank among the ones with the best performance (Brandt *et al.*, 1981; Desiato *et al.*, 1998, this volume).

• Evidence from the second experiment indicates that neglecting massive intermittent and local vertical transport occurring in clouds or cloud systems (deep convection) may be a serious defect, which may have contributed to the large overprediction of surface concentrations in the second release.

• Dispersion predictions using observed meteorological fields differ from those using forecast meteorological fields. These differences are, however, not significant in view of the generally large uncertainties in model predictions.

• The ETEX database can be used to test and improve data assimilation algorithms and has much potential to elucidate this issue.

There is hardly a better application of atmospheric sciences than in the field of weather forecast and environmental safety and it is a challenge to translate scientific progress to the benefit of our modern society. Since the accident in Chernobyl, slightly more than a decade ago, we have made enormous progress in communication and computer technology, so that we are now capable of providing swiftly the information and predictions so desperately needed at that time. However, ETEX has demonstrated the variable quality of predictions and their lack of agreement. This makes real-time atmospheric dispersion modelling an immature technology and no decision maker is encouraged to base his (draconian) decisions on model predictions only. Though we have to accept the intrinsic uncertainties in meteorological predictions, accuracy of dispersion formulation certainly can and must improve. Advancements made, facilitated by increasing computer power and higher resolution models, can be tested with the present data base and may eventually lead to the design of a future experiment, where it is recommended to use other measurements strategies, in particular those which include the determination of 3-D concentration fields and the possibility to make measurement data directly available in order to be used in data assimilation techniques.

Acknowledgments—ETEX is sponsored by the EC, the IAEA and the WMO. The project is managed by the JRC, Environment Institute, which has responsibility for the experiments and the evaluation of the models' performances. ETEX was made possible by the enthusiastic participation of the national weather services and responsible institutes inside and outside Europe. Special thanks go to the site personnel operating the samplers. The release site was made available by the University of Rennes, Radio Communications Faculty. Their contributions are gratefully acknowledged.



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THE FIELD CAMPAIGNS OF THE EUROPEAN TRACER EXPERIMENT (ETEX): OVERVIEW AND RESULTS

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Abstract—As part of the European Tracer Experiment (ETEX) two successful atmospheric experiments were carried out in October and November, 1994. Perfluorocarbon (PFC) tracers were released into the atmosphere in Monterfil, Brittany, and air samples were taken at 168 stations in 17 European countries for 72 h after the release. Upper air tracer measurements were made from three aircraft. During the first experiment a westerly air flow transported the tracer plume north-eastwards across Europe. During the second release the flow was eastwards. The results from the ground sampling network allowed the determination of the cloud evolution as far as Sweden, Poland and Bulgaria. This demonstrated that the PFT technique can be successfully applied in long-range tracer experiments up to 2000 km. Typical background concentrations of the tracer used are around $5-7 \text{ fl/l}^{-1}$ in ambient air. Concentrations in the plume ranged from 10 to above 200 fl/l^{-1} . The tracer release characteristics, the tracer concentrations at the ground and in upper air, the routine and additional meteorological observations at the ground level and in upper air, trajectories derived from constant-level balloons and the meteorological input fields for long-range transport models are assembled in the ETEX database. The ETEX database is accessible via the Internet. Here, an overview is given of the design of the experiment, the methods used and the data obtained. © 1998 Elsevier Science Ltd. All rights reserved.

Key word index: Perfluorocarbons, tracer experiment, long-range transport, model evaluation, database.

1. INTRODUCTION

The European Tracer Experiment (ETEX) was established to evaluate the validity of long-range transport models for real-time application in emergency management and to assemble a database which will allow the evaluation of long-range atmospheric dispersion models in general (Klug *et al.*, 1993; Girardi *et al.*, 1997). The objectives of ETEX were to (1) conduct a long-range atmospheric tracer experiment with controlled releases under well-defined conditions; (2) test the capabilities of organisations in Europe responsible for producing rapid forecasts of atmospheric dispersion to produce such forecasts in real time; and (3) evaluate the validity of their forecasts by comparison with the experimental data. This paper presents an overview of the design of the experiment, the methods used and the data obtained. Detailed discussion and analysis of the ETEX data set are given in subsequent papers in this Special Issue.

2. EXPERIMENTAL DESIGN

2.1. General considerations

The European Tracer Experiment was designed as a major field study to simulate long-range transport of a pollutant in

the atmosphere in western Europe. The ETEX experimental phase comprised two separate releases of perfluorocarbon tracer (PFT). Both utilised a total of 168 ground-based sampling sites in 17 European countries, together with three aircraft. Such a large logistical effort required considerable planning spanning two years, briefly summarised as

- definition of suitable meteorological conditions,
- choice of release site,
- choice of ground-level sampling sites,
- choice, construction, validation and deployment of samplers,
- preparation of sampling tubes,
- establishment and validation of analytical procedures,
- establishment of data management protocols,
- co-ordination of the release, sampling and modelling efforts,
- collation, dissemination and archiving of the resultant database.

2.2. Release site

The release location was selected with the aim of maximising the probability of identifying in advance meteorological situations which would ensure the dispersion of the tracer being within the area covered by the ground sampling stations. In order to conduct a tracer experiment over a distance

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