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Briefs

U.S. Oil Supply Reflects World Events
Successful MFTF-B Technology Demonstration

ii

Improving Tank Track Pads

We are analyzing designs and materials for improving tank track pads to extend their service life and reduce their replacement costs.

1

Atmospheric Ozone: Zeroing In

After nearly a decade of study, we have arrived at two conclusions concerning ozone in the atmosphere: first, that the factors affecting atmospheric ozone concentrations are far more complex than first believed and, second, that compensating effects make the depletion of total ozone less than originally expected.

11

Nuclear Waste Storage: Evaluating the Uncertainties

We have developed a systematic method for evaluating the relative importance of the factors involved in predicting the performance of a nuclear waste repository and for estimating the effects of individual uncertainties on the overall uncertainty in our predictions.

22

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Briefs

U.S. Oil Supply Reflects World Events

In late 1976, U.S. crude oil and petroleum product imports increased to a level at which they equaled domestic crude oil and lease-condensate production. Early 1977 represented a turning point since, even if one included the production of natural gas liquids, imports exceeded domestic production. Henceforth, the country seemed destined to rely on foreign sources to supply the largest share of its demand for oil. At that time, the LLNL Energy and Resource Planning Group developed a graph to monitor domestic production and imports of oil. This plot has been updated periodically with moving four-week averages from the *Oil and Gas Journal*.

It might be expected that the causes of specific variations in crude oil production and imports would be difficult

to ascertain, and this is certainly true for some of the minor fluctuations shown on the plot. The larger perturbations, however, can be attributed with some confidence to particular events, as shown graphically on the following pages.

The upper line (in green) charts the moving four-week average of domestic production of crude oil; this line does not include an additional 1.5 million barrels per day of natural gas liquids that are normally considered part of domestic oil production. The lower line (in brown) plots U.S. imports of crude oil and petroleum products. The numbered circles are centered on significant variations in these two sources of petroleum that can be explained by the events cited briefly in the associated numbered boxes.

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Key Words: crude oils; natural gas; oil supply; petroleum—imports, production.

Abbreviations and Definitions

API	American Petroleum Institute
CIA	Central Intelligence Agency
DOE	Department of Energy
D&N	DeGolyer and MacNaughton, Dallas, Texas
FRB	Federal Reserve Bulletin
h-d-d	Heating degree-day: the number of degrees the daily mean temperature falls below 65°F. This unit is not a day (as in 24 hours); it is, rather, a measure of the coldness of the day and therefore of the amount of heating required.
IEA	International Energy Agency
lease condensate	Liquid hydrocarbons produced with natural gas and separated from the gas at the well on the lease.
NGL	Natural gas liquids
NOAA	National Oceanic and Atmospheric Administration
OAPEC	Organization of Arab Petroleum Exporting Countries (Algeria, Egypt, Iraq, Kuwait, Libya, Qatar, Saudi Arabia, Syria, United Arab Emirates)
OPEC	Organization of Petroleum Exporting Countries (Algeria, Ecuador, Gabon, Indonesia, Iran, Iraq, Kuwait, Socialist People's Libyan Arab Jamahiriya, Nigeria, Qatar, Saudi Arabia, United Arab Emirates, Venezuela)
PIW	Petroleum Intelligence Weekly
PR	Petroleum Review
s-h-d-d	Seasonal heating degree-day: the total number of heating degree-days for all days in the period from July to June. For example, if the heating season had 365 days and all days had a h-d-d of 5°, then $365 \times 5 = 1825$ s-h-d-d. The higher the s-h-d-d, the colder the winter.
TAPS	Trans-Alaskan Pipeline System
USBLS	U.S. Bureau of Labor Statistics

Millions of barrels per day

10.0
9.0
8.0
7.0
6.0
5.0
4.0

1973 1974 1975

75

1976

1977

1978

1979

1980

1981

1982

Successful MFTF-B Technology Demonstration

The MFTF-B, our next-generation Mirror Fusion Test Facility, will contain many state-of-the-art components and operating techniques in a large tandem-mirror experimental device. Prudence dictated that these advanced concepts be demonstrated as early in the construction schedule as practical, to provide design assurance with a minimum of disruption and expense. Accordingly, our plan included a series of Technology Demonstration tests of various subsystems, originally purchased for MFTF and now destined for MFTF-B, to minimize the technical risks associated with operating so far in advance of ordinary engineering practice. These subsystems included the vessel, the cryopanel, the external vacuum, the cryogenics, the local controls, the instrumentation, and the supervisory control from a central systems console.

In February 1982, we completed the last test in this series, the Integrated Systems Test of all the above systems together with the superconducting yin-yang magnet. The results of this very successful demonstration follow:

- The magnet system performed at a design current of 5775 A in full superconducting operation with a good margin below the critical current (above which the magnet ceases to be a superconductor). The magnet temperature was slightly below the expected 4.5 K. The strain gauges on the magnet case tracked almost exactly the predicted values during current buildup, measuring almost 98 MN (22 million pounds) of spreading force against the 0.13-m-thick stainless steel case and the intercoil structure. We also successfully executed two fast dumps and one slow dump of the stored energy and operated with a 20% current imbalance between the two coils (5 kA in one coil and 4 kA in the other).
- The injector magnetic shield for the neutral beam sources performed better than expected, providing a magnetic field of only 100 μ T at the ion source arc chamber instead of the predicted 150 μ T.

- The vacuum vessel provided good vacuum integrity in the pressure range from 1 to 100 μ Pa. We observed only minor leakage during the high-speed pumping tests (both air and deuterium) of the liquid-helium-cooled cryopanel. We achieved an ultimate vacuum of 1.2 μ Pa.

- We also demonstrated the expected external vacuum system characteristics with respect to rough pumping and external cryopumping, achieving a vessel pressure of 100 μ Pa. We readily achieved regeneration of the cryopanel by warming them from 4.5 K to about 15 K and easily pumped away the deuterium released.

- The cryogenic system achieved or exceeded its design performance. The helium refrigerator/liquefier provided 3.2 kW of cooling, compared to the specified 3.075 kW. The liquefaction mode provided 700 l/h of liquid helium, compared to the contract specification of 600 l/h. The measured heat load for the magnet and cryogenic system was 700 W, consistent with the calculated 500 W of magnet heating and 200 W for the cryopumping and valves.

- We fully demonstrated the hardware and software of the hierarchical computerized MFTF-B control system (and gained valuable operating experience) during the tests of the integrated systems. The tests verified the overall design of sensors, remote operating elements, data transfer, data management, displays, console touch-panel operations, timing, discrimination, fault identification, redundant fail-safe computer-crash recovery, and diagnoses with both the hardware and software elements interactively involved with supervisory console operators. The touch-panel controls proved to be most effective and versatile in all the operator control functions exercised during the tests.

- We demonstrated data transfer from local control and monitoring stations to the CAMAC (Computer Automated Measurement and Control) system 150 m away, with light pipes providing very effective high-voltage isolation and noise reduction. Control

verification and relay to the system console worked well and fully demonstrated the software and hardware. We accomplished almost real-time update or modification of displays and exercised effective operating control throughout the technology demonstration testing program.

Successful completion of the Technology Demonstration tests gives added confidence for the design of MFTF-B components and for the use of the fully

computerized control and data-handling techniques. Achievement of this major milestone clears the way for further MFTF-B construction, now scheduled for completion in 1985.

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Key Words: mirror fusion; tandem mirror; yin-yang magnet.

IMPROVING TRACK PADS

Track pads currently
have an unacceptable
incurring growing
we are analyzing
materials to solve

on U.S. Army tanks
high failure rate,
air and replacement costs;
alternative designs and
this problem.

The
tracks
on U.S.

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Army tanks are
faced with rubber
pads (Fig. 1) designed
to reduce vibration, noise,
and damage during travel on
roadways and on cross-country ter-
rain. These pads, which support the
weight of the vehicle, have a limited ser-
vice life. For vehicles weighing over 45 900 kg
(primarily the M48 and M1 tanks), pad failures con-
stitute the major share of track repair and replacement
costs. Because of the growing number of the heavier and
more powerful M1 tanks entering the inventory, these costs,
now about \$650 million per year, are projected to rise to over \$250
million by 1989 (in constant 1981 dollars) unless there are mitigating
changes. Improvements in pad material and design are thus essential to
controlling the expected cost surge.

We are studying the track-pad problem for the U.S. Army Tank-Automotive Com-
mand. In our approach, unique in this application, computer models provide a key
link between laboratory measurements of material properties and the responses of
materials under simulated field conditions. Properties of candidate materials are first
evaluated in the laboratory. The models provide parametric values that are used in
mechanical and thermal models of the track. We then subject candidate materials to
laboratory failure-mechanism tests in the laboratory and use the results to refine the
models. These tests duplicate failure modes identified in field observations. We
evaluate the thermal stability of pad samples by simulating vehicle operation with the
chemical decomposition that may occur in failure. The approach is a marriage of
materials and design in order of feasibility.

Two failure modes indicate that the service life of tank pads may be the most
commonly ignored in testing and modeling: the fatigue and degradation of the
cross-section of the pad. In the past, the track pad has been treated as a
mass specimen in laboratory tests. In contrast, the models require that the
parameters of the track-pad system be defined in terms of the mechanical and
chemical properties of the material. The models are used to predict the failure

Fig. 1

Three views of the T142 tank track, the subject of our study. The octagonal rubber pads are fixed to a steel plate and are attached to the tank track by a threaded steel bolt.

The Mechanical Model

Both our mechanical and thermal analyses of pad materials were based on a cross section of the T142 track used on the M60 series of tanks (Fig. 3). We used the Laboratory's NIKE2D finite-element computer code¹ to model the deformation behavior of the track in the y - z plane (y is the direction of motion of the track; z is perpendicular to the pad's surface). (High-speed motion pictures of the T142 track on an operating tank showed that pad deformation was restricted to this plane.)

A finite-element mesh of the T142 track was used in our mechanical model. We made extensive use of the NIKE2D code's slide-line capability. That is, we incorporated the effect of the roadwheel moving over a section of the track by displacing the roadwheel downward (in the z direction) until it supported the weight of the vehicle and then sliding it horizontally (in the y direction) across its path. The slide-line capability also enabled us to apply frictional loads at the interface between the pad and its supporting surface.

Tension applied by the track was also modeled.

The material properties that are input to the NIKE2D code consist of parameters describing the shear-relaxation behavior of a linear viscoelastic model. As shear-relaxation behavior varies widely among elastomers, these are key parameters in optimizing the pad material selected for a particular track design. They are obtained from the storage and loss moduli of the material. (The storage modulus reflects inherent stiffness, the loss modulus the inherent dissipation of energy as heat.) We measured this modulus by cyclically loading samples in a servohydraulic test machine.

We analyze the output of the mechanical model to obtain two kinds of

information. One is the rate of hysteretic heat generation. Rubber that is cyclically loaded produces heat. The rate of heat generation is a function of temperature. As temperatures in a stressed pad rise very slowly, requiring more than 1.5 h to approach a steady state, they change little during the fraction of a second a pad is loaded and unloaded. This enables us to treat the deformation of the pad at any instant as an isothermal event.

To find the heat-generation rate, we experimentally evaluated the shear-relaxation behavior of the rubber at different temperatures. These parameter values were then input to the model to determine the mechanical response of the material at the same temperatures.

Fig. 2

(a) The T142 track pad as manufactured. (b) Pad showing abrasion failure caused by road wear. (c) Cutting-and-chunking failure, caused by the initiation and propagation of cuts. (d) Blowout failure, caused by high internal pad temperatures that lead to catastrophic rupture.

Fig. 3

Cross-sectional and plane views of the T142 track. Pins through the binoculars are linked by end connectors that join individual track segments. Computer models analyze cross sections of the track.

The rate of heat generation is proportional to the area within the cyclic stress-strain curve. Figure 4 shows an example of this curve obtained for an element of rubber located in the center of the pad. The heat-generation rate thus calculated is input to the thermal model.

The second kind of information provided by the mechanical model comprises the stresses, strains, and strain energies developed in the pad for various operating scenarios. In one typical scenario, the track pad is subjected to the uniform loading and unloading encountered on a paved road. In certain elastomers, this operating scenario can result in blowout failure. (Cutting and chunking does not occur in this scenario because stresses on the track pad are almost entirely compressive. Cuts in rubber tend to propagate in response to tensile stresses.) We calculated the contours of maximum principal stress and the state of stress (represented by a

Mohr's circle) for one element in the region of high deformation. We also used these results in experimental testing to evaluate accumulated damage to the material.

In another operating scenario, the track pad contacts a simulated rigid obstacle typical of the localized loads encountered in cross-country terrain. A region of high tensile stress develops in the pad ahead of the obstacle (Fig. 5). As noted, this type of stress can cause pad failure of the cutting-and-chunking variety. Our high-speed photographic studies have shown that tensile stresses produced in the track pad when it is scrubbed over obstacles are another source of cutting-and-chunking failure. We also applied the NIKE2D code to this scenario.

The Thermal Model

Our thermal model of the track-pad problem was also based on a cross section of the T142 track (see Fig. 3). We used the Laboratory's TACO2D computer code² to perform the thermal analysis for pad sections in the y - z plane. Because of the pad's high length-to-width ratio and the poor thermal diffusivity of rubber, we assumed that heat losses at the end of the pad have relatively little influence on temperatures in interior sections. Thus, for interior sections, heat flow occurs only in the plane section under study. We verified this approach with temperature measurements in the field.

Specifying thermal boundary conditions presented a challenge. Heat is transferred from the pad of a moving tank by the integrated effects of conduction when the pad is in contact with the ground and by convection when it is off the ground. In addition, heat transfer is affected by films (such as dirt) on the pad, its mechanical condition, air movements, and other factors. We found that heat transfer can be adequately modeled by assuming convective heat losses at the pad's surface. We chose a heat transfer coefficient on the basis of surface temperature-time measurements made in the field on a moving M60 tank. Material properties (obtained from laboratory tests) were

Fig. 4

Mechanical finite-element model of the T142 track (right) and a plot of the cyclic stress-strain behavior of an element located at the center of the track pad (left). The area within the stress-strain curve represents heat produced in the material. The heat-generation rate calculated from this is used in the thermal model.

Fig. 5

Contours of maximum principal stress for the operating scenario in which the track pad contacts a rigid obstacle. Large tensile stresses are produced in a localized region within the pad. The state of stress, represented by a Mohr's circle, is shown for this region. (In a Mohr's circle, normal stresses are plotted on the abscissa; positive values are tensile and negative values are compressive. Stresses are in megapascals.)

incorporated as a function of temperature.

We carried out thermal calculations for a track pad operating on a paved road, as in our mechanical-model scenario. The predicted temperature-time response agreed closely with the response measured in the field on an M60 tank. The maximum deviation between the calculated and field data was about 6°C.

Figure 6 shows our calculated constant-temperature contours in the pad after about 6000 s (1.67 h) of simulated operation. The maximum temperature, at the center of the pad, is over 130°C; the surface temperature is about 90°C. Information obtained from the thermal model was also used in the failure-evaluation portion of our analysis.

Material Studies

The material currently used in tank track pads is styrene butadiene rubber (SBR) that contains carbon black as a reinforcing filler material. The properties of SBR can vary over a wide range, depending largely on the amount and

type of carbon black and the cross-link density of the rubber polymer. Our studies to date have been restricted to this material. We have examined both new and field-tested samples from three vendors. We have also tested several of our own formulations of SBR manufactured under laboratory conditions. The thermal and mechanical properties of SBR were studied as a function of temperature; for comparison, the room-temperature properties of SBR follow:

- Thermal conductivity ranged from 0.354 to 0.264 W/m·°C.
- Specific heat ranged from 1420 to 1290 J/kg·°C.
- Density ranged from 1.19 to 1.16 Mg/m³.
- Storage component of complex Young's modulus ranged from 10.0 to 5.5 MPa.
- Loss component of complex Young's modulus ranged from 2.5 to 1.6 MPa.
- Bulk modulus was 1.7 GPa.

The thermal analysis showed that the most significant input parameter was the heat-generation rate, which is

Fig. 6

Calculated constant-temperature contours for the T142 track after 6000 s (1.67 h) of simulated operation on a paved course. Temperatures at the center of the pad exceed 130°C; those at the surface are almost 90°C. The average temperature is 101.5°C. Metallic portions of the track are not color-contoured.

Fig. 7

The T156 tank track shoe currently being used on the M1 tank. This track design differs from the T142 track in that the shoe is a solid block of rubber and the pad is not removable.

sensitive both to material properties and to track design. Varying parameters other than the heat-generation rate caused pad temperatures to vary no more than 4°C after 6000 s (1.67 h) of vehicle operation. Varying the heat generation rate, on the other hand, affected pad temperatures by about 30°C. This variation in temperature can have a significant effect on the pad's ability to resist cutting-and-chunking failure.

Applying the mechanical model to the material properties listed above showed us that both storage and loss moduli play an important role in determining the heat-generation rate. In the materials we studied, structural changes that increased the storage modulus also increased the loss modulus. Moreover, these two properties tended to influence the rate of heat production in opposite directions: increasing the storage modulus decreased the heat-generation rate, whereas increasing the loss modulus increased the heat-generation rate. For a given track design there should be an optimal combination of these properties. With the materials studied and the T142 track design, the stiffest rubber (with the largest storage modulus) had the smallest heat-generation rate. Thus, we believe that increasing the stiffness of the SBR used for the T142 track pads should extend their

service life by significantly reducing their operating temperature.

Tearing energy varied dramatically with temperature, decreasing by about 50% from room temperature to 140°C.

Design Studies

Our studies of alternative track designs are just beginning. Our approach is to study several designs currently in use to establish the character of the problem. In addition to the T142 track, we are examining two quite different track designs: the track used on the British Chieftain tank and the T156 track used on the U.S. Army M1 tank. The track on the British Chieftain tank is a single pin design (one pin per track segment) and has a removable pad.

In contrast to the T142 track, which has a steel body with a removable track pad, the T156 track consists of a solid block of rubber (Fig. 7). When subjected to thermal analysis, the T156 track

reaches a maximum temperature over 160°C (compared with 130°C for the T142), and the surface temperature of the pad is greater than 100°C. The higher temperature results largely from the T156 track's greater heat-production rate (although the thicker rubber on the T156 is also a contributing factor).

Failure Evaluation

Our experimental tests were designed to measure the resistance of candidate track-pad materials to the two types of failure mode (cutting-chunking and blowout) at various temperatures. Cutting-and-chunking resistance was measured by two kinds of test. In the first, we measured the tearing energy (toughness) of the elastomer. (Tearing energy is the change in strain energy of a cracked body per unit of crack ad-

vance; a crack grows when tearing energy reaches a critical value.) This property reflects the inherent ability of the material to resist the propagation of a cut or flaw, and therefore the subsequent removal of a piece of rubber. Our test samples were rubber strips with two cuts perpendicular to their loading axis (Fig. 8, inset). We tensile-loaded the samples to failure at different temperatures.

Figure 8 shows the results of our tearing-energy tests. Tearing energy varied dramatically with temperature, decreasing by about 50% from room temperature to 140°C. This result clearly indicates that a decrease in a pad's operating temperature should substantially improve its service life.

Our second series of tests, just beginning, will evaluate the resistance of a

Fig. 8

Results of laboratory tearing-energy tests. The test samples were rubber strips with two cuts perpendicular to their loading axis (inset). The samples were tensile-loaded to failure at different temperatures. Tearing energy varies dramatically with temperature, decreasing by about 50% from room temperature to 140°C. A decrease in a pad's operating temperature should substantially improve its service life.

pad material to cumulative damage. Damage consists of any mechanical or chemical degradation (cuts and voids or thermal-mechanical oxidation) that promotes the loss of small pieces of material from the track pad. We first condition an uncut strip of rubber by cyclically loading it (for a predetermined number of cycles) to the stress levels predicted by our mechanical model. (Tests are performed at the temperature predicted by the thermal model.) The samples are then tensile-loaded to failure. Comparing the ultimate tensile strength of the cyclically conditioned sample with that of an unconditioned sample gives us a quantitative measure of cumulative damage.

We are also using thermogravimetric analysis (TGA) to measure any weight change in a track pad heated in a known atmosphere. A weight loss may correlate with the chemical decomposition that precedes a blowout. We determined the weight losses in three rubber track pads, obtained from different manufacturers, as a function of temperature. The net loss is small in each case, even at 200°C, well above the operating temperature; at 200°C, the net loss is less than 3%. We are also correlating TGA results with results from the cumulative-damage tests.

Summary and Conclusions

We have developed an experimental-computational method for studying track-pad failure on tanks. In our approach, unique in this application, computer modeling provides a key link between laboratory measurements of material properties and the responses of materials under simulated field conditions. We input the values of material properties to computer models that calculate temperature and stress levels. The materials are then experimentally evaluated for failure resistance at the temperature and stress levels obtained

from the models. Our failure-resistance tests are uniquely based on failure modes identified in field observations.

Our method enables us to evaluate candidate pad materials and designs in a straightforward and relatively inexpensive manner. It can also help to reduce the cost of track pads by identifying critical material properties and their values for use in engineering specifications.

Our studies thus far show that temperature plays a key role in promoting track-pad failure. In addition, we have demonstrated that the tensile stresses associated with cutting-and-chunking failure are produced in typical operating scenarios. As the properties of rubber are strongly temperature dependent, the ability of pads to resist failure declines as their temperature increases. To improve the service life of pads, it is therefore essential to control their temperature while maintaining adequate tearing energy and damage resistance.

Our studies so far have been restricted to several styrene butadiene rubbers. The temperatures reached in these materials are driven by the heat-generation rate. Our analysis shows that increasing the stiffness of these elastomers will prolong pad service life by reducing this rate. □

Key Words: M1 tank; M60 tank; main battle tank; NIKE2D code; T142 track, TACO2D code; track pad.

Notes and References

1. J. O. Hallquist, *NIKE2D: An Implicit, Finite-Deformation, Finite-Element Code for Analyzing the Static and Dynamic Response of Two-Dimensional Solids*, Lawrence Livermore National Laboratory, Rept. UCRL-52678 (1979).
2. P. J. Burns, *TACO2D—A Finite Element Heat Transfer Code*, Lawrence Livermore National Laboratory, Rept. UCID-17980, Rev. 2 (1982). (TACO2D is a modified and extended version of the TACO code written by W. E. Mason, Jr.)

Atmospheric Ozone: Zeroing In

We are beginning to understand why measured changes in the concentration of atmospheric ozone are smaller than those predicted on the basis of known chlorocarbon emissions.

Over the past decade, there has been considerable political, public, and scientific interest in the extent to which man can inadvertently modify stratospheric ozone. Recognizing that even a fairly small perturbation to stratospheric ozone concentrations has potentially significant biological, agricultural, and climatic implications, regulatory actions to protect the stratospheric ozone layer are being considered in the United States and throughout the world.

In the early 1970s, we began to develop numerical models that would enable us to study the chemical and physical processes controlling the stratospheric ozone distribution. Since that time, we have been involved in theoretical studies of the existing atmosphere and assessments of man's past and possible future influence on it. We have made important contributions to both national and international evaluations of man's total impact on the atmosphere.¹

Included in these evaluations are analyses of the potential impact on stratospheric ozone of nitric oxide and water-vapor emissions from aircraft, nitric oxides produced by nuclear explosions, chlorine emissions from the space shuttle, and man-caused global emissions of such gases as nitrous oxide (N_2O), carbon dioxide (CO_2), and

chlorocarbons (especially $CFCl_3$ and CF_2Cl_2). In recent years, the potential effects from chlorocarbon emissions have been of particular concern because, if they reach the stratosphere in large enough quantities, their dissociation by sunlight might significantly affect stratospheric ozone concentrations through ozone-destroying catalytic reactions.

For all its importance, ozone (O_3) represents only a few parts per million of the gases in the atmosphere. If all the ozone in the atmosphere were concentrated into a layer at sea level, the layer would be only 3 mm thick. Most of the ozone is located in the stratosphere: maximum concentrations are found near an altitude of 23 km in the temperate latitudes.

The stratosphere extends from about 16 to 50 km above the earth's surface in the tropics and from 8 to 50 km in the arctic. In the troposphere (the layer below the stratosphere), temperature generally decreases with increasing altitude; however, in the stratosphere, the temperature increases with altitude out to almost 50 km. Thus, the troposphere is turbulent and subject to rapid vertical mixing, whereas the stratosphere is relatively stagnant. As a consequence, substances that reach the stratosphere can stay there for years.

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The importance of the relatively small amount of ozone in the earth's atmosphere lies mainly in its ability to absorb . . . solar ultraviolet radiation.

The importance of the relatively small amount of ozone in the earth's atmosphere lies mainly in its ability to absorb almost all of the short-wave (290 nm or less) and most of the biologically harmful (280- to 320-nm) solar ultraviolet radiation. Ozone stops this radiation from reaching the surface of the earth where it could affect every living thing: plant, animal, and human.

Perhaps the most widely discussed harmful effect of solar ultraviolet radiation is skin cancer. In the United States, there are about 300 000 new skin cancer cases each year, of which about 2500 are fatal. The incidence increases toward the south; in midlatitudes (30 to 45°N), it doubles for each 10° closer to the equator.

Because ozone attenuates solar radiation in the 280- to 320-nm wavelength region so strongly, a small reduction in ozone causes a much larger increase in the ultraviolet flux at the surface. For example, a 10% ozone reduction increases the ultraviolet flux at midlatitudes by about 20%. In terms of increased risk of skin cancer, this is about the same as moving 400 km south, a little more than half the distance from San Francisco to Los Angeles. A 20% increase in the ultraviolet flux would be expected to cause some 60 000 new cases of skin cancer (and 500 deaths) each year in the United States alone.

A reduction and redistribution of ozone might also produce significant climatic effects. In addition to ultraviolet light, ozone also absorbs infrared radiation and some visible light. This absorption helps maintain the world's heat balance and is directly responsible for the temperature inversion that stagnates the stratosphere.

On the basis of simplified, one-dimensional calculations of a climate model, the change in the global mean surface temperature resulting from expected changes in ozone concentration caused by supersonic aircraft and chlorofluoromethanes might be either up or down, and probably would be less than 0.2°C. However, there could be much larger changes regionally, especially at high latitudes.

Furthermore, it should be emphasized that this prediction applies for changes in ozone only. Changes in carbon dioxide concentration can produce much larger temperature changes. Even the chlorocarbon compounds themselves, before they dissociate, can affect global temperatures.

Calculations with two- and three-dimensional models suggest that an increase in surface temperature might increase rain and snow worldwide. However, uncertainties regarding cloud feedback processes and how changes in climate will affect cloudiness make it hard to calculate such rainfall changes accurately.

Modeling the Atmosphere

The processes that determine the distribution of chemical species in the atmosphere take place in three dimensions (four, if we include time). However, the complexity of stratospheric chemistry and the interactions among chemistry, radiation, and fluid dynamics make three-dimensional general circulation studies of stratospheric ozone extremely difficult and expensive. To date, three-dimensional models have been used primarily to study dynamic processes and have included only very simplified chemical schemes.

The basic tool in all previous studies of tropospheric and stratospheric processes and the effects of perturbations has been some form of one-dimensional, coupled transport and chemical kinetics model² to simulate the vertical distribution of atmospheric trace species. Our one-dimensional model³ presently includes 141 chemical and photochemical reactions to determine the concentrations of 47 minor atmospheric constituents. It calculates hemispheric-averaged net vertical transport by an altitude-dependent diffusion coefficient. Multiple scattering effects are included in calculations of the photodissociation rate and are updated at each time step as a function of albedo, altitude, and solar zenith angle.

Our model is designed for time-dependent calculations and can be used in either diurnal or diurnal-averaged

modes. We also can add a radiative-convective model to the transport-kinetics model to study the effects of temperature coupling between changes in composition and reaction rate coefficients. Results from two- and three-dimensional models are to be consistent with the globally averaged changes in ozone calculated for perturbations with the one-dimensional model.

We are currently developing a zonally averaged two-dimensional model containing essentially the same chemistry as the one-dimensional model but with seasonally varying north-south and vertical transport. This model will serve to elaborate on seasonal and latitudinal variations in ozone and other species. A key feature of this model is its modular design, which allows us to analyze its components in detail. Current emphasis is on a careful examination of the major physical and chemical processes encompassed in such models and their proper or practical representation.

Chlorocarbons in the Atmosphere

It was noted in 1974 that the chlorofluorocarbons, CFCl_3 and CF_2Cl_2 (also referred to as CFC-11 and CFC-12, respectively), appear to last long enough in the atmosphere so that their decomposition products could significantly add to the chlorine burden in the stratosphere.⁴ On the basis of present production and emission levels, these two chemicals are still the chlorocarbons of greatest concern.

However, many other chlorocarbons could eventually affect ozone. Large quantities of various chlorocarbons are presently being produced for use as solvents, refrigerants, and propellants in aerosols. The amounts of these substances that eventually reach the stratosphere depend not only on the quantities produced but also on the fraction actually released into the atmosphere and the fraction that survives to reach the stratosphere.

Measurements of CFC-11 and CFC-12 in the troposphere go back almost a decade; Fig. 1 shows the

Fig. 1 Measurements of CFC_{11} (CFC-11) in the northern and southern hemispheres as a function of time.

measurements of CFC-11 collected over this time period by various investigators. The CFC-12 measurements show a comparable increase over this period, but reach much higher concentrations by the year 1980: 320 pptv (parts per trillion by volume) compared with 180 pptv for CFC-11. The concentrations of both compounds continue to increase at a rate of about 10% per year (even though global emission rates have, over the past 9 years, decreased slightly).

Other chlorocarbons have been measured in the atmosphere for a much shorter period of time. Methylchloroform (CH_3CCl_3) concentrations are increasing about as fast as CFC-11 and CFC-12. The present mixing ratio of methylchloroform appears to be about 130 pptv.

Table 1 presents data for worldwide industrial production and for the rate of release into the atmosphere of CFC-11 and CFC-12 since 1975. The annual release of these compounds has decreased slowly since 1976, primarily as a result of the concern about possible ozone changes. Before 1973, industrial

production was increasing by about 10% per year. If we use these historical release estimates in our theoretical model of the atmosphere, the calculated chlorocarbon concentrations agree reasonably well with the measurements described above.

Unless there is some rapid process for their removal, trace gases released into the troposphere will distribute themselves worldwide within a few years; transport into the stratosphere takes much longer. While in the troposphere, a chemical is subject to several removal processes. A water soluble compound, such as hydrochloric acid, can be removed by rain and snow. Hence, its concentration will decrease rapidly with altitude in the troposphere.

Other chemicals are so reactive that their destruction rates in the troposphere are rapid in comparison with transport processes into the stratosphere. Chlorocarbons having carbon-hydrogen bonds or carbon-carbon double bonds (e.g., methylchloroform) react so strongly with hydroxyl (OH) groups in the troposphere that only a small fraction ever reaches the stratosphere. However, completely halogenated chlorocarbons, such as CFC-11 and CFC-12, react very slowly in the troposphere.

The major removal process for CFC-11 and CFC-12 is photolysis in the stratosphere. Measurements of these chlorocarbon species in the stratosphere show a rapid decrease in concentration with altitude, in agreement with theoretical expectations. We have tried to identify other removal processes of comparable efficiency, particularly in the troposphere, but without success.

Ozone is produced in the stratosphere by the photolysis of molecular oxygen (O_2) into free oxygen atoms by solar radiation at wavelengths less than 242 nm. The resulting oxygen atoms react rapidly with molecular oxygen to produce ozone. Since molecular oxygen is one of the major constituents in the atmosphere, the stratospheric production of ozone is relatively insensitive to man's activities.

The destruction of stratospheric ozone, however, comes about primarily through the catalytic effects of various free radical species. The major recognized catalytic loss mechanisms involve NO_x (NO , NO_2), HO_x (H , OH , HO_2) and ClO_x (Cl , ClO) species.

It is generally assumed, on the basis of laboratory data, that the chlorine atoms in the chlorocarbons are rapidly transformed into their chemically reactive ClO_x forms once the chlorocarbons are photolyzed or react with other atmospheric constituents. There are several mechanisms by which ClO_x can catalyze the destruction of stratospheric ozone.⁵ The most important of these catalytic cycles is:

The net effect of this mechanism is to destroy one ozone molecule without using up either the Cl or the ClO molecules involved.

Since the ClO_x comes through the catalytic cycle unchanged, it can react with ozone over and over again. Each chlorine atom can destroy thousands of ozone molecules before it is removed by some other process. Because of this catalytic effect, it takes only a small concentration of these ClO_x molecules to do a lot of damage.

Eventually, the ClO_x molecules are converted into longer lived and less reactive species such as HCl . Such species act as temporary sinks for ClO_x . Once these relatively inactive species work their way back into the troposphere, precipitation can remove them rapidly.

Table 2 shows the atmospheric lifetimes of nine chlorocarbons as calculated by our one-dimensional model. These nine compounds are those of most current interest and represent the wide range of calculated lifetimes. The lifetimes range from 4 years for CHFCl_2 to 385 years for CF_2ClCF_3 . The calculated lifetimes for CFC-11 and CFC-12 of 64 and 108 years, respectively, are in good agreement with estimates based on available measurements.

Table 1 Worldwide production and release rates of CFC-11 and CFC-12 (CFCl_3 and CF_2Cl_2 , respectively) in millions of kilograms per year.

Year	CFC-11		CFC-12	
	Production	Release	Production	Release
1975	323.5	312.9	418.6	412.0
1976	349.9	304.1	449.8	395.7
1977	332.2	306.4	424.4	376.5
1978	321.2	291.0	414.1	347.6
1979	302.0	272.0	400.3	338.0
1980	302.2	257.4	393.4	333.7

Table 2 Calculated atmospheric lifetimes and efficiencies in destroying stratospheric ozone of various chlorocarbons relative to the ozone-destroying efficiency of CFCl_3 (CFC-11).

Species	Atmospheric lifetime, ^a years	Relative efficiency, ^a per kilogram of chlorocarbon emitted
CFCl_3 (CFC-11)	64	1.00
CF_2Cl_2 (CFC-12)	108	0.86
$\text{CFCl}_2\text{CF}_2\text{Cl}$ (CFC-113)	88	0.80
$\text{CF}_3\text{ClCF}_2\text{Cl}$ (CFC-114)	181	0.60
CF_3ClCF_3 (CFC-115)	385	0.52
CHF_2Cl (CFC-22)	28	0.05
CH_2Cl_2	9	0.15
CCl_4	57	1.11
CHFCl_2 (CFC-21)	4	0.04

^aBased on results of LLNL one-dimensional model.

Table 2 also shows the calculated efficiencies of the nine chlorocarbons for destroying stratospheric ozone, relative (by weight) to CFC-11 in each case. Since industrial production statistics are based on weight of production, such relative efficiencies are particularly relevant.

CFC-11 and CCl_4 are the most efficient at destroying ozone per pound of emission. Those chlorocarbons with tropospheric removal mechanisms (e.g., CH_3CCl_3 or CHFCl_2) are much less effective. Other factors affecting the relative ozone-destroying efficiencies are the number of chlorine atoms per chlorocarbon molecule and the chlorocarbon's reactivity as a function of altitude in the stratosphere. Table 2 also shows that there is no direct relation between atmospheric lifetime and the calculated relative ozone destruction efficiency.

Theory vs Measurement

In the years since the possibility that chlorocarbon emissions could damage the stratospheric ozone layer was first identified, model-predicted effects on ozone from these chlorocarbons have undergone several large variations. These variations have been strongly influenced by both uncertainties and improvements in our understanding of stratospheric chemistry.

Figure 2 shows how our estimates of the change in total ozone, assuming constant CFC emissions at the 1976 rate, have fluctuated over the years since we began this study. The calcu-

lated changes in total ozone are relative to a background atmosphere that does not contain these compounds. The current calculations predict a change in the total ozone column of -0.6% by January 1980 and -5.0% at steady state (hundreds of years from now).

Since mid-1979, we have also been including the effect of methylchloroform at the current emission level of 476 metric tons per year. Calculations indicate that about a tenth of the -5.0% predicted change in total ozone at steady state would be due to methylchloroform.

The calculations shown in Fig. 2 are representative of the results found by other models over this time period. While the trend over the six-year period in other models is quite similar to that determined with our model, at any given time the changes in total ozone calculated by different models have differed by several percent.

These differences (as a function of time over this period) primarily reflect changes to the chemistry as new laboratory measurements of rate coefficients became available. Over this six-year period, our understanding of the processes governing the chemistry of the stratosphere has greatly improved. Nonetheless, significant chemical uncertainties still remain, as suggested by the substantial changes to the ozone depletion calculated in the recent past.

Therefore, although the chemistry presently used in the model represents the best current evaluation of existing laboratory data, changes in the chemistry used in atmospheric models seem likely to continue. Yet, after six years of intensive research, the essence of the theory that chlorine from chlorocarbons may significantly reduce stratospheric ozone concentrations remains unchanged.

Ozone in the Last Decade

Ultimately, the validity of the changes in ozone calculated by the various models must be tested against observed trends of both the total ozone column amount and its distribution as a

Fig. 2

Change since 1975 in our one-dimensional model calculations of the expected effect on total ozone at steady state from constant emissions of CFC_3 (CFC-11) and CF_2Cl_2 (CFC-12). The effect of CH_3CCl_3 (methylchloroform) has been included since 1979.

function of altitude in the atmosphere. However, our establishment of a global ozone trend is affected by uncertainties connected with monitoring and evaluating the ozone data. These uncertainties include instrumental and calibration errors, sampling errors, random errors, and long-term natural variations in ozone.

The total ozone column of the atmosphere has been monitored for about 20 years by a network of ground-based measuring stations equipped primarily with Dobson spectrophotometers. Three statistical studies for the detection of total ozone changes during the last decade (1970-1980) have been made recently using time-series analysis.⁶⁻⁸ These studies are based on data from

36 Dobson stations, most of them located in the northern hemisphere.

Due to remaining uncertainties, each of these studies gives a range representing the 95% confidence (2σ) levels for global change in total ozone over the decade. The results from these analyses are shown in Fig. 3. The most recent of these time-series analyses⁸ accounts for possible prior changes in ozone due to the 1961 to 1962 atmospheric nuclear test series.⁹ This analysis yields a measured increase in total ozone over the 1970s of 0.3% with an uncertainty of 1.3%.

Calculations with our model, including the effects of all the important chlorocarbon sources (those in Table 2), indicate a change in total ozone over

Fig. 3

Comparison of statistical trend analyses of ground-based measurements of total ozone and model-calculated changes in total ozone from 1970 to 1980.

the 1970s of -0.5% . As shown in Fig. 3, the results for this case are nearly outside the range of the statistical limits of the ozone trend calculated by the other three studies.

It is possible that the statistical limits fail to account for all existing uncertainties in the ozone measurement or in the trend analysis itself. More probably, the calculations neglect the effects of other natural and artificial causes for the variations in ozone. When we extended our calculations¹⁰ to include the contributions from increasing emissions of nitrogen oxides from aircraft, plus carbon dioxide (CO_2) and nitrous oxide (N_2O) from combustion and other sources, we found that the net overall effect of these sources appears to compensate for the reduction in total ozone caused by the chlorocarbons.

This compensation is due primarily to two effects: CO_2 -induced cooling of the stratosphere, which reduces the effectiveness of catalytic ozone-destruction mechanisms, and increased ozone levels in the upper troposphere and lower stratosphere from aircraft emissions of nitrogen oxides. Including these additional artificial sources produces a calculated increase in total

ozone of 0.1% during the decade. This essentially eliminates any discrepancy between our calculated results and the statistical analyses of total ozone change over the last decade.

Our calculations indicate that most of the ozone destruction due to chlorocarbons takes place in the upper stratosphere, about 40 km above the earth's surface (Fig. 4). Hence, it is at this altitude that statistically significant changes in ozone due to chlorocarbons may first be detectable. At present, however, measurements at this altitude are extremely limited and no firm conclusion can be drawn. Preliminary data from satellites (Hudson, Ref. 1) indicate that ozone concentrations at constant pressure near 40 km may have decreased by about 0.5% per year over the decade, in general agreement with that expected theoretically. However, data from ground-based measurements of upper stratospheric ozone show no significant trend over this period. There is also limited evidence for an increase in ozone in the troposphere, again, in agreement with calculations that included the effect of nitrogen oxides emitted from aircraft. Such findings are still tentative due to remaining observational and theoretical uncertainties, however, and do not warrant any strong conclusions at this time regarding present man-caused influences on the atmosphere.

Ozone in the Future

Because we cannot be sure which of many possible regulatory and economic actions will be taken, there are many possible scenarios for the future emissions of various chlorocarbons. As described earlier, the "standard" assumed scenario is that of constant emissions of the chlorocarbons at current levels. Many others have been developed to examine the impact of various actions. In general, scenarios with uncontrolled growth in chlorocarbon emissions result eventually in large (calculated) reductions in stratospheric ozone. For example, release of chlorocarbons at an exponentially growing rate, similar to that of the early 1970s could, within a

Fig. 4

Calculated change in ozone from 1971 to 1980 as a function of altitude for chlorocarbons only and for chlorocarbons plus other man-made (anthropogenic) source perturbations.

few decades, reduce stratospheric ozone by more than 10%. Scenarios in which only U.S. emissions are reduced show little effect on the overall calculated reduction in ozone. It is evident that controls would be effective only on an international basis.

The International Organization for Economic Cooperation and Development (OECD), of which the U.S. is a member, recently developed a series of 15 simplified general scenarios covering a broad range of future chlorocarbon emission rates.¹¹ These scenarios assume, variously, that emissions of all artificial chlorocarbons will grow, stay level, or decline at the same constant rate from 1980 to 2000, when a change to a different choice of constant rate can occur.

Figure 5 shows the calculated change in total ozone with time for three of these 15 scenarios.¹² Scenario VIII is similar to the standard scenario, except that it includes more chlorocarbon species and its emission rates for CFC-11 and CFC-12 are about 10% lower (1980

levels rather than 1976 levels). This scenario at steady state results in a change in total ozone of -5.8% .

Scenario VII, which assumes a 7% increase in emissions per year after 2000, rapidly depletes the ozone layer.

Scenario IX, on the other hand, reduces emissions by 7% per year beginning in the year 2000. In this scenario, total ozone slowly recovers, returning to the 1980 level in the year 2090.

The maximum calculated decrease in ozone for scenario IX occurs about 15 years after the decline in emissions began. The lag in recovery (the increase in ozone destruction over this time interval) results from the existing chlorocarbon reservoir in the troposphere at the time that emissions began to decline. Calculations that begin the decline in emissions in 1990 rather than 2000 do not reduce ozone as much, showing a maximum difference of 0.5% change in total ozone compared to scenario IX at any given time.

Figure 6 outlines the possible combined impact of chlorocarbons plus

Fig. 5

Changes in total ozone as a function of time for scenarios VII, VIII, and IX relative to a background atmosphere that is free of chlorocarbons.

Fig. 6

Calculated changes in total ozone as a function of time from 1970 relative to a background atmosphere free of chlorocarbons for scenarios involving various combinations of man-caused perturbations.

It is evident that controls [of chlorocarbon emissions] would be effective only on an international basis.

other man-caused influences.¹⁰ In this case, we included scenarios for possible growth in emissions for nitrogen oxides (NO_x from aircraft), carbon dioxide (CO_2), and nitrous oxide (N_2O) in addition to scenario VIII for chlorocarbon emissions.

Although calculations including future chlorocarbon emissions can alone result in significant reductions in ozone, there is very little change in total ozone over the coming decades when other man-caused influences are included. Increasing CO_2 concentration has the largest offsetting effect on the change in total ozone due to chlorocarbons.

Significant changes in the ozone concentration as a function of altitude are still found, even though the change in total ozone is less than 1%. The changes in ozone with altitude would have potentially significant climatic implications.

Chemical uncertainties could affect the magnitude and relative importance of various man-made sources. Recognized uncertainties concerning modeling techniques seem unlikely to alter the calculated changes in total ozone for the combined scenarios by more than a few percent. The uncertainties regarding the choice of scenarios for future emissions of the artificial sources seem much more significant.

Summary

Nearly a decade of research and data-gathering has produced a much clearer understanding of the complex processes responsible for maintaining the ultraviolet-absorbing ozone layer in the upper stratosphere. We have developed a one-dimensional atmospheric model that includes 141 chemical reactions in its estimates of the concentrations of 47 different chemical species. Statistical analyses of the measured changes in atmospheric ozone indicate that these changes are much smaller than the model calculations lead us to expect when only the effects of chlorocarbons are considered. When we add in the effects of aircraft emissions and carbon dioxide from combustion, our calculated ozone concentrations agree

much more closely with the available measurements. ■

Key Words: atmosphere; chlorocarbons; ozone; stratosphere; supersonic aircraft; troposphere.

Notes and References

1. See for example: National Research Council, *Halocarbons: Effects on Atmospheric Ozone*, National Academy of Sciences, Washington, D.C. (1976); National Research Council, *Stratospheric Ozone Depletion by Halocarbons: Chemistry and Transport*, National Academy of Sciences, Washington, D.C. (1979); R. D. Hudson and E. I. Reed, Eds., *The Stratosphere: Present and Future*, NASA Reference Publication 1049 (1979); R. D. Hudson, Ed., *The Stratosphere 1981: Theory and Measurements*, World Meteorological Organization, Global Ozone Research and Monitoring Project, Rept. No. 11 (1981); Coordinating Committee on the Ozone Layer, *Report of the Fourth Session*, United Nations Environment Programme, UNEP/CCOL/4/7 (1980).
2. J. S. Chang and D. J. Wuebbles, "One-Dimensional Coupled Transport and Chemical Kinetics Models of the Stratosphere," in *Proc. NATO Advanced Study Institute on Stratospheric Ozone*, Albufeira, Portugal, October 1-13, 1979.
3. See for example: J. S. Chang, A. C. Hindmarsh, and N. K. Madsen, "Simulation of Chemical Kinetics Transport in the Stratosphere," in *Stiff Differential Systems*, R. A. Willoughby, Ed. (Plenum Press, New York, 1974), pp. 51-65; F. M. Luther, J. S. Chang, W. H. Duewer, J. E. Penner, R. L. Tarp, and D. J. Wuebbles, "Potential Environmental Effects of Aircraft Emissions," U.S. Department of Transportation, Rept. No. FAA-EE-79-23 (1979); and D. J. Wuebbles, *The LLNL One-Dimensional Transport-Kinetics Model of the Troposphere and Stratosphere: 1981*, Lawrence Livermore National Laboratory, Rept. UCID-19185 (1981).
4. M. J. Molina and F. S. Rowland, "Stratospheric Sink for Chlorofluoromethanes: Chlorine Atom Catalyzed Destruction of Ozone," *Nature*, **249**, 810 (1974).
5. D. J. Wuebbles and J. S. Chang, "A Study of the Effectiveness of the ClX Catalytic Ozone Loss Mechanisms," *J. Geophys. Res.*, **86**, 9869 (1981).
6. G. C. Reinsel, "Analysis of Total Ozone Data for the Detection of Recent Trends and the Effects of Nuclear Testing During the 1960s," *Geophys. Res. Lett.*, **8**, 1227 (1981).
7. D. S. St. John, S. P. Bailey, W. H. Fellner, J. M. Minor, and R. D. Snee, "Time Series Search for Trend in Total Ozone Measurements," *J. Geophys. Res.*, **86**, 7299 (1981).
8. P. Bloomfield, M. L. Thompson, G. S. Watson, and S. Zeger, as cited in *The Stratosphere: Present and Future*, R. D. Hudson, Ed., World Meteorological Organization, Global Ozone Research and Monitoring Project, Rept. No. 11 (1981).
9. J. S. Chang, W. H. Duewer, and D. J. Wuebbles, "The Atmospheric Nuclear Tests of the 1950s and 1960s: A Possible Test of Ozone Depletion Theories," *J. Geophys. Res.*, **84**, 1755 (1979).
10. D. J. Wuebbles, F. M. Luther, and J. E. Penner, *Effect of Coupled Anthropogenic Perturbations on Stratospheric Ozone*, Lawrence Livermore National Laboratory, Rept. UCRL-86870 (1982).
11. *Emissions Scenarios for CFCs*, Organization for Economic Cooperation and Development, Rept. ENV(81)14 (1981).
12. D. J. Wuebbles, *Chlorocarbon Emission Scenarios: Potential Impact on Stratospheric Ozone*, Lawrence Livermore National Laboratory, Rept. UCID-19171 (1981).

Nuclear Waste Storage: Ev

We now have a systematic method for evaluating the uncertainties and sensitivities involved in predicting the performance of a nuclear waste repository.

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The national strategy for disposing of nuclear waste is to encapsulate it in long-lived packages and to isolate it in underground repositories such as that depicted in Fig. 1. Given this method of waste management, our task was to develop a tool for determining the uncertainty in our predictions of the effectiveness of such isolation.

Nuclear waste disposal schemes necessarily involve relatively untried and unproven technology for three main reasons: nothing quite like this has been done before, the time scales over which nuclear waste must remain isolated are long (tens of thousands of

years), and the medium in which the waste is to be emplaced (the underground) is extremely variable. Hence, every scheme proposed must be evaluated theoretically on the basis of the best information available, some of which contains considerable uncertainties.

In such a situation, it is important to identify the principal parameters that govern movement of waste from the repository and to establish the range of uncertainties associated with these parameters. This information is essential for estimating the uncertainty in our performance estimates. It also

luating the Uncertainties

enables us to decide which parameters must be investigated more thoroughly and which are already known accurately enough.

One common way to define the effectiveness of nuclear waste isolation is to estimate the amount of radiation exposure the population is likely to acquire as a result of potential leaks into the biosphere. This definition involves a large number of untestable assumptions about future human uptake of radioactive materials from the environment. For example, who can say today the extent to which it may become possible, a thousand years from now, to remove dissolved radioactive materials from water?

To avoid dealing with such assumptions, we chose instead to define isolation effectiveness in terms of the peak radioactivity release rate from the repository into the biosphere. Since these repositories are underground, this

calls for an analysis of nuclide migration via groundwater flow. Therefore, our principal objectives were:

- To establish models for predicting nuclide migration and the underground flow of water.
- To determine the major parameters controlling underground water flow and nuclide migration.
- To establish a method for quantifying and calculating the uncertainties in the release rate of potentially hazardous nuclides.
- To determine the importance, to the final result, of each of the various subsystems and parameters that influence the peak radioactivity release rate.

Approach to a Solution

Our first step in accomplishing these objectives was to describe a generic nuclear waste isolation system in terms of a series of subsystems and modeling parameters. Figure 1 shows the three

Fig. 1

Pictorial representation of a nuclear waste repository showing its three subsystems used in our modeling (the waste form and package, the repository and nearby site, and the regional aquifer).

subsystems we used: the waste form and package, the repository and nearby site, and the regional aquifer (underground water-permeable layer).

The next effort consisted of obtaining and interpreting raw data on waste forms and geologic media (mostly from literature surveys and responses from experts), incorporating this information into the system models, and, finally, determining representative values of the uncertainties in the various input parameters that make up the system models. For the most part, we chose ranges of the parameters appropriate to a repository in salt. We used a log-normal distribution in estimating uncertainties because it reasonably represents the data (which usually covered several orders of magnitude).

After compiling the data, we performed deterministic and probabilistic parametric analyses of the system models. The deterministic calculations were straightforward and direct applications of the system models. The probabilistic calculations determined uncertainties in the radioactivity release rates using the same models. In the latter calculations, we represented each distribution by a set of discrete ele-

ments and combined the probabilities representative of each element to form the overall distribution of probabilities.

In performing these calculations, we used the MACRO2 code (see box on p. 25). In reviewing the tentative conclusions we have drawn from these calculations, it is important to keep in mind the limitations of this study, which was based on simple models and generic data (specific to no particular site or region) and was conducted mainly to demonstrate the usefulness of the method. Refined models and specific data should produce correspondingly more definitive results with little additional effort.

Subsystem Models

To avoid making assumptions about how radioactive materials in the environment find their way into the human body, we based our definition of system performance on the peak rate of radioactive release from the repository (measured in curies per year). When the repository is first filled, the radioactivity contained is a maximum but the leakage is zero. Hence, the release rate (the product of radioactivity and leakage rate) is also zero. After a time, in a failure scenario involving corrosion or dissolution of the waste container, leakage begins and grows progressively worse up to a maximum rate. As the quantity of radioactive material in the repository declines, the leakage rate will decline.

MACRO2 Overview

In everyday affairs, the wisest course usually is to avoid the obvious hazards and not worry too much about remote possibilities. Sometimes, however, as in planning for nuclear waste management, we must know the probabilities of even very unlikely events and determine which components of this probability are relatively unimportant and which have a large effect on the final outcome.

The MACRO2 computer program enables us to organize the task and complete the computation in a few minutes on the Cray-1 computer. MACRO2 is not specific to the waste-management problem but could be used to perform both sensitivity and uncertainty analysis on the collection of deterministic models that characterize any system. It contains algorithms that automate simple sensitivity analyses and govern the generation, propagation, listing, and plotting of probability distributions through a string of successive models.

The usual way to perform an uncertainty analysis by computer is with a Monte Carlo program, essentially a gambler's approach that runs the same calculation over and over with random variation in the input parameters to obtain a probable distribution of the outcome. This approach works well in determining probabilities in the midrange, e.g., deciding between a probability of 20% or 10% for a particular effect.

In waste management, however, we need to know the whole range. Although we are interested in midrange

probabilities, we also need to quantify the remote possibilities—to decide whether a probability is one in a hundred or one in a thousand. The usual Monte Carlo approach is slow in providing such answers, simply because it spends most of its time on the common or midrange possibilities and seldom hits one of the very unlikely events.

In our new approach with the MACRO codes (MACRO1 and MACRO2), we break the probability function for each component effect into a finite number of slices, or bins, which together constitute a good discretized approximation of the true probability distribution. We then direct the computation to spend an equal amount of time on each of the bins. Thus, we avoid lavishing time on the midrange of the distribution and gain time for the fringes.

MACRO codes operate on these discretized approximations for each of a set of component effects to arrive at an overall probability distribution for successively more general effects.¹ The number of bins in one of these composite probability distributions is the product of the number of bins in each of the component distributions.

When the number of bins becomes unwieldy, it is possible to rediscretize the new probability distribution. Although this somewhat degrades the degree of approximation, it shortens considerably the time required for the calculation.

Another factor that can reduce the release rate is radioactive decay. The radioactivity is highest during the first few hundred years and decreases exponentially with time. Once the leakage reaches its maximum rate, radioactive decay and the exhaustion of the repository stock can only reduce the rate of radioactivity release. Hence, the rate of release passes through a maximum, and it is this peak release rate that serves as a measure of system performance.

Yet another factor influencing the release rate of a particular isotope is its chemistry, in terms of both the rate at which it leaches out of the waste form and the rate at which it moves in an aquifer. In theory, it might be necessary to calculate a separate release rate for each isotope. In practice, we were able

to group isotopes with similar chemistry and calculate release rates for only three groups of nuclides; actinides, halogen fission products (technetium-99 and iodine-129), and other fission products.

As waste forms, we considered both spent fuel and high-level waste, using typical reactor parameters in calculating the amounts of the various nuclides present.² We assumed that the spent fuel would be enriched uranium (3.2% uranium-235) that had been exposed for 1100 days at an average reactor power of 30 MW/MT. For the plutonium-storage fuel cycle, we assumed that the high-level waste was derived from fuel reprocessed 160 days after removal from the reactor. We assumed that reprocessing removed all the krypton and xenon, 99.9% of the

... we based our definition of system performance on the peak rate of radioactive release from the repository.

iodine, and 99.5% of the uranium and plutonium in the spent fuel. The waste was also taken to include core structural materials such as cladding and spacers, and nitrogen impurities in the fuel. Table 1 lists the typical radioactive constituents of high-level waste.

For definiteness, we chose an inventory of radioactive waste (appropriate to a typical 10-year-old, medium-size repository) equal to that resulting from the generation of a million megawatt years of electricity, or 1 TWe·y. The total radioactivity in such an inventory amounts to some five billion curies. For simplicity and to reduce the problem to manageable proportions, we also made a number of simplifying assumptions. Any one of them could be refined or expanded as needed or as additional information becomes available. For example, we assumed that this inventory is stored in a single point rather than spread over several square kilometres, leaving to future studies the task of investigating the effects of a more realistic distribution.

In our calculation, we also assumed that the leakage rate from the waste form and package had already reached a constant or equilibrium value governed by two parameters, the release initiation time (T_0) and the release duration (T_D), also called the dissolution or leach time. The release initiation time includes the age of the waste at the time it was buried, the time for the repository to become resaturated and repressurized, and the canister life (corrosion time). Both T_0 and T_D implicitly include the possible effects of changes in temperature or water chemistry.

Water flow underground is governed by Darcy's Law,³ whose input parameters are pressure difference (head), length (or pressure gradient), and hydraulic conductivity and whose output is the Darcy velocity. By dividing the Darcy velocity by the effective porosity, we can obtain the pore water velocity. We assume that the transport of nuclides is governed by the advection-diffusion equation⁴ over one-dimensional pathways with longitudinal dispersion.⁵ We further assume that

Table 1 Inventory of radionuclides in high-level waste (4500-MCi activity, 10 y after removal from reactor core). Nuclides with activities below 0.01 MCi have been omitted.

Radionuclide	Activity, MCi				
	100 y	1000 y	10 000 y	100 000 y	1 000 000 y
²⁴³ Am	0.55	0.51	0.23	—	—
²⁴¹ Am	4.26	1.02	—	—	—
²³⁸ Pu	1.42	0.01	—	—	—
²³⁷ Np	0.01	0.01	0.01	0.01	0.01
¹⁵³ Eu	2.69	—	—	—	—
¹⁵¹ Sm	17.3	0.01	—	—	—
¹²⁶ Sn	0.02	0.02	0.02	0.01	—
⁹⁹ Tc	0.45	0.45	0.43	0.32	0.02
⁹⁰ Sr	207	—	—	—	—
⁷⁹ Se	0.01	0.01	0.01	—	—

we can apply the same retardation factor for all nuclides in the same decay chain.

To solve the nuclide-transport equation, we use Green's function with semi-infinite boundary conditions,⁶ using the pore water velocity, retardation factor, dispersion constant, and path length as input parameters. We compute the flux of waste by integrating with respect to time over the path function (Green's function) and the dissolution function (which is assumed to be constant). Then, we multiply by the waste's activity (which depends on time and on the nuclide, group) to determine how much radioactivity this flux represents. The maximum over time of this product is our measure of system performance.

The mean transit time T_T for a pulse of waste to traverse a path of length L with pore water velocity v and retardation factor B is simply:

$$T_T = LB/v.$$

The time spread caused by longitudinal dispersivity is the square root of the second central moment:

$$S = T_T \sqrt{2\alpha/L},$$

where α is the dispersivity length. For serially connected pathways (such as the repository site and the regional aquifer), the total transit time is the sum of the individual transit times, and the square of the total spread is the sum of the squares of the individual spreads. Figure 2 shows how these calculations tie the various subsystem models together.

Parametric Analysis

The purpose of this analysis was to find out which parameters are dominant in determining the performance of a repository. We believe that a global

Fig. 2

Flow chart illustrating the process of calculating repository performance (defined as peak activity release rate).

parametric analysis with a few high-level (lumped) parameters, a "bird's-eye view of the terrain," is more informative than a complex array of local sensitivity analyses, which can be like getting acquainted with a forest by studying individual trees. Table 2 lists the parameters we used and their ranges.

The contour plots in Fig. 3 present the deterministic results of the analysis. Figure 3a, which is for actinides from spent fuel with little dispersion (a T_T/S ratio of 10), shows that the peak activity release rate depends only on the transit time (and is independent of the dissolution time) when the transit time is larger than the dissolution time (gold area).

A comparison Figs. 3a and 3b (e.g., the contour for 10 Ci/y marked in maroon) shows that the actinide activity release for spent fuel is about ten times larger than that for high-level waste. This is to be expected because most of the uranium and plutonium have been removed from the high-level waste. Our analysis verifies and quantifies this expectation.

To put some of these numbers into perspective, imagine a repository that releases mixed nuclides at a constant rate of 100 Ci/y into a river flowing at 1000 km³/y. (Such a flow is typical for the Colorado River.) The resulting concentration of radionuclides in the river water would be 100 aCi/cm³, less than a thousandth of the natural activity of the earth's surface.⁷

A comparison of Fig. 3a with 3c shows that varying the time-to-spread ratio from 10 to 1 has little effect on the peak activity rate contours. For a 10-km aquifer, this corresponds to varying the longitudinal dispersivity from 50 m to 5 km. Hence, longitudinal dispersivity is relatively unimportant in determining repository performance. This is a welcome indication since dispersivity is quite difficult to determine in the field.

General conclusions that can be drawn from the results of our deterministic parametric analysis include:

- Performance (peak activity rate) is usually more sensitive to the larger of the transit time or the dissolution time.
- Actinide release rate is greater for spent fuel than for high-level waste.
- Dispersivity has little influence on performance.

We also performed a probabilistic parametric analysis for the same parameters, with an assumed

Table 2 Parameters and their ranges used in our parametric analysis of a hypothetical nuclear waste repository.

Transit time (T)	10^3 – 10^6 y
Dissolution time (T_D)	10^3 – 10^6 y
Release initiation time (T_0)	500 y
Transit time/spread (T/S)	1, 10
Inventory type	Spent fuel, high-level waste
Inventory amount	1 TWe · y
Nuclide groups	Technetium and iodine; actinides; other fission products

hundredfold uncertainty in both the transit and the dissolution times. The result confirmed the conclusions drawn from the deterministic analysis.

Subsystem Analysis

As a further application of our model, we performed a probabilistic consequence analysis of the system and a failure scenario. The failure scenario assumes that water flows into the repository from an underlying aquifer and out again through a fracture zone, associated with a sealed shaft and tunnel, to an overlying aquifer that leads to the biosphere. The purpose of this analysis is to determine how each subsystem (waste form and package, repository site, and regional aquifer) contributes to the uncertainty in the system performance and which ones have the most influence in determining it. The values of the parameters that characterize these pathways and the waste form and package appear in Table 3.

Hence, longitudinal dispersivity is relatively unimportant in determining repository performance.

Fig. 3

Peak activity release rates for actinides as a function of transit time and dissolution time. (a) Repository filled with spent fuel, assuming a time vs spread ratio (τ_T/s) of 10. (b) Repository filled with high-level waste, assuming the same τ_T/s ratio of 10. (c) Repository filled with spent fuel, assuming a τ_T/s ratio of 1.

Table 3 Median values of the subsystem parameters used in our analysis of the performance of a nuclear waste repository.

Waste form and package	
Inventory type	Spent fuel
Inventory amount	1 TWe.y ^a
Release initiation time	500 y
Dissolution time: technetium and iodine, and fission products	5000 y
Dissolution time: actinides	10 ⁵ y
Repository site — fracture zone below repository	
Length	300 m ^a
Area	10 ⁴ m ²
Hydraulic conductivity	0.1 m/y
Repository site — fracture zone associated with shaft and tunnel	
Length	1200 m ^a
Area	100 m ²
Hydraulic conductivity	1 m/y
Longitudinal dispersivity	50 m
Effective porosity	0.001
Retardation factor: technetium and iodine, and fission products	10
Retardation factor: actinides	100
Regional aquifer	
Length	10 km ^a
Water velocity	2 m/y
Longitudinal dispersivity	2000 m
Retardation factor: technetium and iodine, and fission products	100
Retardation factor: actinides	1000
Head difference between upper and lower aquifers	10 m ^a

^aGiven, no uncertainty.

We analyzed each subsystem by computing its performance with probabilistic input parameters, most of which were assumed to range a factor of ten above and below the median values given in Table 3. We set each of the stipulated parameters in turn to its median value, let the rest of the parameters vary over their ranges, and computed the resulting variation in the subsystem's performance. Figure 4 shows how we condense the results into a set of bar graphs that project the probability distributions for the peak activity release rate.

Figure 5 represents the performance of the aquifer, based on the assumption that the whole inventory of the repository (all 2000 MCI) is instantaneously released at the input of the aquifer 50 years after being removed from the reactors. The results show that allowing the dispersivity to range from 400 m to 10 km has little effect on the performance of this 10-km aquifer.

Figure 6 compares the performances of the subsystems and the total system. Comparing cases 1, 2, and 3 with case 4, the combined system, shows that for the fission products and actinides the regional aquifer is the dominant (most influential) subsystem.

Figure 7 gives the results of an uncertainty sensitivity analysis for the complete system. Only the most influential parameters from each system were included here. The results show that the uncertainty in performance of the complete system is most sensitive to the uncertainty parameters in the dominant subsystem. This consistency bolsters our confidence in this method for identifying important subsystems and parameters. For the case considered, reducing the uncertainty in the water velocity in the aquifer will significantly reduce the uncertainty in the overall performance result. Information of this sort could be valuable in developing site-characterization plans.

The methods described above enable us to draw the following conclusions for the hypothetical system studied:

- Uncertainty in dispersivity has little effect on the uncertainty in the performance of the complete system.
- Subsystems seldom act as equal barriers; there is usually one dominant subsystem.
- The overall uncertainty in system performance is most sensitive to the uncertainty in the parameters of the dominant subsystem. These are: the water velocity in the aquifer for all nuclides, the waste dissolution time for technetium and iodine, and the retardation in the aquifer for actinides and fission products.

It follows from the last conclusions that the most effective way to improve overall system performance or reduce its uncertainty is to concentrate research and design effort on the dominant subsystem. It is important to note, however, that there are situations in

Fig. 5 Uncertainty sensitivity analysis of the regional aquifer subsystem, indicating the range of the peak release-rate results for two of the three radionuclide groups (technetium-iodine and other fission products) for several different cases. Restricting dispersivity to a single value has little or no effect on the overall uncertainty, suggesting that dispersivity is less influential than other factors in determining the peak release rate.

Fig. 6

Comparison of the effectiveness of separate subsystems in determining the peak release-rate results for two of the radionuclide groups (actinides and fission products other than technetium and iodine). All the bars for subsystem groups that include the regional aquifer are about the same, clearly indicating that this is the dominant subsystem.

Fig. 7

Uncertainty sensitivity analysis for the complete system, with various factors held constant and the rest allowed to vary. As in Fig. 6, this figure presents the peak release-rate results for two of the radionuclide groups (actinides and fission products other than technetium and iodine).

which there is no dominant subsystem (e.g., a well bypasses the aquifer). It should also be kept in mind that this study examined only one scenario and made no attempt to determine the probability that such a scenario would take place.

Summary

Using simple models and typical data, we have developed a method for using uncertainty analysis in directing waste-isolation research and development. Figure 8 indicates schematically how this use of uncertainty analysis could be applied. Our work was concentrated in the areas represented by the colored boxes.

From our subsystem study, we can say that the most effective way to enhance the overall system performance or reduce the uncertainties associated with it is to make improvements in the dominant subsystem. From the parametric analysis and the subsystem study, we see that longitudinal dispersivity has less influence than most of the other parameters considered.

One way to demonstrate the utility of our methods would be to repeat the analysis using real data representing specific sites and repository designs. Such an analysis should include quantitative input elicited from experts on the failure modes, models, and probability distributions that characterize the specific system. The study should establish the quantitative effects of assumptions built into the system models. Finally, the sensitivity and uncertainty analyses should be integrated into a decision-analysis framework, including an estimate of the impact of further research and development. ■

Key Words: MACRO2; mathematical modeling; nuclear waste; nuclear waste disposal; repositories; sensitivity analysis; uncertainty analysis; underground storage.

Fig. 8

Flow diagram illustrating the use of uncertainty analysis in making waste-isolation decisions. Colored blocks indicate those sections of the overall analysis involved in our development work.

Notes and References

1. L. L. Edwards, *MACRO1: A Code to Test a Methodology for Analyzing Nuclear-Waste Management Systems*, Lawrence Livermore National Laboratory, Livermore, CA, Rept. UCRL-52736 (1979).
2. C. W. Alexander et al., *Projections for Spent Fuel to be Discharged by the U.S. Nuclear Power Industry*, Oak Ridge National Laboratory, Oak Ridge, TN, Rept. ORNL/TM-6008 (1977).
3. J. M. DeWiest, Ed. *Flow Through Porous Media*, (Academic Press, NY, 1969), Ch. 1.
4. B. L. Berman et al., *Analysis of Some Nuclear Waste Management Options*, Lawrence Livermore National Laboratory, Livermore, CA, Rept. UCRL-13917 (1978) Vol. II, Appendix F.
5. J. Bear, *Hydraulics of Ground Water* (McGraw-Hill, New York, 1979), Ch. 7.
6. S. G. Oston, *Finite Medium. Green's Function Solutions to Nuclide Transport in Porous Media*, Lawrence Livermore National Laboratory, Livermore, CA, Rept. UCRL-15040 (1979).
7. M. Eisenbad, *Environmental Radioactivity*, (Academic Press, New York, 1973), Ch. 7.

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