

# FACT SHEETS

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## CENTER FOR HUMAN RADIOBIOLOGY

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## CENTER FOR HUMAN RADIOBIOLOGY

## Fact Sheet on

Theory of the Induction of Bone Cancer by Radiation

The purpose of this work is to provide a logical synthesis of all the data concerning the induction of osteosarcoma by radiation in man, animals and cell culture in terms of cellular, molecular and systemic mechanisms. We focus upon osteosarcoma and radiation for two reasons: (1) They are of prime interest to the Center in its effort to learn from the human radium cases how to derive solidly-based predictions for the risk to man at any dose level following any kind of exposure from internal emitters, such as strontium, plutonium, thorium, or uranium, and (2) bone cancer as induced by alpha or beta radiation appears to be one of the best defined problems in cancer induction. The dosimetry is excellent; there is a wealth of data in mice, rats, dogs and man for both long term and short term, long range and short range ionizing radiation; the cells at risk have been identified; and bone cancer has a very low natural incidence with no known complications with other environmental carcinogens. These favorable conditions, plus powerful new computer techniques, plus basic research on mechanisms, plus the crucial human data, lead us to believe that the Center for Human Radiobiology may be in a position to produce such a synthesis.

The second step in this plan, A Theory of the Induction of Bone Cancer by Alpha Radiation, has just been published (Radiation Research 71, 149-192, 1977 - the July issue) and reprints are available. This paper shows that two initiation steps, plus one promotion step in a single cell at bone surface can, together with cell killing, provide a good fit to both the dose distribution and the time distributions of osteosarcomas in man and in dog. (The first step in the plan was the synthesis of metabolic data for man which provided the internal dosimetry for the bone cancer theory (Alkaline Earth Metabolism in Adult Man, Health Physics 24, 125-221 (1973), ICRP Publication 20)).

The cellular model has just been related to a promising new molecular model which involves direct action of alpha particles upon DNA. This DNA model may provide the desired link to low doses, low dose rates, and low LET radiation.

Recent progress (1 person-year)

The magnitudes of the linear and the quadratic terms of the dose-response of the Marshall-Groer model of cancer induction have been calculated assuming that the two initiation events correspond to double strand disruption of each of two DNA molecules within the cell nucleus. This calculation is being published in the Alta Symposium volume of the SIMS organization soon to come out. This promising mechanism leads to a new expression for dose response which is linear below a dose  $\eta$  (the order of 40 rads) square between dose  $\eta$  and the mean lethal dose for a single cell (the order of 100 rads), and which reaches a constant plateau value for all doses much in excess of the mean lethal dose for a cell. The response of the model is a tumor rate - the number of tumors expected per unit time per number of cases then at risk. This tumor rate is multiplied by

the number of cases at risk at each endosteal dose and each time before it is compared to data. Therefore, the comparison between model and data is three-dimensional (tumor rate, dose rate, and time) and corrects for competing risks of death.

So far, the model has been found to fit well the data for

- 1) Ra-226 and Ra-228 in man (CHR data)
- 2) Ra-226 in beagle dog (Salt Lake Radiobiology Laboratory data)
- 3) Ra-224 in man (the German cases of Spiess and Mays)

Both (1) and (3) appear to require that the endosteal dose be an order of magnitude less than current estimates on the basis of simple dosimetry for cells 0-10  $\mu\text{m}$  from bone surface. These results may indicate that

- a) the cell at risk is some 25  $\mu\text{m}$  off bone surface, rather than 0-10, OR
- b) the diffuse component is the effective component for Ra-226 AND there is a cleared layer of bone about 5  $\mu\text{m}$  wide on most long term bone surfaces,
- c) there is considerable escape of thoron and not much preference for radium at the bone surfaces of the German Ra-224 cases, OR
- d) the mean lethal dose for cells from alpha particles is much greater in vivo than in vitro. Recent work on DNA repair makes this a possibility, but there is as yet no other evidence.

The Ra-224 data for all age groups analyzed together indicate that the promotion rate,  $\lambda$ , may be as large as 10-15%/year. This contrasts with the value of 1-2%/year found for the high dose Ra-226 cases. Inhibition of resorption in the latter cases is a distinct possibility. The Ra-224 cases are now being reanalyzed according to age.

4) The fourth set of data fitted to the model has been the Ra-224 mouse data from Neuherberg Germany (Luz, Hug, et al.). A good fit was obtained only if the time for tumor growth was allowed to vary a factor of ten inversely to the time of protraction of a given dose; a long-continued irradiation caused the tumors to grow more rapidly than an absence of continuing radiation. This is reminiscent of Blum's work on ultraviolet skin tumors in mice, and Finkel's observations of fast tumor growth for Sr-90 in mice contrasted with slow tumor growth following Y-90 (half-life a few days) using serial x-rays of the tumors. No serial x-rays were made at Neuherberg, but Mays at Salt Lake has been alerted to take serial x-rays of the new Ra-224 beagles to look for this effect. It may be that a distribution of tumor growth times dependent on continuing radiation will be necessary for the short-lived mouse even if it is not necessary in the dog or in man. If this effect is confirmed, it may be the first evidence we have seen that the immune response has anything to do with bone tumors. The effect would be only upon tumor growth time,  $g$ , and so far only in mice.

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Fact Sheet on  
Tumor Mechanisms

Mechanistic models are needed to extrapolate dose-response relationships of radium and plutonium to low doses. This involves studies on the interrelationships of radiation and the immune responses using direct measurements on blood samples from the radium patients as well as studies of cells in vitro.

Oncogenic Transformations in Vitro (2.5 person-years)

We have shown that cells from the C3H 10T1/2 cell line, obtained from a mouse embryo, become malignantly transformed with a dose-dependent frequency when irradiated by 5.6 MeV alpha particles.<sup>(1)</sup> This radiation is similar to that emitted from plutonium-239. A Tandem Van de Graaff machine which provides a beam of ionizing particles, well defined in geometry and energy, is being used as a probe to study the mechanisms by which cells in vitro are killed or transformed to become malignant. Dose-response relationships are being generated as a function of energy and cell geometry. The dimensions of the nuclei of cells lining human bone surfaces are being determined by light and electron microscopy.<sup>(2)</sup> These results, together with our in vitro studies, suggest that the flattened cells adhering to bone collagen may be the most important in the production of bone tumors. The malignant potential of  $\alpha$ -particle irradiation is being compared with the most common carcinogenic product of combustion, benzo-a-pyrene (BP), in the same in vitro system. The results, so far, indicate similar morphological transformation for the two agents but a higher frequency of transformation with BP.<sup>(3)</sup> A dose response for BP is being generated. In addition, normal cells and analogues of Vitamin A are being used in attempts to inhibit transformations.<sup>(3,4)</sup> Our results indicate that under certain conditions, normal cells can inhibit the formation of transformed foci.

Immunocompetence of Radium Dial Painters (1.5 person-years)

The immunocompetence of dial painters who had been exposed to intake of  $^{226}\text{Radium}$  is under study with the following immunological assays:

- 1) Mitogen-induced lymphocyte DNA synthesis of peripheral blood lymphocytes, specific for detecting the competence of both cellular and humoral immunity;<sup>(5)</sup>
- 2) Immunofluorescence studies for the detection of tumor specific antibodies in the sera of these patients using specific antisera and osteosarcoma cell lines;<sup>(6)</sup> and
- 3) Functional chemotactic assays to determine the immunocompetence of monocytes and neutrophils. The purpose is to determine whether signs of immune deficiencies in these patients can be observed before clinical diagnosis of tumor.

## References

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## CENTER FOR HUMAN RADIOBIOLOGY

## Fact Sheet on

Metabolic Studies of Internally Deposited Radionuclides:I. Measurements In VivoRadium (2.5 person-years)

i) The body content of  $^{226}\text{Ra}$  is determined by counting the 1.76 MeV  $\gamma$ -rays emitted by  $^{214}\text{Bi}$  (RaC). Measurements are made with large NaI(Tl) detectors in underground shielded rooms. The patient either lies on a curved bed of 1.5 m radius or sits in a reclining chair. Thirty-six cases have been counted in both geometries in order to calibrate the latter. The minimum detectable amount ( $3\sigma$ ) of radium in the chair geometry is 4.7 nCi, which corresponds to an intake of 1.43  $\mu\text{Ci}$  to the blood fifty years previously. Sequential measurements are made in order to determine the retention at long times after exposure.

ii) There are about 200 women employed as dial workers prior to 1930 who have been unwilling or unable to come to CHR for a body radioactivity measurement. About half have indicated that they would not object to a measurement near their home. To perform these measurements, a mobile whole body counter has been prepared. The counter consists of an NaI(Tl) detector in a shadow-shielded geometry mounted in a semitrailer. Field trips have been made to Ottawa, IL, Orange, NJ, and Waterbury, CT. A total of 16 of these previously unmeasured cases have been counted, of whom two had radium contents above the minimum detectable activity of 10 nCi (mobile counter).

Thorium (1 person-year)

A study population of 558 former workers at a thorium refinery has been defined on the basis of likely exposure. The chest contents of  $^{212}\text{Bi}$  (ThC) have been investigated for 81 cases (to Sept. 30, 1978), of whom 31 showed measurable amounts (limit of detection  $\sim 160$  pCi). Although almost all the cases have demonstrated measurable thoron exhalation, there is no significant correlation between the amount of emanating  $^{224}\text{Ra}$  (the parent of thoron) and the chest content of  $^{212}\text{Bi}$  for the 31 cases. Efforts are underway to analyze by least squares the  $\gamma$ -ray spectra, which contain contributions from other gamma-ray emitters in the decay chain, in order to obtain some information on the degree of radioactive equilibrium between inhaled  $^{232}\text{Th}$  and its daughters in vivo.

Transuranic elements (0.3 person-year)

i)  $^{239}\text{Pu}$ : An 18-cm diameter gas-filled proportional counter is used to detect the 17 keV uranium L x-rays which follow the  $\alpha$ -decay of plutonium. Calibration factors for inhaled plutonium have been determined from a simple phantom, and they were verified by the results of an IAEA-sponsored intercalibration experiment in which volunteers inhaled "mock plutonium," an aerosol labelled with  $^{103}\text{Pd}$ .

A method for determining skeletally-deposited plutonium by external counting at the skull has been developed. Calibration factors were obtained from measurements on the exhumed skulls of three plutonium injection cases. Radiochemical analysis has shown that the concentration of plutonium in the skull is approximately equal to the average concentration in the entire skeleton, and the skull-counting technique has a limit of detection of approximately 10 nCi in the whole skeleton, or one-fourth the maximum permissible skeletal burden (To 77).

ii)  $^{241}\text{Am}$ : The body content and distribution of  $^{241}\text{Am}$  have been investigated in eleven subjects. Nine had contents below the maximum permissible body burden of 50 nCi. In 1977 one subject contained some 900 nCi of an initial burden of about 1800 nCi inhaled in 1965. The distribution of activity in the skeleton was consistent with deposition on bone surfaces, and approximately 10% of the initial burden remained in the lungs (To 76). Another subject had swallowed two point sources of  $^{241}\text{Am}$  of the type used in domestic smoke detectors. The sources were recovered intact after passage through the GI tract, with more than 99% of their original activity. Since the exact amount of  $^{241}\text{Am}$  in the sources could be determined after their voiding, we were able to verify the calibration equation used for determining the activity in vivo (Ru 77).

iii)  $^{249}\text{Bk} - ^{249}\text{Cf}$ : The retention of  $^{249}\text{Cf}$  in the chest of a man who accidentally inhaled  $^{249}\text{Bk} - ^{249}\text{Cf}$  has been followed for 1630 days. A sum of two exponential functions provided a reasonable description of the chest content to 743 days (the longer-lived component had an effective half-life of about 1360 days), but the decline between then and 1630 days has been somewhat slower. The actual behavior must have been more complex because of the growth of  $^{249}\text{Cf}$  in vivo from the much larger amount of the parent  $^{249}\text{Bk}$ . Evidence is slowly accumulating for the entry of some  $^{249}\text{Cf}$  into bone.

#### Other radionuclides (0.3 person-year)

The low background in the underground counting rooms and consequent high sensitivity of the detectors enables us to follow the retention and distribution in vivo of injected radiopharmaceuticals for extended periods. Serial body radioactivity measurements of two ANL employees who were injected with  $^{75}\text{Se}$ -labelled selenomethionine in 1974 indicated an effective half-life of 62-64 days initially (corresponding biological half-life 130-140 days), but changing to 70 days (biological half-life 166 days) after about 200 days post-injection. Profile scans with a slit-collimated detector demonstrated the presence of radioactivity in the skeletal muscle as well as in a region from 30 to 60 cm from the vertex, which was presumed to include lung, pancreas, spleen and heart. This region contained 40-50% of the body content, while the lower legs contained about 10%. The distributions could be determined until about 500 days post-injection for one subject and until about 1100 days for the other. The distributions were quite similar and remained stable over the periods of observation.

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## CENTER FOR HUMAN RADIOBIOLOGY

## Fact Sheet on

Metabolic Studies of Internally Deposited Radionuclides:II. Measurements In VitroRadium (5 person-years)

Analysis of data on the concentrations of  $^{226}\text{Ra}$  in highly trabeculated bone (vertebra) and in cortical bone (femoral shaft), has demonstrated some agreement with the ICRP 20 metabolic model. Some evidence for a radiation effect on retention was obtained.

The results of measurements of  $^{226}\text{Ra}$  in excreta and diet have shown that at 40 to 50 y after intake the coefficient of excretion ranges from about 1% to 5% of the body content per year, averaging about 2%/y for former dial painters and about 1%/y for subjects who acquired radium by injection.

The concentrations in liver, adrenals, pancreas, lung, kidney, brain, thyroid and spleen from a subject with a terminal body content of 1120 nCi, were 4.9, 14.7, 15, 52, 76, 130, 210 and 940 pCi/kg, respectively. Concentrations in bone were two to three orders of magnitude higher. A detailed study was made of radium in an eye from this subject. The concentrations were about 3000 pCi/kg in the sclera, choroid and iris, but less than 1000 pCi/kg in the other structures.

Routine measurement of radon in the breath of subjects has been discontinued due to the high variabilities in exhalation rates, especially those associated with the digestive process (Ru 78), and similar postprandial changes have been observed in the exhalation of radon originally inhaled from the environment and dissolved in body fluids and tissues. It has been discovered that very high levels of radon (10–30 pCi/litre) may be found in houses which have unpaved crawl spaces (e.g. especially split-level houses). Average daughter-product concentrations in two houses tested have approached 0.01 WL.

Thorium (0.5 person-year)

Significant levels of thoron were found in the breath of most of 80 former workers in a thorium processing plant. The results, expressed as the equivalent amount of freely emanating  $^{224}\text{Ra}$  at the mouth of the subject, range up to about 160 pCi; the limit of detection is less than 1 pCi. Some persons who had lived for many years near the plant, but never worked there, showed levels of around 2 pCi.

Measurable levels of  $^{232}\text{Th}$  (by activation analysis) were found in lung and lymph node taken at autopsy from a former thorium worker who had no detectable body content of thorium daughter products by external counting. The significance of these levels has not yet been assessed.

### Transuranic Elements (1.5 person-years)

The burden and macrodistribution of plutonium in the skeleton and its concentration in certain soft tissues have been determined in the remains of an 18-yr old woman (40-010) who received plutonium by injection in 1945 and who died 518 days later. The skeletal burden was  $54 \pm 2\%$  of the amount injected. As expected, the concentrations in trabecular bone were much higher than those in cortical bones, but the bones of the skull and the clavicle are quite representative of the entire skeleton. The results may not be typical because the subject was suffering from Cushing's syndrome. The concentrations in soft tissue were a factor of about 100 lower than those in bone. The concentration in hair increased markedly with the distance from the scalp. The macrodistribution will be compared with that for a man (40-015) who received plutonium by injection in 1946 and whose remains were recently obtained.

An investigation is being made of the effect of the oxidation state of plutonium on absorption from the gastrointestinal tract (La 78). For mice that had been starved prior to intragastric administration of the plutonium solutions, the fractions absorbed were  $2.0 \pm 0.3 \times 10^{-3}$  for Pu(IV) and  $3.0 \pm 0.7 \times 10^{-3}$  for Pu(VI). The value for Pu(IV) is a factor of about 100 higher than those reported previously for the rat and the ICRP value for man; the value for Pu(VI) is about a factor of 10 lower than the one reported previously.

Urinary excretion rates of  $^{241}\text{Am}$  (40 to 100 fCi/day) have been determined for two individuals who received plutonium by injection and whose plutonium excretion rates (4.7 to 7.6 pCi/day) had been previously determined. The  $^{241}\text{Am}$  was formed in vivo from the decay of the  $^{241}\text{Pu}$  that was in the injected plutonium.

### Uranium mill tailings (1 person-year)

A study is being funded by the Nuclear Regulatory Commission to assess the impact on the human food chain of radionuclides from tailings piles of uranium ore processing mills in New Mexico. Samples of cow dung and grass collected from land irrigated with uranium mine water showed levels of  $^{226}\text{Ra}$  5 to 20 times those in samples from a control area. The concentrations of  $^{226}\text{Ra}$  in various soft tissues and bone from rabbits trapped near a tailings pile were about twice those in control samples, while cattle grazed within 2 km of the tailings pile had only slightly higher levels than controls.

The levels of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the exposed rabbits were not significantly greater than in control rabbits. The  $^{210}\text{Pb}$  levels in the exposed cattle were somewhat greater than in the controls, but the study is incomplete. Vegetables from this area had appreciably higher levels of  $^{210}\text{Pb}$  than did those from places such as New York City. The excess radiation dose to individuals consuming meat and vegetables exclusively from these sources, could approach the limit of 25 mrem/y set by the USEPA.

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## CENTER FOR HUMAN RADIOBIOLOGY

## Fact Sheet on

Progress of Medical StudiesRadium cases (5 person-years)

Medical examinations are continuing at about the same rate as before; six are ordinarily scheduled each week, and the times of examination are coordinated with times of measuring radium burden. Frequency of re-examination of radium patients is determined by the radium burden and the age of the patient; however, all persons under follow-up are contacted at least annually by telephone or letter with questionnaire. Since a major thrust of the program is correlation of pathologic changes with radium burdens, ascertainment of radium content is of prime importance and efforts are made to obtain the best feasible medical profile of each person in the study.

1. A few individuals remain too anxious to be willing to visit the Center, and a small number have declared their active hostility. The mobile unit has enabled us to obtain measurements of some of these persons in several parts of the country. Several persons have willed their bodies to the Center and the exhumation program has been opened up in the midwest; both of these activities benefit from the rapport that has been established between the study group and our junior staff, while the senior staff is trying to cement relations with physicians in the critical areas, along lines of the successful seminar last January for those in the Ottawa, Illinois, region.

2. In addition to an exhaustive medical history and physical examination, a full profile of biochemical analyses (SMA-24 and thyroid profile, plus others as indicated); and a full set of skeletal x rays including special views of mastoids and paranasal sinuses (total about 160  $\mu$ rad mean dose to marrow) on at least one examination. Careful audiograms are taken, including bone conduction, on account of the vulnerability of mastoids to radium damage.

The coding of pathologic states for computer retrieval, according to the standard nomenclature (SNOP), has been done on the 5400 cases on whom files exist in the Center, yielding over 128,000 entries. Most of the present work now consists in coding conditions found in hospital records that are being obtained, and in first examinations of new entries. About half of these files contain minimal information (this might arbitrarily be defined as less than 10 entries) and the remainder contain an average of 47, with apparently quite uniform recording of common conditions likely to be identified in a complete examination. It has been recognized that SNOP file is at least an excellent index to the records; our study is being continued to assess the probability that an existing condition will have been recorded in a given category of patients.

A file of tumor diagnoses is being compiled from the SNOP listings. The present file includes 2016 entries in 835 cases. In all cases where the entry appears to require verification (comprising 108 of the above cases), the original records are being studied. By keeping this tumor diagnosis file up to date it will be possible to retrieve current data on any subgroup of patients in the radium study.

Former thorium workers (3 person-years)

Medical and radioactivity examinations are planned for over 400 living persons (all males) who worked at a thorium refining plant for one year or longer. Measurements of body radioactivity and thoron exhalation show a considerable difference between individuals and reasonably good reproducibility between measurements on individuals made at different times.

The in vivo measurements that are made are not yet translatable into body and organ thorium content, therefore each person at the time of examination is advised of the willed body program with a frank explanation of the scientific reasons for it. Several individuals have indicated willingness to participate.

The same medical and laboratory examinations are given as in the radium study, with the exception of detailed audiometry and the x-ray survey, which in thorium studies is limited to chest, abdomen, and any other films that are required for particular indications that become evident at examination. In addition, all of the participants in this study are given respiratory function testing including CO diffusion, rates and total lung volume as well as the routine spirometric and voluntary ventilation measurements. It is assumed that the major mode of intake of thorium has been by inhalation.

Serial measurements are desirable, in particular to evaluate changes in respiratory indices, and will probably be carried out at three-year intervals along with medical re-evaluation and radioactivity measurements.

## CENTER FOR HUMAN RADIOBIOLOGY

## Fact Sheet on

Bone StudiesMicrodosimetry at Endosteal Bone Surfaces (1.5 person-years)

Models of bone tumor induction proposed by Marshall and Groer predict a lower dose to bone cells at endosteal surfaces than expected from autoradiographic measurements. Because of its low spatial resolution, autoradiography provides only an indirect measure of endosteal dose rate and may be the reason for the discrepancy. To improve upon this, direct measurements of the energy released from bone surfaces are now being made using alpha particle spectrometry. Because of its high spatial resolution, spectrometry also allows us to determine the distribution of radium as a function of depth below the bone surface and, thus, to test a specific hypothesis explaining the disagreement between predicted and observed dose: The presence of a layer of bone, adjacent to the surface which is depleted in its radium content. We can determine whether or not the radium distribution is uniform by examining the spectrum shape. The shape also gives evidence as to whether a non-uniform distribution has a high or a low value adjacent to the bone surface. To date, we have found evidence of depleted layers in some bone samples and evidence of layers with enhanced concentrations in others. The relative frequency with which these types occur is yet to be determined.

The spectrum from a piece of radium bearing bone receives contributions from radium and each of its alpha emitting daughter products. In order to determine the concentration of radium or the dose rate from it, the radium contributions must be separated out. Because these spectra are quite complex, separation is a formidable task which we are attacking with computer methods and specialized measurement methods which allow us to determine the radium contribution directly but which require months of data collection. Computer separation has been successful in certain special spectra and we are now developing it to handle more general spectrum shapes.

Bone Surface to Volume Ratio Measurements (0.25 person-year)

The total area of endosteal bone surfaces enters the calculation of dose to bone surface cells. The surface area is calculated as the product of surface to volume ratio and bone volume. We have been measuring the surface to volume ratios in cancellous bone from 10 normal humans aged 19 to 59 years who died accidentally. The measurements are based on a new technique which utilizes the image produced by bone on plastic films when the bone and plastic are in tight contact and are irradiated by thermal neutrons. Past measurements in CHR have been made using microradiographic images which are known to be systematically low in value because the image is produced by relatively thick sections (about 100 microns) and, thus, have relatively low resolution. The neutron image is produced by a bone layer which is only 1 or 2 microns thick and the method should give results equivalent to those which would be obtained from direct measurements on sections of comparable thickness. A comparison between measurements made by the two methods shows that they differ by -3% to +21% and are correlated with the thickness

of the section used to produce the microradiographic images. This correlation agrees with theoretical prediction but the percentage difference between the methods is less than predicted. We believe that the method based on neutron induced images gives values which are as accurate as those produced from measurements on high resolution scanning electron micrographs.

#### Bone Remodeling Rates (0.1 person-year)

Bone remodeling plays a significant role in radium metabolism and is known to be reduced in dogs exposed to high radiation levels. Autoradiographic evidence has indicated reduced remodeling rates among patients whose doses are in the carcinogenic range. More data are needed, however, to specify the relationship between remodeling rate and radiation dose. Autoradiographic methods require long exposure times and therefore can be used only when the body burden is quite high. Thus, a dose response relationship, based on autoradiographic data, cannot extend over a broad dose range.

We are now using a non-autoradiographic method which is applicable to all dose levels. It is based on the numbers of intact and fragmented osteons which are visible in rib cortices. The osteons and fragments provide a record of the entire remodeling history of a cortical bone section and with the aid of proper calibrations give the average remodeling rate for that section. So far, we have applied the method to six cases of varying dose level and have found confirmation of the earlier autoradiographic results; i.e., the remodeling rate appears to be depressed when the dose level is high. Low dose levels have shown no depression of remodeling rate.

#### Microradiographic Assessment of Bone Damage (0.1 person-year)

Microradiography offers a method of studying bone damage which does not require the decalcification step normally used in histology. This preserves the radium content of the section which can then be used for autoradiography. This permits correlations between terminal radium concentration and damage to be made. We are surveying microradiographs from bone taken surgically, at autopsy or obtained by exhumation, for evidence of bone damage. Damage categories that we look for are plugged Haversian canals, unusually large or unusual numbers of resorption cavities, hypermineralized osteons, mosaic and fibrous bone in marrow spaces and unusual amounts of resorption at trabecular bone surfaces. These are present to varying degrees in our population, especially among patients who received high doses.

## CENTER FOR HUMAN RADIOBIOLOGY

## Fact Sheet on

Microdosimetry of Paranasal Sinus and Mastoid Air Cell Carcinomas (1.3 person-years)

Among radium cases, there is an unusually high risk of carcinomas in the paranasal sinuses and mastoid air cells. But, little is known about the dose delivered to the epithelial cells at risk because the target cell locations and the amounts of radioactivity producing the dose are not well known. This problem is complicated by the fact that several sources of radioactivity are present: Radium and its daughter products lodged in bone and radon and its daughter products contained in the air spaces and on the surfaces of, and within, the mucosal membranes. Radon and its daughter products have often been assumed to be more important than radium and its daughter products in bone, but there are no quantitative dosimetric data to support this. Furthermore, radon concentrations in the sinus and mastoid air spaces and in the mucosal membranes cannot be measured during life, or with great reliance post mortem.

Two efforts are now being made to solve these problems. The first is a histological study of the mucosal membranes in the paranasal sinuses and mastoid air cells. From this, we expect to determine the distributions of cell to bone surface and cell to epithelial surface distances and the populations of target nuclei so that practical microdosimetric calculations can be carried out given the distributions of radioactivity. Second is an autoradiographic study of the terminal distribution of radium in the walls and septa of the mastoid air cells and paranasal sinuses. From this and surface to target cell distances, we expect to determine the terminal dose rate to target cells within range of the bone surface due to radioactivity deposited in the bone.

Histological study of two low level radium cases and several unexposed individuals is now being carried out. So far, we have made at least one sampling of each paranasal sinus and mastoid. The observations show that the target cells reside almost exclusively in the epithelial membrane atop the lamina propria. Glands at intermediate distances are rare. The thicknesses of the lamina propria and epithelium vary widely within a particular site and between sites as well. In the two radium cases, we have observed areas in which the lamina propria is virtually absent, areas in which it is tens of microns thick and other areas in which it is hundreds of microns thick. Epithelial thicknesses have generally been less than 100 microns and usually a few tens of microns. These data indicate that radium and its daughter products in bone and radon and its daughter products in the air spaces, on the epithelial surface and dissolved within the mucosal membrane may all be important sources of radiation dose but will vary in importance from site to site according to variations in the thicknesses of the lamina propria and the epithelium.

Populations of target nuclei in the two radium cases are also quite variable. So far, the total cross sectional area of epithelial cell nuclei has ranged between about 10 and 40 percent of the cross sectional area of the epithelium. This variability should be reflected in a variability of tissue radiosensitivity from site to site. A further observation is that the nuclei are concentrated toward the bottom of the epithelium when the epithelium is thick and are thus shielded somewhat from alpha particles emitted by radon and its daughter products in the air spaces and on the epithelial surfaces.

Autoradiographs are being made and evaluated for the mastoid regions in 14 cases with mean skeletal doses in excess of 1500 rad. Autoradiographs of 7 of these, 3 with mastoid carcinomas on the side opposite the sampling site and one with a sphenoid carcinoma, have been examined. All show a nearly complete absence of hot spots in the air cell septa, but substantial hot spot activity in the lateral and medial walls of the temporal bone. It, thus, appears that formation of the mastoid air cells had ceased by the age of first exposure (14-21 years) and that bone remodeling was absent. Dosimetrically, this means that the mastoid mucosa were irradiated almost exclusively by a diffuse deposit rather than by diffuse and hot spot deposits, the common situation throughout the skeleton.

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Fact Sheet on  
Energy-Related Stresses on Great Lakes Fishes (4 person-years)

The emphasis of this program has been the quantification of direct effects of cooling systems. Through a series of field experiments in Lake Michigan we have shown that:

- Most salmonid and forage species are attracted to thermal effluents during spawning seasons, while brown and rainbow trout are attracted periodically throughout the year. Individual salmonids are not permanent residents of any one discharge but they do reside temporarily in many of the discharges encountered during normal migrations.
- Fish densities in plume regions vary with normal fluctuations in inshore abundances, but densities in plumes often exceed those in ambient areas by a factor of 10 or more. The highest densities of fish often occur at plume-ambient interfaces and at intermediate plume temperatures.
- Plume-resident salmonids "control" their body temperatures (thermoregulate) by avoiding high temperatures and by partitioning residence time between warm and cool temperatures, but they do not maintain constant body temperatures for more than a few hours. Despite wide fluctuations in temperature exposure, mean body temperatures often approximate optimum metabolic temperatures.
- Temperature selection is a complex function of water temperature, time of year, fish size and recent thermal exposure (acclimation). Juvenile trout select and acclimate to higher temperatures than large trout.
- Plume-resident rainbow trout increase their acclimation temperature at least 2°C and up to 10°C over ambient, and maximum increases in acclimation occur when water temperatures are low.
- Sport fishermen also concentrate at thermal discharges and warm water fisheries now account for 1-2% of the total salmonid catch in Lake Michigan. We estimate that 10-20% of all the salmonids caught in Lake Michigan have resided in thermal effluents.
- Studies of impingement and entrainment at water intakes show that very large numbers of alewife and smelt are killed annually in Lake Michigan, but the significance of this unnatural mortality is unknown. Estimates of fish mortalities at intakes are used to identify sensitive populations and relatively destructive intake designs.

The information gained from previous studies is being applied to new experiments and simulation models designed to predict the impacts of thermal discharges and water intakes.

- Since elevated fish body temperatures result in predictable increases in metabolic rates, we are simulating the energetic costs of plume residence

by salmonids. Preliminary projections (Table 1) show energy (food) deficits for plume-resident trout in winter, when forage densities are very low. At other times of the year forage densities are high enough to offset the increased demand for food by plume-resident trout.

Table 1. Energy requirements of two brown trout tracked at Point Beach in 1977.

	1.6 kg Female	2.7 kg Male
Track dates	9 February—2 March	13 April—3 May
Total track hours	410	452
Hours in ambient	276	391
Hours in plume	134	61
Ambient temperature (°C)	2	6
Mean body temperature	13 (15—18)	11 (7—14)
Physiological costs (calories/hr)		
Ambient	77	615
Plume	1064	1196
Cost index (plume/ambient)	13.8	1.9

—The indirect effects of winter plume-residence by trout may be the utilization of fat reserves (energy store), loss of weight and reduced gamete production. Increased forage consumption during other seasons could amount to a very significant increase in lakewide predation on alewife and smelt.

—Increase in temperature exposure, metabolic rate and food consumption of plume resident salmonids (and other organisms) may result in significant changes in pollutant cycling and transfer to man. Preliminary experiments indicate significant increases in body fat and radiocesium concentrations in plume fish during the fall. We are initiating experiments to quantify changes in accumulation of inorganic, organic and radioactive pollutants by plume resident trout.

—An EPA sponsored study (RER, EIS, University of Michigan) is projecting the impacts of cooling system on forage fish populations and salmonid production in Lake Michigan. Mathematical simulations incorporate the ANL intake data base, and fish population data from the U.S. Fish and Wildlife Service.

Fact Sheet on  
Cycling and Effects of Trace Elements in Plankton and  
Benthic Communities (4 person-years)

Similarities between trace element concentrations in Lake Michigan water and the lowest levels known to cause toxic effects in aquatic biota were compared to select "high risk" trace elements for investigations of uptake, cycling and effects. Zinc, cadmium, mercury and copper were selected. To date, ANL studies have confirmed the incorporation of zinc and cadmium by plankton and measured their toxic effects on cultured phytoplankton and natural assemblages of zooplankton at elemental concentrations less than one order of magnitude above ambient levels in Lake Michigan.

Uptake and Cycling Studies

Recently we developed a coordinated program to quantify the role of lake biota in the cycling of trace elements and to measure long-term variations (trajectory) in natural structure and function within Lake Michigan. These studies center on trace element concentrations in biota, incorporation rates, bioaccumulation rates, and trophic transfer processes in the planktonic and benthic communities.  $^{65}\text{Zn}$  and  $^{109}\text{Cd}$  tracers are used to study cycling simultaneously with the in situ effects experiments (described below). We anticipate combined cycling and effects research will improve our ability to predict both the fate and effects of future trace element inputs to the Great Lakes.

The role of benthic fauna in trace element cycling and the potential for effects on benthos are studied with microcosms which consist of the top 30 cm of intact sediment cores from Lakes Erie and Michigan that are incubated at 4°C and spiked with  $^{65}\text{Zn}$  and  $^{109}\text{Cd}$ . Thus far, we have kept sensitive benthic fauna, i.e., *Mysis* and *Pontoporeia*, alive for ~2 months and the approach appears promising.

A new onboard system has been developed to simultaneously map total chlorophyll a and total seston mass while underway. The technique will improve our ability to obtain representative measurements and to understand the nature of heterogeneous water masses and it will provide useful baseline information applicable to our prediction of trace element fate and effects.

Effects Studies

Studies of pollutant effects are being performed in experimental microcosms, ponds, lakes, and in situ enclosures of various kinds. An in situ method using polyethylene carboys as enclosures is being used to study

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pollutant effects on natural plankton communities in large lakes. The effects of enclosure in translucent carboys at optimal depths for three weeks are not greater than those observed in larger enclosures in two Canadian lakes (ELA lakes 223 and 382). Percentage similarity (PS) values of zooplankton communities 3 weeks after cadmium enrichment in large enclosures in ELA lake 223 were within the 95% confidence limits predicted from the results of our 3-week experiments in Lake Michigan (Fig. 1).

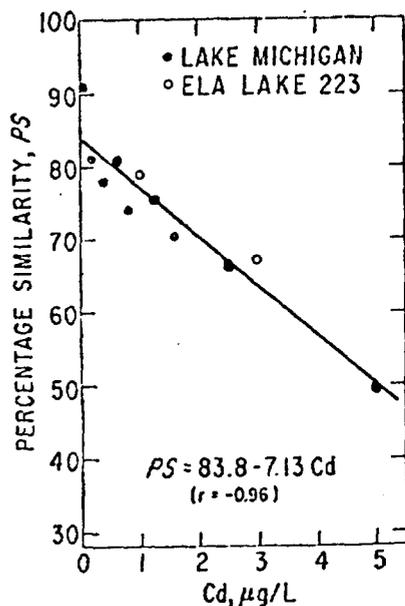


Fig. 1. Effects of cadmium on percentage similarity of crustacean zooplankton communities three weeks after cadmium enrichment of enclosures in Lake Michigan and ELA lake 223.

Preliminary studies on the effects and interactions of zinc and cadmium enrichment on Lake Michigan plankton demonstrate the value of using large and small enclosures to simulate pollutant effects in lakes. However, more detailed studies of the effects of using different sizes of enclosures simultaneously in different lakes are needed to determine the utility of small enclosures. Studies are in progress in Lake Michigan and ELA lake 382, where a 3-hectare bay was isolated in May 1978 in preparation for a cadmium effects experiment including large and small enclosures. We anticipate that our in situ experiments with small enclosures will complement the experiment with the entire bay.

## ECOLOGICAL SCIENCES SECTION

Fact Sheet on  
Impact of Fossil Fuel Combustion on Crop Plants (4 person-years)

Projected increases in the use of coal for generation of electrical power have caused concern regarding the environmental impact of combustion emissions. Gaseous and particulate emissions from coal combustion place large quantities of sulfur (primarily SO<sub>2</sub>), nitrogen, carbon dioxide, and heavy metals in the environment. Secondary reactions produce ozone and acid rain, all of which are of concern as environmental contaminants. Due to the relative sensitivity of many plant species to SO<sub>2</sub> in the atmosphere and the importance of agriculture in the midwest, a project concerning SO<sub>2</sub> effects on midwestern crop species is being pursued.

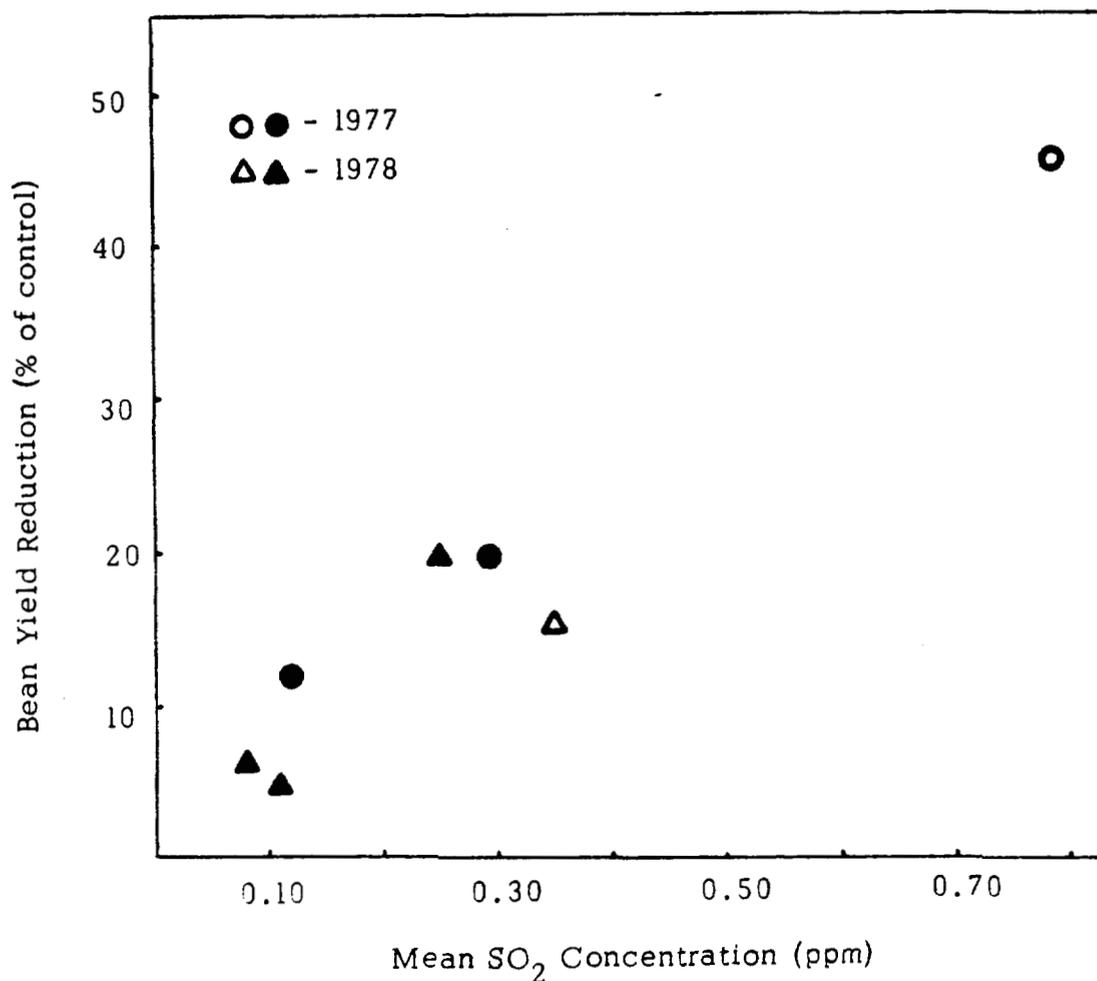
In the summer of 1977 three 80' × 100' field plots of soybeans were periodically fumigated by controlled release of SO<sub>2</sub> through a system of pipes suspended over the plant canopy. The fumigations were performed on 24 days between early July and late August for an average of 4.75 hours per day. The 1978 experiment was similar with 4 plots exposed to 19 fumigations averaging 3.8 hours per day. The mean SO<sub>2</sub> concentrations during fumigation were 117, 300 and 786 ppb in 1977 and 94, 109, 255 and 353 ppb in 1978. Only the plot receiving 786 ppb in 1977 exceeded the national secondary air quality criteria for SO<sub>2</sub>.

Some chlorosis and bronzing of leaf tissue were observed in the plot receiving 786 ppb of SO<sub>2</sub> in 1977, and mild chlorosis was observed in the 353 ppb plot in 1978; no other plots exhibited visible injury. However, bean yields were reduced in all the fumigated plots, regardless of whether or not visible injury occurred (Fig. 1). The bean yield reductions were primarily due to decreases in seed size, numbers of filled seeds per pod, and numbers of pods per plant. The protein and oil contents of the seeds in 1977 were not extensively affected by the SO<sub>2</sub> treatments, although the seed concentrations of several essential elements were altered.

The effect of SO<sub>2</sub> fumigation on photosynthetic rate of leaf tissues was apparently contingent upon the age of the tissues, their prior exposure to SO<sub>2</sub> and the prevailing environmental conditions. In 1977 photosynthesis was reduced as much as 63% at the highest SO<sub>2</sub> concentrations, while a short-lived stimulation of photosynthesis was occasionally noted at the lowest SO<sub>2</sub> concentrations. Post-treatment carryover of the photosynthetic effects was noted only in the tissues exhibiting visible damage. A survey of the incidence and severity of fungal plant pathogens in the SO<sub>2</sub> treated plots in 1977 did not indicate any significant SO<sub>2</sub> related effects. A preliminary study indicated some promise for the use of aerial color and color infrared photography in detecting soybean seed yield reductions.

Dissertation research of P. M. Irving involved a study on the effects of acid precipitation through application of simulated acid rain (pH 3.0) and control rain (pH 5.4) to plots of soybeans receiving no SO<sub>2</sub> and plots receiving ~230 ppb SO<sub>2</sub>. A total of 4.5 cm of precipitation simulant was applied over a 9-week period. Results indicate that acid precipitation causes no decreases in yield of SO<sub>2</sub>-exposed and unexposed soybeans. Data from this field study and from chamber experiments suggest the possibility of increased productivity of soybeans receiving acid precipitation due to a fertilizing effect by the higher nitrogen and sulfur found in acid precipitation.

Fig. 1. Effect of SO<sub>2</sub> fumigation on soybean yields in 1977 and 1978. Open symbols indicate plots where visible damage to leaves was observed.



Fact Sheet on  
Oxidation State of Plutonium in Natural Waters (3 person-years)

Differences in environmental behavior between different oxidation states of plutonium are expected to be as great as are the differences between uranium and thorium. Thus the accurate prediction of the fate and radiological significance of a plutonium input requires a knowledge of the distribution of oxidation states both within the source and after equilibration with the recipient water system. Since the oxidation state of plutonium cannot be reliably predicted on theoretical grounds, an experimental program was initiated in 1977 to measure it in Lake Michigan, the coastal waters around Great Britain, and a series of Canadian lakes. The analytical technique chosen separates Pu(III) and Pu(IV) from Pu(V) and Pu(VI), an optimum separation since either of the two lower oxidation states should be much more insoluble than either of the higher oxidation states.

Analyses of the concentrations of plutonium in lower and higher oxidation states in samples from 2 or 3 depths at two Lake Michigan stations were carried out during the summer and fall of 1977. For all hypolimnetic samples the mean concentration of Pu(III,IV) was found to be  $0.08 \pm .03$  fCi  $L^{-1}$  and of Pu(V,VI) was  $.34 \pm 0.07$  fCi  $L^{-1}$ . No seasonal trends in the ratio of higher to lower oxidation states can be discerned. Little or no Pu(V,VI) was detectable in particulate material filtered from the water or in surficial sediment. Thus the distribution coefficient ( $K_D$ ) for Pu(III,IV) in Lake Michigan appears to be  $1-2 \times 10^6$  while that of Pu(V,VI) is  $\leq 10^4$ , and the  $K_D$  for total plutonium is  $2-4 \times 10^5$  as previously reported.

A previous study in a limited series of natural waters showed considerably higher total plutonium concentrations in the more acidic systems. With the cooperation of the Freshwater Institute of Environment Canada, water samples were obtained from a series of lakes ranging in pH from 4.8-8.5. Within the alkaline pH range (7-8.5), Pu(V,VI) concentrations were found to be relatively constant ( $0.1-0.7$  fCi  $L^{-1}$ ), while those of Pu(III,IV) were found to be much more variable and ranged from 0.1 to 8.3 fCi  $L^{-1}$ . In two of the cases (both oligotrophic-calcareous), the Pu(V,VI) accounted for more than 50% of the dissolved plutonium as it does in Lake Michigan. In the acidic natural waters, Pu(V,VI) accounts for  $\leq 10\%$  of the dissolved plutonium. The occurrence of high concentrations of Pu(III,IV) is accompanied by high concentrations of dissolved thorium which suggests solubilization of the IV oxidation state by naturally occurring ligands.

A cooperative program with the Fisheries Radiobiological Laboratory (Lowestoft, Suffolk, United Kingdom) allowed the collection and analysis of a similar set of data in the ocean. Most of the plutonium in these samples originated from the discharges of the Windscale Nuclear Fuel Reprocessing Plant, in contrast to the fallout origin of the plutonium in Lake Michigan and the Canadian lakes. The oxidation state of plutonium in solution for all

seawater samples was observed to be a mixture of Pu(III,IV) and Pu(V,VI) with between 60 and 95 percent of the total being Pu(V,VI). The highest percentages of Pu(V,VI) occurred within the Irish Sea, the lowest in the open ocean. Plutonium associated with colloids was never a major fraction of the total: approximately 20% of the Pu(III,IV) and less than 5% of the Pu(V,VI) being removed by passage through a 10,000 molecular weight (0.0025  $\mu$ m) dialysis membrane after filtration through a (0.22  $\mu$ m) membrane filter. As was found in Lake Michigan the plutonium on particles was found to be almost entirely Pu(III,VI). Concentration factors for the two oxidation states of plutonium onto particles were generally in the range of  $1-5 \times 10^6$  for Pu(III,IV) and  $<5 \times 10^3$  for Pu(V,VI), which are quite similar to those for Th(+4) and U(+6). Concentration factors measured for americium onto the same particles were almost identical to those of Pu(III,IV) suggesting the adsorption of Pu(III,IV) and americium were similar processes for a wide range of particle types.

Since 5% of the plutonium discharged from Windscale appears in the Irish Sea as Pu(V,VI), the question of the oxidation state present at discharge was examined. Samples of Windscale effluent were analyzed for plutonium oxidation states and no evidence was found to suggest that more than 1% of the current plutonium discharge is Pu(V,VI). The 5% Pu(V,VI) observed in the environment would seem to be a product of the environmental oxidation of Pu(III,IV) rather than to be a remnant of a Pu(V,VI) discharge.

The possibility of direct oxidation of Pu(IV) by seawater was confirmed in laboratory experiments using tracers of unknown oxidation state added to filtered seawater. Oxidation rates of 0.5 to 3 percent per day were observed. These rates are adequate to account for the Pu(V,VI) observed in the environment.

Since Pu(V,VI) appears to be generated in the environment, the long-term stability of the remaining 95% of the released plutonium still residing, as Pu(+4), on the seabed was examined by measuring the plutonium content of interstitial water squeezed from cores collected near Windscale. The concentrations of both upper and lower oxidation states of plutonium measured in the uppermost slice (0-5 cm) were similar to the corresponding concentrations measured in overlying water. Below 5 cm the Pu(V,VI) practically disappeared while the Pu(III,IV) decreased in direct proportion to the decrease in plutonium on the solid phase. Since Pu(V,VI) was detected in the interstitial water of the surface layer, the return of plutonium from the surficial sediments as Pu(V,VI) cannot be ruled out. The Pu(V,VI) in the overlying water could originate from the oxidation of either (a) 5% of the currently discharged plutonium or (b) a smaller fraction of the large inventory of plutonium residing in the surficial sediments. The constancy of Pu(III,IV) concentration factor ( $\sim 10^6$ ) with depth argues against the formation of a more soluble reduced species of plutonium, either Pu(III) or an organic complex of Pu(IV), after burial in the sediments.

Argonne National Laboratory  
 ATMOSPHERIC PHYSICS SECTION

Fact Sheet on  
Improved Sodar Capabilities (0.5 person-year)

The ANL acoustic sounders have been modified to enable the quantitative evaluation of thermal turbulence in the Planetary Boundary Layer (PBL). With the acquisition of a Digital Equipment 1134 minicomputer as well as an analog-to-digital converter, the ability to quantify the scattered acoustic signal has become a straightforward reality. The challenge lies in the 'calibration' of this data to give meaning to the sensed quantities.

The calibration falls into two categories: determination of the total acoustic power transmitted into the atmosphere and evaluation of the response of the system to incoming (i.e., scattered) acoustic energy. With these two functions, the returned acoustic energy can be related to the RMS temperature fluctuations using the acoustic scattering cross-section equation.

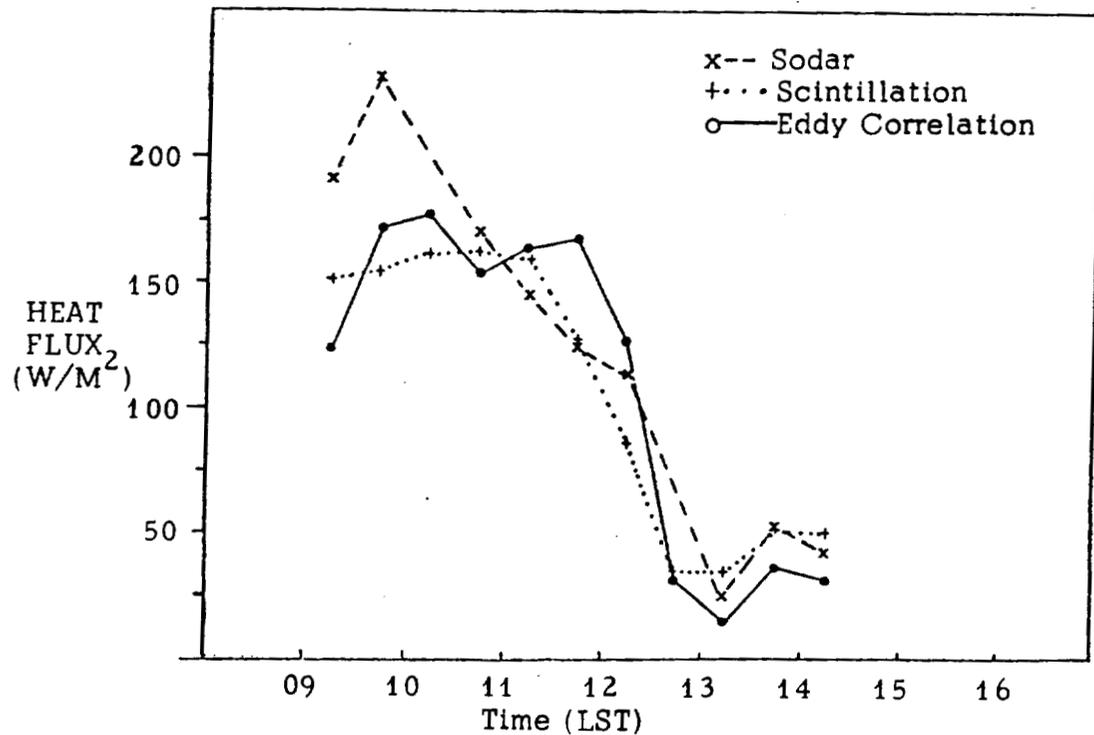
With a calibrated microphone suspended above the sodar in various positions, the beam pattern is mapped out; similarly the output power of the sodar antenna along the beam axis is measured. Integration of the product of the normalized beam pattern magnitudes and the output power over the solid angles defined by the beam pattern measurements provides an estimate of the output power. Inverting the arrangement, which means placing the microphone beside the antenna and a small speaker aloft, gives a measurement of the sodar sensitivity at a given acoustic pressure level (determined by the microphone). Measurements at several amplitudes provide a calibration curve and permit determination of the magnitude of the scattered acoustic pressure fluctuations. The ratio of returned to output power for a monostatic system can then be related directly to  $C_T^2$ , the temperature structure parameter.

Knowledge of this parameter is useful to the efforts of this Section in various ways. For example, the magnitude of  $C_T^2$  in the elevated inversion capping the mixed layer is a measure of the temperature difference across this interface; its variation with height in the lower PBL is directly related to the surface heat flux. When the condition of free convection is approached,  $C_T^2$  in the lower PBL varies with height in a manner determined primarily by the sensible heat flux.

A test of the free convection relationship was carried out at ANL in June of 1978. Measurements of heat flux determined via the sodar, by eddy correlation techniques already well established, and by scintillation of a laser beam over a known path in the PBL were taken. Results indicate that this method can indeed give reasonable estimates of the surface heat flux, particularly from about 1000 to 1500 hrs. LST. The figure below shows the derived values of heat flux using the three methods through the daytime hours on June 14, 1978. These results are only preliminary, but encouraging.

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The sodar system has been extended at present to make heat flux estimates automatically and continuously over a given time period and store the values on magnetic tape. Using this arrangement and a doppler sodar under development, similar evaluations can be made of mechanical turbulence through  $C_v^2$ , leading to estimates of the dissipation rate. Also, mean wind speed estimates up to approximately 500 meters can be made by use of the doppler capability.



Argonne National Laboratory  
ATMOSPHERIC PHYSICS SECTION

Fact Sheet on  
The Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) model  
(0.5 person-year)

The ASTRAP model has been developed for application to studies of long-term regional-scale dispersion of air pollution, primarily that resulting from sulfur pollutants released by fossil-fuel combustion. ASTRAP combines the statistical trajectory method of estimating horizontal dispersion, first developed in Europe and initially applied here by Jimmy Sheih, with improved parameterizations: wet removal is a function of the half power of the precipitation rate, and the rate of transformation of sulfur dioxide to sulfate, the stability profile, and the deposition velocities for  $\text{SO}_2$  and  $\text{SO}_4^-$  are all given typical diurnal variations. The latter parameterizations incorporate the results of the field studies of this section. An additional improvement in ASTRAP is the capability to estimate net mass fluxes, such as across the Atlantic coast or the U. S. — Canadian border, as well as net fluxes to the surface by wet and dry processes.

Sensitivity tests performed with a partial emission inventory of coal-combustion sources of sulfur pollutants indicate that the diurnal variations in the parameterizations can have somewhat mixed effects in simulations. The variations in deposition velocities increase seasonal average surface concentrations by 10-20% over the case in which deposition velocities are held constant; the variation in the transformation rate decreases  $\text{SO}_4^-$  concentrations 5-10% while having little effect on  $\text{SO}_2$  concentrations; and the variation in stability profiles decreases  $\text{SO}_2$  concentrations 20-40% and sulfate concentrations 15-25%. The reason that the stability variation has the greatest effect is that the nocturnal inversion cycle simulated by the stability profile variation effectively decouples pollutants aloft from surface processes during part of the diurnal cycle. When all diurnal variations are included (Figs. 1a-b) simulated concentrations are 20-40% lower than for the case of no diurnal variations (Figs. 2a-b). Since wet deposition is a sink for both  $\text{SO}_2$  and  $\text{SO}_4^-$ , increasing the rate of wet removal always decreases concentrations. A change from 50% removal by a rainfall of  $2 \text{ mm hr}^{-1}$  to 50% removal at  $4 \text{ mm hr}^{-1}$  increases atmospheric concentrations by 10-20%. A change of transport winds, precipitation fields, and diurnal variations of parameters from summer to winter reduces  $\text{SO}_2$  concentrations by 30% and sulfate concentrations by 60%.

The ASTRAP model has been used to investigate several questions for EPA, including the relative importance of primary sulfate (emitted from stacks) and secondary sulfate (formed by transformation in the atmosphere) and the relative importance of coal combustion and oil combustion in resulting sulfate concentrations. This latter area is of interest because oil combustion may release a considerably higher portion of sulfur as primary sulfate than does coal combustion.

A budget from part of this study indicates that for coal combustion sources in the eastern U.S. in summer, 37% of the sulfur is scavenged by precipitation, 22% by dry deposition, 24% passes into the Atlantic, 12% into Canada, and 4% into the Gulf.

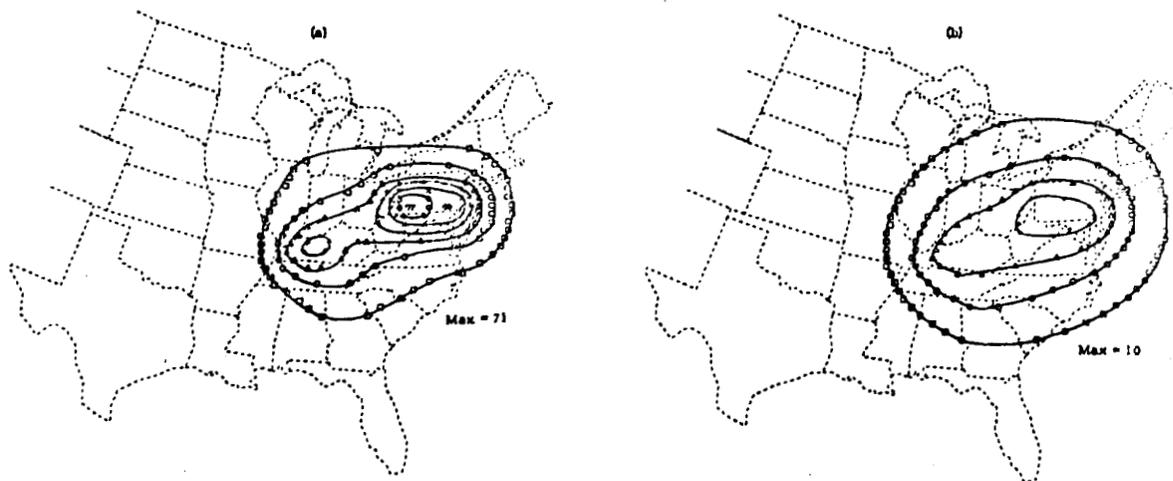


Fig. 1. Seasonal average concentrations of (a) SO<sub>2</sub> and (b) SO<sub>4</sub> resulting from coal combustion during summer, with diurnal variations in parameterizations. Minimum contour and contour interval are 10 μg/m<sup>3</sup> for SO<sub>2</sub> and 2 μg/m<sup>3</sup> for SO<sub>4</sub>.

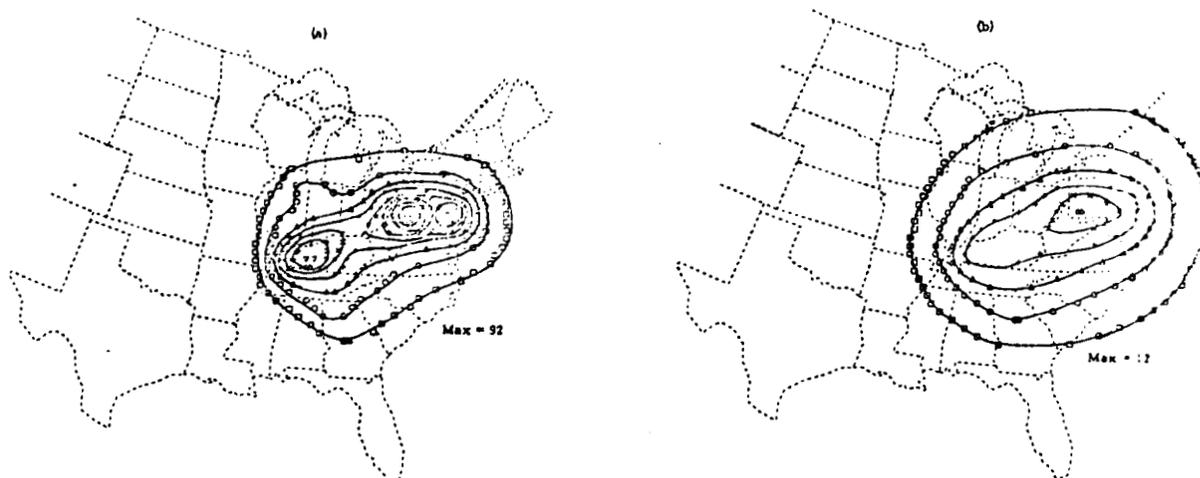


Fig. 2. Same as Figure 1, except that parameterizations have no diurnal variations.

## FUNDAMENTAL MOLECULAR PHYSICS AND CHEMISTRY SECTION

## Fact Sheet on

Molecular Radiation Physics (about 7 person-years)

This program is directed at extending the knowledge of elementary molecular processes involving electronic excitation. Methods of research are in part theoretical and in part experimental. Work concerns the excitation processes of molecules by energetic agencies (photons, electrons, and other charged particles) as well as the properties of the initial species such as excited states, ions, radicals, and secondary electrons. The resulting information is essential as a prerequisite to sound and detailed modelling of succeeding chemical reactions that occur in irradiated molecular substances including biological materials. The program may be broken into five closely related subprograms as follows.

## A. Electron collisions (2.5 person-years)

The goal of the work is to measure cross sections for inelastic collisions of electrons with molecules at electron energies below about 1 keV. Differential cross sections are measured by analysis of electron energy losses. Integrated cross sections for slow incident electrons are measured by various other techniques.

## B. Photoabsorption and photoionization cross sections (1.6 person-years)

Absolute cross sections for photons in ultraviolet and far ultraviolet regions of polyatomic molecules are measured. The data are related with electron energy-loss cross sections taken under certain conditions. Results of subprograms A and B are the most basic data for the theory of initial-species yields.

## C. Fundamental radiation physics and chemistry (1.8 person-years)

Using methods of modern theoretical physics, we study many elementary processes and properties, some of which are subjects of experimental study in our group. Some of the current work concerns 1) spectral distribution of the oscillator strength of atoms and molecules, 2) new methods of computing continuum electron wavefunctions in anisotropic molecular fields, for photoionization and electron collision calculations, and 3) basic quantities in radiation physics such as stopping power, yields of initial species, and electron slowing-down spectra.

## D. Theoretical study of secondary electrons (0.7 person-year)

Energy and angular distribution of secondary electrons are essential inputs to radiation-physics modelling. A number of methods of analysis

of experimental data have been developed and are used to test data reliability.

E. Analysis of the initial products formed in photoionization (0.5 person-year)

Photoelectrons as well as ions resulting from absorption at various fixed wavelengths are analyzed. Results elucidate intricate details of photoeffect and also provide an important classification of different states of ions, i.e., the most important of the primary species under irradiation in general.

## FUNDAMENTAL MOLECULAR PHYSICS AND CHEMISTRY SECTION

## Fact Sheet on

Molecular Physics Basic to Atmospheric Chemistry (about 3 person-years)

The program aims at providing basic data for characterization, measurements, and chemical behavior of atmospheric pollutants and related substances at the molecular level. Methods of research are largely laboratory-experimental, and in part theoretical. Most of the pollutant molecules considered are related to coal combustion and other non-nuclear energy technologies.

The work is divided into three subprograms addressed to different scientific aspects.

## A. Chemical physics of atmospheric pollutants (1.3 person-years)

The first area of study concerns mechanisms and kinetics of cluster formation in high-pressure gases, a topic of well-known importance in atmospheric chemistry. The second area of study concerns free radicals in the atmosphere as reaction intermediated. Major tools of study are a quadrupole mass spectrometer and a variable-wavelength light source. A high-pressure supersonic jet is used to produce a good amount of small molecular clusters.

## B. Spectroscopic properties of pollutants and related molecules (1.4 person-years)

Detailed spectroscopic information about polyatomic molecules is essential for development of new detection methods and pollution-control techniques. We mobilize expertise existing in the group in the areas of photoabsorption spectroscopy, electron-impact spectroscopy, and photoelectron spectroscopy. Examples of study have been  $N_2O$ , hydrocarbons, and fluorocarbons.

## C. Theoretical chemical physics (0.3 person-year)

Theoretical methods are used to interpret some of the experimental work generated under subprograms A and B, and also to generate necessary data on molecules that are difficult for laboratory study. Topics of current work concern electron-molecule at low energies and some basic aspects of cluster formation.

## FUNDAMENTAL MOLECULAR PHYSICS AND CHEMISTRY SECTION

## Fact Sheet on

Atomic and Molecular Physics for Energy Systems

## Atomic Physics Related to Fusion Energy (1.7 person-years)

The objective of the program is to generate reliable atomic data for use in fusion technology. The efforts may be divided into two subprograms.

## A. Data for Tokamak and other magnetic-confinement studies

Properties of highly stripped atomic ions, including energy levels, transition probabilities, and electron-collision cross sections, are needed for plasma diagnosis as well as for plasma modelling, especially for the understanding of the plasma cooling due to contaminants from walls. These contaminants are often heavy metal elements and will take the form of the highly stripped ions in plasmas. The expertise existing in the group in accurate calculations on these properties specifically including many-electron correlation effects and relativistic effects is applied to this current task.

## B. Data for heavy-ion induced inertial-confinement fusion

As a part of the Argonne project for heavy-ion induced pellet fusion, calculations of several kinds of cross sections are being performed. One kind concerns charge transfer between heavy ions, which have to be stored in a storage ring; charge transfer processes govern the lifetime of the stored ions. Another kind concerns collisions of high-energy heavy ions with molecules of background gases including air and hydrogen. These collision cross sections determine the requirement of the level of vacuum for ion accelerators.

## Electron Collision-Cross Sections for Modelling of MHD Plasma Conductivity (0.4 person-year)

In a coal-fired open-cycle MHD power generator, there are numerous molecular species and a relatively small number of ions and electrons. The efficiency of the generator depends crucially on the electrical conductivity of the weakly ionized plasma, which in turn is governed by low-energy electron interaction with molecules. A new method of calculation developed in our group is excellently suitable for study of this topic, and is being applied to increasing number of molecules.