ENVIRONMENTAL CONSEQUENCES OF ATMOSPHERIC
KRYPTON-85

MASTER

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ABSTRACT

Krypton-85 is a radioactive inert gas produced during normal operations of the nuclear fuel cycle. The quantities of krypton-85, that will be produced in the next century, are sufficient, if released, to alter the electrical state of the atmosphere. The principal hypothesis is that an anthropogenic alteration of the electrical state of the atmosphere could alter other meteorological phenomena and lead to significant environmental impacts. The goal of this project was to explore some areas of basic science related to the evaluation of that hypothesis. The approach was primarily theoretical. The following topics were addressed, (1) a first approximation model to estimate the effects of krypton-85 on the electrical state of the atmosphere; (2) an analysis of the pathways between krypton-85 production and the atmosphere; (3) an analytical model for fair weather atmospheric electricity, and (4) a dipole model for atmospheric electricity. The results will provide a framework on which detailed models can be built. The results should provide better understanding of some topics in atmospheric electricity.

The initial proposal was submitted to ERDA in July 1975. Funding for the proposal became available during 1977. In the midst of the final negotiations on the contract, the principal investigator learned that he had received a one-year NSF Science Faculty Professional Development Award. It was agreed to begin the contract, then have a one-year suspension of work.

The contract was renewed for one additional year by supplementing the funds carried over with new funds to bring the annual rate back to the original support level.

The goal of this contract was not to produce a definitive report on the environmental consequences of atmospheric krypton-85 but rather to do some of the preliminary work on the scientific basis for such an assessment. An ionizing agent such as krypton-85 would directly modify the electrical conductivity of the earth's atmosphere. This perturbed conductivity profile would lead to adjustments of the atmospheric electrical field and air-earth current. At present, there is not an adequate model to describe the chain of phenomena linking the electrical state of the atmosphere to the microphysical processes of charge separation; to the cloud microphysics of precipitation charging; to the dynamical motions of ions and charged precipitation in a cloud dynamics model; to the conversion of mechanical energy to electrical energy in an electrified cloud; to the transport of electrical energy from a single storm cell to its immediate environment; to the patterns of electrical current flow in the earth-ionosphere cavity; to the resultant ambient electrical field in which a second storm cell originates at a location thousands of kilometers away from the first cell. That comprehensive model is clearly beyond the scope of this contract. The objectives of this work were (1) theoretical development of atmospheric electrical theory from first principles to provide a framework for further electrification models and constraint equations to test the physical reality of present models; (2) identification of significant scientific publications related to this issue; (3) an analysis of existing empirical atmospheric electrical data and (4) a one-year measurement program to extend the base of empirical data.

Within a few generations the leaders of nuclear nations will have the capability to manipulate the environmental radiation background of the atmosphere in the limited sense that they could increase the background by releasing radioactive inert gases. The consequences of such releases are not fully known. The environmental consequences of such releases, if any, would be in addition to other anthropogenic influences on the earth's climate.

I. Theoretical Developments

A. First-Approximation Model to Estimate the Effects of Krypton-85 on the Atmosphere

This model focuses attention on the lower atmosphere by assuming that the earth and the ionosphere are concentric spherical surfaces. Current flow between them is essentially radial. The total current passing from the earth to the ionosphere in either direction is taken to be the sum of the currents from various areas on the earth's surface. Each area is assumed to have a horizontally uniform electrical conductivity profile, which will differ from one area to another. This model treats mountainous area as uniform plateaus. The output of the total thunderstorm activity in the atmosphere is assumed to be constant. This has the effect of decoupling the thunderstorm generators from their electrical environment. If, as some authors have suggested, there is a feedback between thunderstorms and ambient atmospheric electric fields, this model underestimates the effects.

The equations that result from the model are as follows:

1. The electric field as a function of height in a horizontally uniform area labeled by index i , $E(i)$ is equal to the air-earth current density in that region $(J)_i$ divided by electrical conductivity at height, $\sigma(h)_i$, in that area. This is expressed as

$$E_i(h) = \frac{J_i}{\sigma_i(h)} \quad (1)$$

Equation (1) is a restatement of Ohm's Law.

2. The vertical air-earth current density in area i is equal to the ratio of the total resistance between the earth and the ionosphere (R) to the columnar resistance of a one-square meter column of air extending from the earth to the ionosphere in area i times the product of the total surface area of the earth (A) and the summation of the air-earth current density in each area J_i times the fractional area covered by area, (f_i) .

$$J_i' = \frac{R}{R_i} A \sum J_i f_i \quad (2)$$

The current flow will be larger in areas of below average columnar resistance.

3. The ratio of the electric field at height (h) and over an area of i in an atmosphere contaminated by krypton-85 $E_{ik}(h)$ to the same parameter in an identical atmosphere uncontaminated by krypton-85 ($E_i(h)$) is the product of three ratios. The first is the ratio of the total resistance of a contaminated atmosphere R_k to the total resistance of an uncontaminated atmosphere. The second is the ratio of the columnar resistance of area i for uncontaminated air, R_i , to the same parameter for contaminated air. The third term is the ratio of the local conductivity with contamination $\sigma_{ik}(h)$, to the conductivity without contamination.

$$\frac{E_{ik}}{E_i} = \frac{R_k}{R} \frac{R_i}{R_{ik}} \frac{\sigma_{ik}(h)}{\sigma_i(h)}$$

The effect of krypton-85 is to decrease the total resistance and the columnar resistance and increase conductivity. Therefore, we can predict

$$\frac{R_k}{R} \leq 1, \quad \frac{R_i}{R_{ik}} \geq 1, \quad \frac{\sigma_{ik}}{\sigma_i} \geq 1$$

The vertical distribution of krypton-85 and cosmic-ray ionization allows the specification

$$\frac{\sigma_{ik}}{\sigma_i} = 1 \quad \text{for } h \geq 10 \text{ km}$$

$$\frac{\sigma_{ik}}{\sigma_i} < 1 \quad \text{for } h = 0 \text{ km.}$$

The result of the calculation of the ratio of the electric field with and without krypton-85 for a typical land station and a typical ocean station are:

Height	Ocean	Land
0	.7642	.8393
2 km	.8691	.8393
4 km	.9560	.9114
6 km	.9970	.9505
10 km	1.0011	.9544

The columnar resistance over the ocean is reduced by 16% and the columnar resistance over land by 12%.

The five percent reduction in the electric field at 10 km above continents is due to a corresponding decrease in the air-earth current. The tendency is for more current to sink into ocean areas. The net result is that at cloud base (2 to 4 km in altitude) the reduction in the electric field over land and oceans are quite similar. The reduction are projected to be maximum at the ocean surface and minimum at 10 km above the oceans.

B. An Analysis of the Pathways Between Krypton-85 Production and the Atmosphere

Figure I seeks to illustrate the various decisions that affect the atmospheric concentration of krypton-85. The only significant source of this isotope is nuclear fission of uranium, thorium and plutonium. The only significant removal mechanism is the transformation of krypton-85 to rubidium-85 by nuclear beta particle decay. The atmospheric concentration is determined by the rate of release and the rate of decay of this isotope.

The major sources of krypton-85 are on the top line; they include civilian nuclear power, military and research reactors, nuclear weapons production and nuclear explosions. In the past, plutonium production has accounted for most of the atmospheric krypton-85. Projections for the future indicate that civilian nuclear power reactors will become the dominant source of krypton-85 in the future.

Each nation using nuclear reactors will contribute a share to the global inventory of krypton-85. The summation symbol with subscript N indicates that nuclear decision in many individual countries produce the end result.

The steps between the production and release of krypton-85 for different paths can be outlined as:

1. Nuclear explosions. Atmospheric nuclear explosions leave 100% of the krypton-85 that they produce in the atmosphere. Unless underground explosions are accidentally or deliberately vented, the krypton-85 they produce will remain trapped in the explosion-produced cavity and decay there.
2. Plutonium production reactors. The fuel from these reactors will be chemically reprocessed to recover the plutonium. There is no indication in the literature that any country is controlling the krypton-85 produced by this activity. Recent measurements of atmospheric krypton-85 suggest that worldwide plutonium production rates are down from those of a few years ago.
3. Military and research reactors. The spent fuel from these reactors is often reprocessed because the highly enriched uranium they use is too valuable to discard. The scale of this effort is small and krypton-85 controls are not presently used.
4. Civilian nuclear power. Only a small portion of the spent fuel from civilian nuclear power reactors has been removed from the fuel cartridges and reprocessed. Consequently, the krypton-85 contained in these fuel elements constitutes a major reservoir of this isotope. After 75 years of storage, less than one percent of the original krypton-85 will remain. At present, the United States is enforcing a moratorium on civilian nuclear fuel reprocessing. There will also be a small percentage of failed fuel elements leaking krypton-85 and other isotopes. These materials will be disposed of either by further incapsulation or by reprocessing. At the design stage of a nuclear fuel reprocessing plant there is the opportunity to design and install an effective krypton capture system. Once a plant is in operation, the off-gas system will be contaminated with radioisotopes, notably ruthenium, and changing of the system to accommodate a krypton-85 capture system may not be feasible. A properly operating krypton capture plant should remove 99% of the krypton from the nuclear fuel reprocessing plant. However, to achieve a 99% reduction of the annual output, the krypton capture system must not malfunction in a year of continuous operation. It is more reasonable to expect a system to be down (nonfunctional) less than 10% of the year. Thus, an operational system could be expected to remove between 90 and 99% of the krypton-85 in the reprocessed nuclear fuel. This implies that a nation operating a nuclear fuel reprocessing plant without krypton-85 controls will be releasing

from 10 to 100 times the amount of krypton-85 released by plants with mandatory controls.

Decision Characteristics

Because the same process that produces krypton-85 also produces plutonium, any decision affecting krypton-85 controls will be made at the national level, on a nation-by-nation basis. These decisions will probably be made semi-independently of one another. Decision-makers in nations with nuclear capabilities will certainly know of other countries' decisions regarding krypton-85 controls. They will not necessarily make the same decisions.

The basic premise is that decision-makers wish to know the consequences and/or risks associated with each available course of action. Continuation of the status quo is often one of the available courses of action. The decision-makers attempt to choose a course of action that leads to an acceptable level of risk at an acceptable economic and social cost. The basis for comparison, in any case, should be the status quo, with reasonable projections for future events. This choice is justified by the observation that indecision or postponement of a decision is equivalent to a deliberate choice of the status quo.

For those decisions involving a choice between two alternatives, there will be a threshold for action. The status quo will be maintained unless there is an unacceptable risk resulting from that course of action and there is an alternative course of action that reduces the risk at an acceptable social and economic cost.

The desired output of the decision-making process is the correct choice between two alternatives. This is called an error-free decision. The projections on which an error-free decision can be based do not have to be perfect, only accurate enough to choose correctly between two alternatives. In particular, those aspects of the prediction that could swing a decision from one alternative to another deserve the most concentrated attention.

C. An Analytical Model for Fair Weather Atmospheric Electricity

The reasons to focus attention on the interpretation of atmospheric electrical measurements include their potential use as a global monitor of certain aspects of meteorological phenomena. The universal time variation is good evidence that the fair weather atmospheric electric field responds rapidly to events around the globe. Fair weather atmospheric electricity shows modulations synchronized with solar phenomena of both periodic and nonperiodic types. The existence of these

correlations highlights a gap in our present knowledge of the interrelations between cloud physics and atmospheric electricity.

A solution has been obtained for Maxwell's equations for time variations with a range of characteristic periods from a few seconds to many years and for regions of the troposphere and stratosphere that are free of current sources. This corresponds to regions where fair weather atmospheric electricity measurements are made. The results are given in terms of a harmonically analyzed separable solution in spherical coordinates for an exponential conductivity profile with boundaries in the stratosphere and on the surface of the earth. A first-order analytical solution has been found for global-scale phenomena with characteristic lengths of more than 15 degrees of latitude. The temporal and spatial variations of the electric field at the boundaries determine the coefficients of the solution. The solutions is sensitive to the ratio of conduction current to displacement current. At low altitudes for short periods, the displacement current dominates. In this region, upward-propagating electric fields are essentially unattenuated below the altitude where conduction currents are dominant. In the conducting region, which lies above the other region, electric fields are exponentially attenuated. This is consistent with a model of a dielectric atmosphere surrounded by a conducting shell, provided that the thickness of the dielectric layer depends on frequency. The dielectric region is nonexistent for field variations with periods longer than a day.

The magnitude of the fair weather atmospheric electricity field is determined by the balance between the integral of all processes removing charge from the earth and the returning currents. The relationship between cloud processes, lightning and charge transfer from the earth needs further investigation.

D. Dipole Model for Atmospheric Electricity

This theory treats the atmosphere as a polarized dielectric with electric and polarization vector fields. Current flow will be described as a polarization current rather than a conduction current.

In the dipole model, the atmosphere consists of dipoles that are formed as ion pairs drift apart under the influence of the atmospheric electric field. The dipole moment of each set of ion pairs is given by the quantity of charge (1 electronic charge) times the distance of separation. The direction of these dipoles will be such as to produce a depolarizing field with the net effect of reducing the initial polarizing field in a manner identical to a discharging current.

Some ions are not paired with atmospheric ions but rather with

induced charges on the surface of the earth. In this case, the dipole extends from the surface of the earth to the singular charge. The dipole model inherently conserves the total charge of the atmosphere-earth system.

Figure 2 represents some aspects of the dipole model. Panel 1 of Figure 2 illustrates a conventional model for atmospheric electricity. Positive charges are shown above the conducting surface of the earth. The earth's surface has an induced negative charge. The electric field in this and the other panels is oriented toward the earth. Panel 2 of Figure 2 shows a number of ion pairs represented as dipoles orientated in the direction of the field. The left side of panel 3 shows that singular charges produce dipoles orientated away from the earth. These can be treated as the source of the ambient electric field. Panel 3 also shows ion pair dipoles which act to depolarize the dielectric atmosphere.

The ionic polarization current transmits current in the direction of the electric field through a series of discrete steps. Panel 2 of Figure 2 illustrates this point. Only ion pairs that will ultimately interact are illustrated. Three ion pairs are generated at time $t = 0$ in a vertical electric field. Before $t = 0$, there is downward electric field but no ion pairs and no downward directed dipole moment. Shortly after time zero, there are three dipoles orientated with the field direction. Each pair has separated only a small distance and the total dipole moment per unit volume is still negligible. As time progresses the middle pair neutralizes one of the ions of the upper and lower pairs in Figure 2 panel 2. The intermediate charges disappear and finally the dipole moment has the value of unity charge times the separation distance at time t . As they continue to drift with the field, or encounter other ions, the process transfers positive charge in the direction of the electric field, representing an equivalent current. The electric field due to the dipole is a depolarizing field that reduces the driving electric field in the same way a discharging current decreases the field. Individual ion pairs drift only tens of meters before encountering other ions. This distance is less than the tens of kilometers between the earth's surface and the conducting regions of the upper atmosphere. Clearly a large number of bombardment induced transient ion pairs must participate in the transfer of a single positive charge from the upper atmosphere to the earth. The process is sufficiently different from metallic conduction that it should be described as a bombardment induced ionic polarization current.

The basic equations for the dipole model are an extended

set of Maxwell's equations that include terms for the non-electromagnetic sources of fields and currents. The absence of "true space charge" and "true currents" in this model provides some simplification of the equations for the fields. The electric field, E , includes the conservative part of the field as well as a part due to electromagnetic induction. Ohm's law takes the form

$$\frac{dP}{dt} = \sigma_{\text{eff}} E + I \quad (1)$$

where $\frac{dP}{dt}$ is the polarization current,

σ_{eff} is the effective ionic dipole conductivity, and I represents various currents produced by non-electromagnetic sources. The atmospheric polarization vector field, P , is given by

$$P = P_C + P_I \quad (2)$$

where P_C is the polarization vector corresponding to the ionic dipole current, and P_I is the polarization vector corresponding to the currents of nonelectromagnetic origin.

Faraday's law of induction is extended to include non-electromagnetic sources:

$$\oint_C E \cdot dl = - \frac{d}{dt} \int_S u_0 H \cdot dS = \oint_C \frac{dP}{dt} \cdot \frac{dl}{\sigma_{\text{eff}}} - \oint_C I \cdot \frac{dl}{\sigma_{\text{eff}}} \quad (3)$$

the symbol $\oint_C E \cdot dl$ represents a contour integral of the vector E along curve C and the symbol

$$\iint_S H \cdot dS$$

represents the surface integral of vector H over any surface bounded by the curve C .

Ampere's law takes the form

$$\oint_C \mathbf{H} \cdot d\mathbf{l} = \iint_S \epsilon_0 \frac{d\mathbf{E}}{dt} \cdot d\mathbf{S} + \iint_S \sigma_{\text{eff}} \mathbf{E} \cdot d\mathbf{S} + \iint_S \mathbf{I} \cdot d\mathbf{S} \quad (4)$$

The equation for the Poynting vector is given at

$$\oint_S \mathbf{E} \times \mathbf{H} \cdot d\mathbf{S} = - \frac{\partial}{\partial t} \iiint_V \frac{1}{2} (\epsilon_0 \mathbf{E}^2 + \mu_0 \mathbf{H}^2) dv$$

$$- \iiint_V \left[\frac{dP}{dt} \right]^2 \frac{dv}{\sigma_{\text{eff}}} = \iint_S \mathbf{I} \cdot \frac{dP}{dt} \frac{dv}{\sigma_{\text{eff}}}$$

where $\iiint_V Z dv$ is the volume integral of the scalar Z over the volume V and

$\oint_S \mathbf{T} \cdot d\mathbf{S}$ is the surface integral of the vector $\mathbf{T}$ over the surface S of the volume V .

II. Significant Publications

A number of publications in the last year contributed to the background information relating atmospheric ionization to cloud physics.

In 1978, an English translation of symposium entitled "Atmospheric Electrical Effects of Krypton-85 Released into the Environment"¹ was made available through the efforts of James Hughes of ONR. The symposium was held in December 1976 near Tokyo, Japan and was previously only available in Japanese. Copies of the 180 page proceedings are available from the author.

There were a number of publications related to the effects of atmospheric electrical field or charges on cloud microphysical processes.

A significant review volume "Microphysics of Clouds and Precipitation" was written by Pruppacher and Klett.² This volume contains a review chapter on the electrical state of the atmosphere and its effects on cloud microphysics.

A recent paper by Freire and List³ calculates the collision enhancement for droplet pair with electrically reduced approach speed. The authors conclude, "The magnitude of the collision efficiencies calculated here, together with the past work of Schlamp et al., among others, leaves no doubt that electrical effects involving vertical electrical fields offer a very powerful explanation for the fast growth of small cloud droplets beyond the size at which condensation ceases to be effective. To neglect electrical effects in the natural growth of small cloud droplets is no longer justified." These calculated phenomena occur at relatively low electric fields and extend into the range of fields expected inside clouds in an ambient fair weather electric field.

Several other papers have been written on closely related topics^{4,5}.

A paper by Spangler and Rosenkilde⁶ presents calculations of the effect of radioactive aerosol particles resulting from fresh debris from a nuclear explosion on a simple infinite cloud thunderstorm model. They conclude that very large ionic currents would prevent significant electrification of their cloud model. The ionization rates are many orders of magnitude greater than the natural background ionization rate. That result was anticipated by others. The results of this simple modeling experiment have little bearing on the evaluation of the more subtle cloud physical phenomena addressed by Pruppacher and others.

III. Data Analysis

The reduction of the data from strip charts to numerical values is far behind schedule. Only a few months of Mauna Loa data have been reduced and analysis of such a small data sample has not been attempted. Computer programs for file management were prepared.

IV. Measurement Program

With the active cooperation of the staff of Mauna Loa Observatory atmospheric electrical measurements were continued for the period of this contract. The electric field, air-earth current, positive and negative electrical conductivity were measured. Several instruments were repaired, maintained and/or calibrated this year.

FOOTNOTES

1. H. Ishikawa, Ed. Atmospheric Electrical Effects of ^{85}Kr Released into the Environment, Atmospheric Electrical Society, Tokyo, Japan, 1976. (Translation available from W. Boeck, Niagara University, N.Y. 14109)
2. H. R. Pruppacher and J. D. Klett, Microphysics of Clouds and Precipitation, D. Reidel, 1978.
3. E. Freire and R. List, "Collision Enhancement for Droplet Pairs with Electrically Reduced Approach Speed," J. Atmos. Sci., 36, 1777-87, 1979.
4. R. J. Schlamp, S. N. Grover, H. R. Pruppacher and A. E. Hamielec, "A Numerical Investigation of the Effect of Electric Charges and Vertical External Electric Fields on the Collision Efficiency of Cloud Drops. Part II," J. Atmos. Sci., 36, 339-349, 1979.
5. F. Herbert, "A Theoretical Model to Describe the Motion of Aerosol Particles Due to the Combined Action of Inertia, Brownian Diffusion and Phoretic and Electric Forces," J. Atmos. Sci. 1744-50, 1978.
6. J. D. Spangler and C. E. Rosenkilde, "Infinite Cloud Model of Electrification by the Precipitation Mechanism in the Presence of High Rates of Ion Generation," J.G.R., 3184-90, 1979.

Figure I.

Figure 2.

Panel 1. Atmospheric ions and induced charges.

Panel 2. Ion pairs illustrated as dipole.

Panel 3. Atmospheric ions and induced charges illustrated as dipoles.