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Evaluating a Fluorosis Hazard after a Volcanic Eruption

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ABSTRACT. The August, 1991 eruption of Mt. Hudson (Chile) deposited ash across southern Argentina and contributed to the deaths of thousands of grazing sheep. Early ash analysis revealed high levels of fluoride, a potential ash constituent toxic to humans and animals. In order to evaluate fluorosis as the cause of sheep deaths and to examine the possibility that similar ash and airborne toxins could also have an effect on the human population, we conducted an investigation that included health provider interviews, hospital record review, physical examination of sheep, determination of sheep urine fluoride levels, and complete constituent analysis of ash samples collected at proscribed distances from the volcano. Ash deposited farthest from the volcano had highest fluoride levels; all fluoride measurements were normal after rainfall. There were no signs or symptoms of fluorosis observed in sheep or humans. Sheep deaths resulted from physical, rather than chemical properties of the ash.

EXPOSURE to excessive amounts of fluoride may cause adverse health effects for humans and animals. Recently, improper fluoridation of a city water supply was associated with human death in Alaska¹ and, in Idaho, the fluoride content of grazing pastures exposed to industrial pollution is being monitored for a possible long-term effect on animal health.^{2,3} The plume dispersed by winds after a volcanic eruption contains volcanic ash that may also be a source of fluoride at levels that are potentially toxic.^{4,5} Given that grazing animals are more likely to be exposed to such ash, acute adverse

health outcomes for these animals may be a sentinel to fluoride contamination, which would ultimately manifest itself as chronic human fluorosis.

Background

On August 12, 1991, Mount Hudson, located in southern Chile, approximately 100 kilometers (km) from the Argentine border, began a 3-d cycle of eruptions that sent a column of ash 18 km into the air. Hudson's previous eruption, exactly 20 y earlier, had produced an ash column 7 km high and had extruded lava that melted glacial ice; this melting triggered large mudflows that led to the deaths of 11 people and numerous animals.⁶ During the recent eruption, winds from the northwest car-

ried ash over Santa Cruz province, resulting in ash deposition as far away as the Falkland Islands.

Approximately 50 000 people were clustered in several towns along the periphery of the volcano impact zone (the area of greatest ash accumulation). Although no volcano-related human deaths were recorded, Argentine news media reported that massive numbers of sheep were dying and predicted that hundreds of thousands of the 2.5 million sheep pastured in the volcano impact zone would die from poisons carried by the ash (especially fluorides) and from gastrointestinal obstruction after ash ingestion.^{7,8}

According to newspaper and government reports, the people of Santa Cruz feared contamination of their food and water supply and loss of the wool production industry. Some speculated that the deaths, caused by toxic ash, among the more exposed animal population were sentinel events for chronic health effects that would eventually affect exposed humans in the ashfall area.

Early ash analyses in Argentina determined that levels of fluoride, considered to be the most toxic ash constituent, were elevated.⁶ Moreover, researchers hypothesized that, geographically, fluoride from the ash would be the most widely distributed toxic agent because it adheres to the smallest particles that the wind transports. In addition, the initial effects of the ash were compounded by typical spring winds of 100–150 km/hr and by the drought that the affected area of Argentina was experiencing for the third year.

A cooperative investigation between the Argentine Ministry of Health and the Centers for Disease Control and Prevention (CDC) was begun 1 mo after the eruption to evaluate fluorosis as the possible cause of sheep deaths, to examine the possibility that similar ash and airborne toxins could also have an impact on the health of the human population, and to assess the likelihood of long-term ash-related effects on humans, animals, and plants.

Method

During the week of September 16, 1991, researchers conducted a field investigation that included interviews and medical record reviews, clinical assessments of sheep, and laboratory analyses of ash constituents and sheep urine fluorides. Open-ended interviews were conducted with local physicians and hospital staff, veterinarians, ranchers, municipal government and civil defense personnel, and national agricultural representatives. The health status of sheep was evaluated through direct physical examination of animals that appeared to be ill and by analyzing urine samples for fluoride concentrations. Evaluation of the remaining sheep was limited to clinical field observations. The range of ash exposures was assessed by collection of ash samples at 50-km intervals along Highway 281, which runs between the border of Argentina and Chile and the Atlantic coast.

Samples were collected between September 18 and 21, more than 4 wk after the last recorded ash fall; in four instances we attempted to obtain samples of ash that had remained undisturbed (virgin ash) from horizontal sur-

faces (on buildings and natural rock formations) that were protected from direct wind. We collected other ash samples from around the bases of plants and also brushed samples from the leaves of plants that animals used for forage. Seven samples, presumably collected during the initial ashfall period and donated by local public health authorities, were also analyzed by the National Institute for Occupational Safety and Health (NIOSH). In Table 1 are enumerated the various parameters of ash that were analyzed, which include inorganic anions and heavy metals as well as particle sizing and determination of radioactivity levels.

We evaluated bioavailable fluoride using two extraction methods. NIOSH Method 7903,⁹ a procedure that uses a weak carbonate-bicarbonate eluent for extraction, was used to analyze for fluoride and other anions. This method is believed to reflect the amount of fluoride that would be available in ash-contaminated water sources. In addition, the Environmental Protection Agency's Procedure #1311, which uses acetic acid extraction, was used to simulate an *in vivo* sample (i.e., the potential for release of toxin by stomach acids after ash ingestion). The Argentine Ministry of Health laboratories in Buenos Aires analyzed sheep urine fluorides ($n = 5$ pooled samples) for fluoride concentration.

All ash samples underwent particle sizing and fiber identification, using polarized light microscopy. We used stereomicroscopy to size the coarsest particles and then representative portions of each sample were immersed in Cargille liquids, examined microscopically, and were sized according to a calibrated scale in the eyepiece. Analysis for free silica was performed by Data-Chem Laboratories, using NIOSH method 7500⁹ to quantify quartz and cristobalite.

Radioactivity was measured using a gross alpha- and beta-counting system that had been calibrated previously for efficiency by using a strontium-90 beta source and a plutonium-239 alpha source. Gross

Table 1.—Potential Ash Constituents That Were Evaluated in Ash Samples Collected September 1991, Vicinity of Mt. Hudson Eruption, Chile and Argentina

Fluoride*	Cobalt	Silver
Sulfate	Chromium	Sodium
Chloride	Copper	Tin
Nitrate	Iron	Tellurium
Phosphate	Lithium	Thallium
Quartz	Magnesium	Titanium
Christobalite	Manganese	Tungston
Aluminum	Molybdenum	Vanadium
Arsenic	Nickel	Yttrium
Barium	Lead	Zinc
Beryllium	Phosphorus	Zirconium
Calcium	Platinum	Fiber, size
Cadmium	Selenium	Radioactivity

Notes: Analysis conducted by the Centers for Disease Control and Prevention (CDC); the National Institute for Occupational Safety and Health (NIOSH); the Division of Physical Science and Engineering (DPSE), Cincinnati, Ohio; and by DataChem Laboratories, Salt Lake City, Utah.

*NIOSH method 7903 and EPA Procedure 1311.⁹

gamma-emitting isotopes were measured on a spectrometer that had been calibrated previously.

Results

Humans. Reports from physicians, clinic workers, and the region's hospital administrator did not suggest an increase in lower respiratory tract infections, e.g., pneumonia, among inpatients, or an exacerbation of reactive airway diseases such as asthma in susceptible individuals during the 1 mo following the eruption. Interview data confirmed that complaints of upper respiratory and mucous membrane irritation, manifested by dry cough and watery eyes, were common. Such minor irritant complaints were attributed to constant resuspension of fine ash by strong winds. On the basis of data from September, the number of outpatient visits and hospitalizations for pulmonary diseases following the eruption did not exceed pre-eruption levels of visits and hospitalizations. Health care providers observed that neither smokers nor children—two potential high-risk groups for respiratory sequelae—were represented disproportionately among those seeking medical care.

Donated and homemade face masks and goggles were worn extensively during the first few weeks. Several medical providers and patients reported adverse psychological symptoms, including feelings of uncertainty and loss of control. These symptoms appeared to be occurring particularly in areas near the Chilean border where ash depth reached 10 to 15 cm and in coastal communities where there was no significant ground accumulation, but where fine ash was constantly being resuspended by high winds. There were no human deaths attributed to ash-related effects.

Animals. Observation and clinical examination of sheep did not detect any signs or symptoms of fluorosis (e.g., lameness or diarrhea). All the dead and critically ill sheep observed were ewes located in areas where the ash covered much of the vegetation. Most of the sheep were dehydrated and thin; many pregnant ewes appeared to be near term. No evidence of ocular lesions (suggesting ash-induced corneal damage that would impair ability to locate food) or dental involvement (indicative of chronic fluorosis or ash abrasion) was observed. Our examinations revealed no ash in oral cavities, indicating that the animals did not attempt to consume the ash. Beneath heavy winter coats these sheep were quite thin. We could not approach free-grazing sheep for examination, but could only observe them for general signs and symptoms of fluorosis, such as diarrhea, poor appetite, diminished alertness, or abnormal gait. We did not observe any of these diagnostic signs or symptoms. Lambs with newly erupted teeth lacked dental lesions that are consistent with fluorosis (e.g., mottling and discoloration).

Veterinarians in Perito Moreno, Argentina, located 120 km from the volcano, had performed necropsies on an undetermined number of sheep and reported no large accumulations of ash in either the respiratory or gastrointestinal tract and only small amounts in the rumens of several animals. We found no evidence of significant mechanical obstruction of the gastrointestinal tract in the

Argentine animals, a finding similar to that of studies conducted following the eruption of Mt. St. Helens', in 1980.⁹ Instead, sheep deaths were attributed by local Argentine veterinarians to enterotoxemia and ketosis.

We observed and, in some cases, examined other species, including cats, working dogs, goats, and various domestic or wild birds. None of these species manifested evidence of fluoride toxicity.

Fluoride concentrations of four pooled urine samples ranged from 1.7 to 5.3 ppm with a mean of 3.4 ppm. These urinary concentrations are within the normal ruminant range of 1–8 ppm reported in other field studies.¹⁰ Only one pooled sample, collected 100 km from the volcano in Los Antiguos, had an elevated fluoride level of 15 ppm, which is less than the 20 ppm considered the upper limit of normal by other reference texts.¹¹

Ash. Laboratory analyses for the constituents listed in Table 1 indicated that most of these substances were nondetectable or below the limit of measurement. The amount of free silica, the causative agent in silicosis, was minimal. Glass was the only fiber identified by microscopic examination. Table 2 gives measurable concentrations of the remaining constituents. These values are all within the normal range of amounts found in crustal rock.¹² Gross alpha, beta, and gamma levels were higher than similar measurements made in the past at other volcanic sites, but these levels probably reflect the high normal background radiation level for this area.^{14,15}

Fluoride levels, measured by the U.S. Environmental Protection Agency's (EPA's) and the National Institute for occupational Safety and Health's (NIOSH's) analytical methods, were below established toxic levels. Both the early samples and those collected 1 mo post-eruption are plotted in Figure 1, according to their distance from Mount Hudson. Fluoride levels in ash samples collected shortly after the eruption increased in direct proportion to the distance the ash was ejected from Mt. Hudson. Among samples collected later, the sharp decline in fluoride concentration, beginning at about 200 km from the volcano, reflects the leaching effects of a heavy rainfall at that location on September 18.

In Figure 2 are shown the fluoride levels of both sample series, compared with ash particle size (ash size varied from < 2.5 μm to > 1 mm in diameter). Fluoride levels are highest for the smallest particle sizes in both series. Moreover, we observed an inverse relationship between particle size and the distance the ash was ejected from Mt. Hudson (Fig. 3). Therefore, smaller particles, which were ejected farthest, contained the most fluoride. For example, at the sample site most distant from the volcano (550 km), we observed that the smallest particle size (< 2.5 μm) had the highest fluoride concentration (52 ppm).

Discussion

Fluorosis. Fluorosis in animals after volcanic eruptions was observed as early as 1694 in Iceland.^{4,16} The first evidence that Andes volcanoes could produce fluoride-laden ash occurred in Lonquimay, Chile, in 1988.¹⁰ Fluoride is considered an essential element for human, animal, and plant health; however there is no

Table 2.—Analysis of Quantifiable Ash Constituents from Mt. Hudson, Chile, Collected in Santa Cruz Province, Argentina, 1991

Constituent (ppm)	8/12–8/20 (7 samples)		9/17–9/18 (13 samples)	
	Mean*	Range	Mean*	Range
Fluoride†	22.7	ND-52	16	ND-28
Fluoride‡	11	ND-25	9	ND-16
Sulfate	578	64–970	395	5–2 200
Chloride	115	13–240	96	3–450
Aluminum	1 886	200–4 100	7 175	1 100–14 000
Barium	10	2–16	40	2–130
Calcium	2 970	420–3 600	8 592	1 500–40 000
Iron	3 564	550–6 300	9 100	2 100–20 000
Magnesium	1 690	200–3 300	2 668	510–6 000
Manganese	83	13–160	176	45–330
Phosphorus	437	130–650	390	260–710
Sodium	1 579	550–3 200	1 216	460–2 000
Titanium	274	37–440	579	180–1 100
Vanadium	9	6–12	24	5–51
Zinc	7	5–10	18	4–39
Gross alpha§	11.7	6.1–15.0	10.3	5.8–18.7
Gross beta§	37.3	31.0–42.7	38.2	30.4–51.7
Gamma§	1.7	1.3–2.1	1.2	0–2.1

Note: ND = not detected.
 *Mean of all samples within limits of detection.
 †EPA Procedure #1311.
 ‡NIOSH Method 7903.
 §Picocurie of radioactivity/gm of ash (pCi/gm).

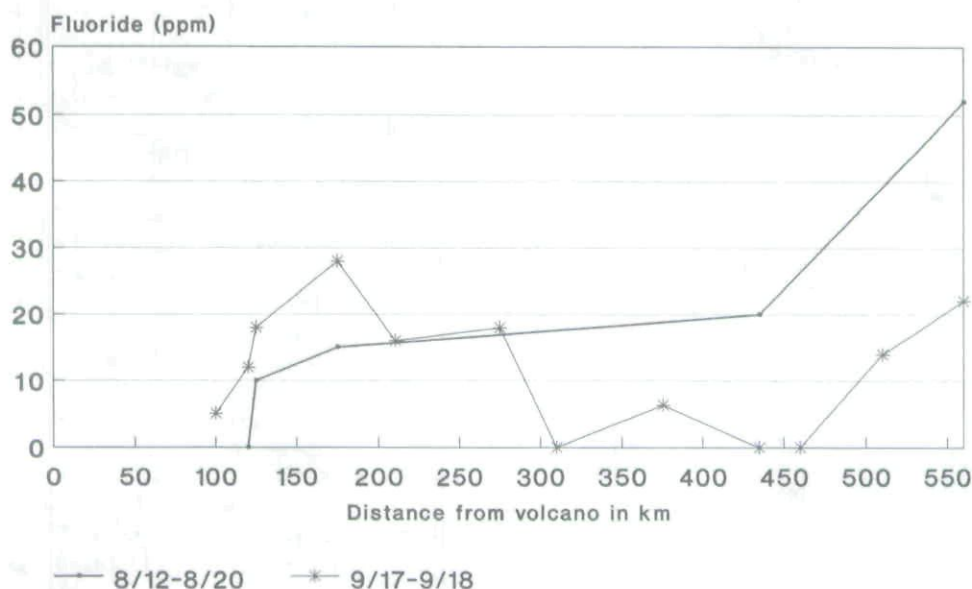


Fig. 1. Fluoride content of volcanic ash, according to distance the ash fell from Mt. Hudson, Chile, 1991.

record of a natural environment so lacking in fluoride as to cause deficiencies in grazing animals.¹¹ A single dose of 50–70 mg fluoride/kg body weight of a soluble fluoride or the ingestion of 6–20 mg fluoride/kg body weight for several days may result in acute toxicity in cattle. An average-sized cow that ingested Mt. Hudson ash would need a daily dietary intake of more than 30 kg of the most concentrated ash in order to manifest acute fluorosis. Sheep may be even more tolerant of dietary

fluoride. Research results show that a total dietary intake of 60 mg/kg of fluoride can be ingested by breeding ewes, and feeder lambs can ingest 150 mg/kg with no adverse health consequences.¹⁷ When these levels are exceeded, either through inhalation of gases containing organic fluorides or ingestion of excess fluoride, fluorosis can occur as either an acute or chronic toxic syndrome.^{18–20}

Humans and animals exhibit similar signs and symp-

toms of fluoride intoxication, resulting from respiratory or intestinal exposure. Acute inhalation exposure usually causes reversible nasal and ocular inflammation, but may lead to death from pulmonary edema or cardiac arrhythmias. High oral doses may cause hemorrhagic gastroenteritis and death. Chronic poisoning in humans and animals can occur similarly, regardless of the route of exposure. Mottling of tooth enamel is the most sensitive indicator of chronic fluorosis; teeth that are just forming will react to as little as 2 ppm ingested per d. Experiments have shown that ingesting 60–120 mg/d, or as little as 10 ppm, for 3 y has led to dental fluorosis in sheep.²¹ Other chronic manifestations include gastric irritation with vomiting or diarrhea, skeletal changes ranging from increased bone density to exostosis of the long

bones, and gait impairment.²² Long-term exposures may also lead to interstitial nephritis, paresthesia, paresis, and convulsions.¹⁹ Endemic chronic fluorosis that is dependent on geographic location has been reported in humans and animals.²³ Fluoride continues to accumulate in the body as long as excessive ingestion or exposure continues to occur.³

Exposure potential

Potential for fluoride exposure is a reflection of dose (e.g., fluoride concentration and ash depth) and duration of exposure (e.g., leaching effect of rainfall, redistribution by wind), as well as nutritional status and kidney function. As our results demonstrate, concentration was

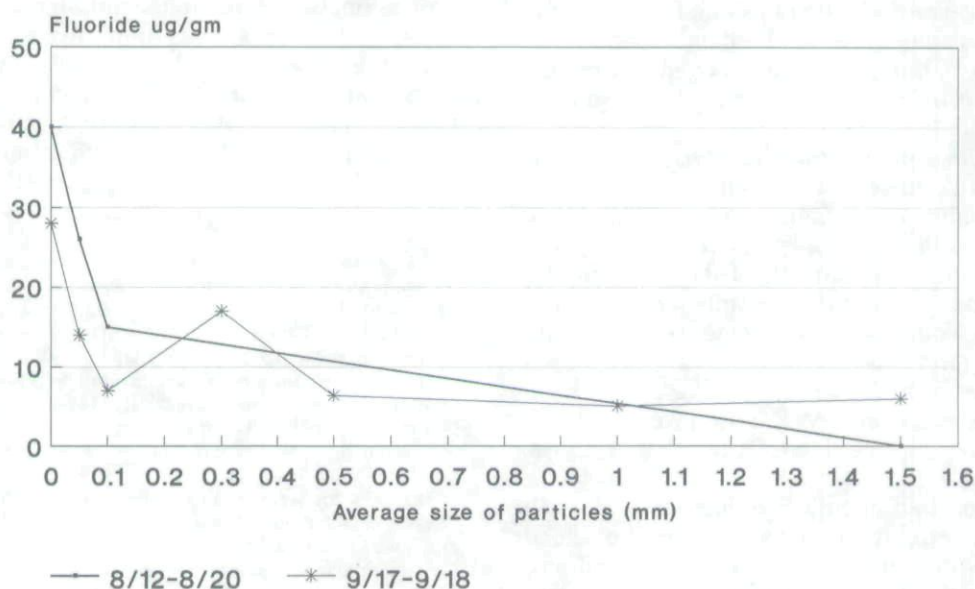


Fig. 2. Fluoride content, according to ash particle size, Mt. Hudson, 1991.

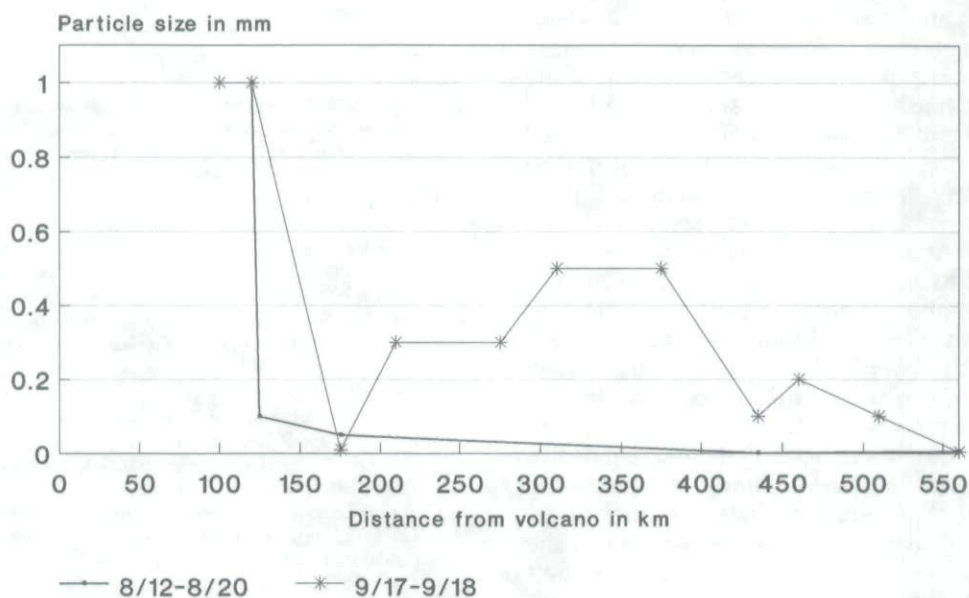


Fig. 3. Average size of ash particles, according to distance from Mt. Hudson, 1991.

maximal when the ash depth was less than 0.1 cm. Routine soil samples, collected randomly, could easily miss ash that had drifted into the leaves of the low-lying types of plants favored by grazing sheep. Soil analyses that fail to consider this drifting may underestimate the fluoride exposure of sheep as well as the increased potential for plant uptake of toxic ash constituents. Plant roots can absorb fluoride from soil with varying degrees of efficiency and then concentrate it in their leaves or absorb it directly through their leaves. Therefore, measurements of soil fluoride levels alone will not predict plant fluoride concentrations.²⁴ In vivo concentrations of less than 100 ppm will not usually adversely affect the external appearance of the plant.¹⁶

The potential to be exposed to fluoride through contact with Mt. Hudson ash was highest during the immediate post-eruption period. The rains that fell during the time investigators were in the field not only leached fluoride from the ash, but physically removed it from leaf surfaces where it may have been ingested by grazing sheep.

Given the findings of this study (i.e., absence of clinical signs of fluoride toxicity, normal mean urine excretion of fluoride, and concentrations of potentially toxic ash constituents within normal limits), it is unlikely that fluorosis caused the large numbers of sheep deaths. If a toxic element such as fluoride had adhered to the ash particulates, one would expect subsequent exposure and adverse effects from fluoride to be maximal at ash deposition sites farthest from the volcano; however the sheep that died after the eruption of Mt. Hudson were pastured nearest the volcano, in the deeper ash fall areas of Argentina.

Physical, rather than chemical properties of the ash were more likely responsible for the thousands of sheep deaths. Indeed, as observed in past volcanic eruptions, fluorosis has been falsely blamed for animal deaths that were actually the result of poor herd management practices.²⁵ Ranchers reported that the winter of 1991 was particularly severe in the Mt. Hudson area, with especially cold temperatures and unusually deep snowfalls. Given the economically depressed wool market, a higher than usual population of sheep was alive at the beginning of this harsh winter, and sheep were forced to forage on land that had been significantly overgrazed during the preceding 3 drought y. Deaths of sheep during normal winters are usually not systematically recorded in Argentina, and such losses may vary greatly among flocks. Informal reports of previous winter losses ranged from 10% to 30% of flock size. Estimates of mortality rates since the eruption have varied from 15% to 35%. These animals were chronically malnourished, and when the ash covered their already limited food sources, many probably succumbed to starvation or, among the pregnant ewes, to toxemia.

Given this scenario, it is unlikely that the sheep deaths were sentinel events for adverse human health effects. However, because the animal population is much larger and more exposed than the human population in the volcano impact zone, animals are the logical sentinels for adverse health outcomes in humans. The use of animal herds as sentinels is most successful when adequate

pre-disaster and post-disaster surveillance systems, which collect information on production parameters (e.g., milk, meat, and wool output) and reproduction patterns, are in place.

Team approach to health risk assessment

At present, there is a high likelihood that future volcanic activity will occur in the Mt. Hudson area.²⁶ Assessing health risk and allocating disaster relief resources are best handled through a team approach that includes preparing for, rather than simply reacting to, the emergency.²⁷ This investigation of fluoride toxicity and other studies of volcanic effects can be accomplished most effectively through a team approach that uses volcanologists, geophysicists, physical scientists, and public health professionals. Veterinarians and animal health officials are essential components of this team when large numbers of dead or ill animals disrupt the safety or economy of the food chain or when the effect of a volcanic eruption on an animal population may be a sentinel event for subsequent adverse human health effects.

* * * * *

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Use of trade names is for identification purposes only and does not constitute endorsement by the Public Health Service or the U.S. Department of Health and Human Services.

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