

## Zinc Cadmium Sulphide Dispersion Trials

Report by the Academy of Medical Sciences to the Chief Scientific Adviser, Ministry of Defence on the Zinc Cadmium Sulphide dispersion trials undertaken in the United Kingdom between 1953 and 1964.

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## Executive Summary

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1. We have investigated the possible effects on health of the dispersion of Zinc Cadmium Sulphide particles as a simulator of biological warfare agents. These dispersion tests took place between 1953 and 1964. We estimate that, during this period, some 250 kg of this material was dispersed from the land based sites listed in Table 4, (mainly RAF Beaulieu airfield and Porton Down); and up to a further 4600 kg from ships and aircraft mainly over BDE Cardington, Bedfordshire and over the English Channel and the North Sea.

2. Similar tests were performed in the United States. The US National Research Council (NRC) published, in 1997, an extensive report on their tests, which came to the conclusion that no health hazard had arisen there.

3. We have reviewed the NRC report and concluded that their study is essentially applicable to the United Kingdom. However, more recent published work indicates that even their extremely low estimate of possible lung cancer risk from ZnCdS inhalation is exaggerated.

4. We have also reviewed the extensive studies made at Shipham in Somerset where there has been substantial contamination with cadmium from old mine works. This is relevant because cadmium is the component of Zinc Cadmium Sulphide that gives rise to the possibility of toxicity. The data here are reassuring, as there have been no ill health effects attributable to cadmium toxicity found at Shipham.

5. We have reviewed all the available information on the British trials and estimated levels of exposure to cadmium both immediately around the land dispersion site and widely from the dispersion from sea and air.

6. Significant soil contamination is likely to have occurred only in very restricted areas at the land dispersion sites where the dispensers were washed. These sites were not inhabited at the relevant time.

7. The cadmium released into the atmosphere from the 44 long range trials for which data are available and extrapolated to a total of 76 trials (to allow for trials with incomplete information and where the data may be missing altogether) is about 1.2% of the total release of cadmium into the atmosphere from UK industry over the same period.

8. Estimates of inhalation exposure to individuals have been calculated on the same basis. Although this cannot be done accurately, our 'worst case' estimates are smaller than the calculated exposures in the United States. They correspond in aggregate to about 10 µg cadmium over the eight years of testing. This is the amount of cadmium that is inhaled in a normal urban environment in a period of between 12 and 100 days (or from smoking a total of 100 cigarettes).

9. Of the four personnel involved in the dispersion procedures (who must have been exposed to much higher levels of cadmium than anyone in the general population) two

are known, and one believed, still to be alive and in apparently good health; and the fourth died of a heart attack in his 70s.

10. We conclude that the cadmium exposure arising from these trials did not significantly increase the level to which the population is normally exposed.

11. Although the disquiet felt by populations in Southwest England when the knowledge of this programme became public in the early 1990s is entirely understandable, the available evidence indicates that there was, in the event, no danger to health involved.

## Introduction

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During the early period of the 'Cold War', from 1953 to 1964, a programme of zinc cadmium sulphide dispersion tests was conducted by the British Ministry of Defence, who perceived that Great Britain was at risk of attack by forces from the former communist countries using biological weapons. Being an island, it was considered that discrete dissemination of toxic biological agents over the entire country might be possible, without the risk of spreading the biological agents over neighbouring countries. Studies were therefore conducted - as they were in other countries that perceived themselves to be at risk - principally the USA, Canada and Scandinavia - to determine whether small particles could be spread over large areas from aircraft or vehicle emissions. An insoluble tracer, zinc cadmium sulphide (ZnCdS) was chosen to simulate the biological agents principally because it fluoresces strongly when exposed to UV light and could therefore be detected as single particles when collected on sampling devices positioned around the country.

When information about these trials became public in the 1990s concern was expressed in both the United States and the United Kingdom about the possibility that they had given rise to health hazards. A detailed toxicological assessment of the US Army's zinc cadmium sulphide dispersion tests was undertaken by the National Research Council (NRC) in the United States and their report, which was able to reassure the American Congress on this point, was published in 1997.

The Chief Scientific Advisor for the Ministry of Defence has now invited the Academy of Medical Sciences to undertake an independent review of the zinc cadmium

sulphide trials that were carried out in the United Kingdom between 1953-64.

The terms of reference for this independent review were:

1. To review the published toxicological assessment of the US trials which was carried out to address concerns raised about their potential effects on public health. In particular, to become familiar with the scale and exposure levels of the US trials and the manner and scope of the toxicological assessment of the conclusions.
2. To consider the scale and exposure levels of the UK trials in the context of the conditions of the trials carried out in the US.
3. Based on the outcome of these reviews, to comment on whether the UK trials posed any risk to public health.
4. To provide a report to the Ministry of Defence Chief Scientific Adviser comprising:
  - A summary of the UK trials for the lay reader putting them into the context of similar trials carried out in the US.
  - A summary of the toxicological assessment of the US trials, including comment on its veracity and relevance to the UK trials.
  - Conclusions with respect to the potential health risk posed by the UK trials.

To carry out the independent review, the Academy appointed a small group comprising Professor Peter Lachmann (President of the Academy and Emeritus Professor of Immunology, University of Cambridge) as Chairman, Professor Dame Barbara Clayton (Honorary Professor in Metabolism, University of Southampton), Professor Paul Elliott (Head of Department of Epidemiology and Public Health and Head of Division of Primary Care and Population Health Sciences, Imperial College) and Dr Clive Phillips (Lecturer in

Farm Animal Production Medicine,  
University of Cambridge).

In carrying out our work we were greatly helped by Dr Rick Hall and his colleagues at DERA at Porton Down whom we visited on 29 September 1999 and who shared with us the available data on the individual trials as well as the reviews of the trials that were carried out for the Ministry of

Defence by Dr G F Collins and by Dr G B Carter. We also made contact with Dr R H Titt who was himself involved in carrying out the trials and who was able to give us information about other staff members involved. Mr C John at DERA also made available to us his model for calculating the transport, dispersion and deposition of fluorescent particles based on the SCIPUFF model published by Sykes *et al.* in 1997.

## Review of the toxicological assessment of the UK trials

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The published study "Toxicological assessment of the US Army's zinc cadmium sulphide dispersion tests" (National Academy Press, Washington 1997) is a substantial review running to more than 350 pages. Its final conclusion, given by the committee chairman, Dr Rogene Henderson, was "After an exhaustive, independent review requested by Congress we have found no evidence that exposure to zinc cadmium sulphide at these levels could cause people to become sick. Even when we assume the worst about how this chemical might behave in the lungs, we conclude that people would be at a higher risk simply from living in a typical urban, industrialized area for several days or, in some cases, for months".

Although there is great difficulty in estimating the level of individual exposure in either the US or the UK trials, it does seem clear that exposure in some large American cities (St Louis, Minneapolis and Winnipeg) was higher than in any United Kingdom town.

In the NRC assessment cadmium sulphide (CdS) was studied rather than ZnCdS. It is reasonable to accept that CdS would be more toxic than ZnCdS, which is highly insoluble. It is also reasonable to accept that it is impossible to distinguish between cadmium from the Army's tests and cadmium derived from other sources.

Cadmium accumulates in the body, especially in the kidneys and to a much lesser extent in the liver. About 5% of cadmium is absorbed through the gut. When cadmium is absorbed through the lungs or the gut it becomes bound to albumen or to the cystine-rich protein, metallothionein. In the kidneys, the lysosomes of the proximal tubules degrade

the metallothionein and so release the bound cadmium. Once there, the kidneys cannot make enough metallothionein and the free cadmium will damage them. The most sensitive indication of cadmium-induced renal damage is an increased urinary excretion of low molecular weight proteins, and particularly of beta-2-microglobulin, as there is a decrease in renal tubular absorption. There is uncertainty about the fate of ZnCdS when it is breathed deep into the lungs. It is known that cadmium ions can enter the cell surface fluid and pass into cells in the lung. Like other transition metals cadmium participates in oxidation and reduction reactions.

The NRC report includes substantial input from the public on health concerns but there is no mention of kidney disease. The main health concerns expressed in the United States related to cancer. The NRC came to the view that lung cancer was the only form of cancer where any risk was plausible. They performed a risk estimate for lung cancer and concluded that in the most highly exposed American city, St Louis, (total potential inhalation dose of cadmium 24.4µg) exposed subjects could have a life time increased lung cancer risk not more than 1.5 in a million ( $1.5 \times 10^{-6}$ ). This risk estimate is based on the risks of industrial workers exposed to cadmium by inhalation reported by Thun *et al.* (1991). There is however now available more recent

work by Sorahan *et al.* (1995) and Sorahan & Lancashire (1997) which casts substantial doubt on even these estimates. Sorahan and his colleagues found an increased lung cancer incidence in cadmium exposed workers only when they were also exposed to arsenic. Even high



exposure to cadmium alone (>13µg Cd inhaled dose per day over many years - i.e. inhaling every two days the total dose from ZnCdS in St Louis) showed no increase in lung cancer incidence - although at this level there was an increase in chronic non-malignant respiratory disease, as there was in their control group of workers exposed industrially to iron and brass.

We conclude from these data that there is really no increased risk of lung cancer attributable to inhalation of ZnCdS in either the US or UK tests.

The NRC calculated the total (time-integrated) potential inhalation doses of cadmium in the US cities affected by the tests. The highest levels were calculated to be 24.4µg cadmium (from 156µg ZnCdS) in St Louis, 14.5µg in Winnipeg and 6.8µg cadmium in Minneapolis.

Within the limits of the information we believe the NRC case was well argued and we fully accept their conclusion, as given by Dr Henderson and quoted at the beginning of this section.

This compares with an average yearly inhalation dose from other sources between about 30 and 250µg (0.1µg-0.8µg/day). Smoking adds significantly to the inhaled dose (2-4µg/20 cigarettes). The NRC therefore concluded that the ZnCdS tests did not exceed background exposure even by inhalation.

The total background intake of cadmium is much higher (12-84µg/day in urban areas). Most of this comes from food (10-60µg/day) and water (where the cadmium concentration varies from <1 to 10µg/litre).

We did not find the animal data to be particularly helpful as species' differences are always a problem. However we noted that no deaths were seen in dogs fed ZnCdS at levels as high as 10g/kg or in rats fed at a level of 20 g/kg - truly enormous doses!

## Cadmium toxicity to humans in the United Kingdom

Most of the literature on the toxicity of cadmium relates to smelting where the main route of exposure is through the lungs. For example, Belgium is the principal producer of cadmium in Europe and environmental contamination in areas near zinc smelting plants, which release cadmium into the environment, is severe. In these highly contaminated areas, there is evidence to suggest increased body burdens of cadmium among a proportion of the exposed populations, with some evidence of increased urinary excretion of beta-2-microglobulin and some loss of bone density among people with the highest urinary cadmium concentrations (Buchet et al, 1990; Staessen et al, 1999). Such a severe degree of contamination would not have arisen from the ZnCdS releases. There are also areas where extensive mining operations have led to contamination particularly affecting the soil. In the UK, the highest level of soil contamination due to former mining activities has been found in the Somerset village of Shipham. The health of Shipham residents has been extensively investigated and we consider Shipham to be a useful example to examine in regard to the possible hazards of soil contamination with cadmium. An extensive study was made in 1979 of the inhabitants' health, as well as the health of volunteers living in the nearby town of North Petherton in which there was no exposure to heavy contamination with metals. The pollution was associated with old mine workings and in more recent years the soil has been disturbed by the development of pasture. Many houses and gardens are on polluted land but the Cd in the soil is tightly bound up in minerals and is not readily available for uptake by locally grown produce.

The cadmium content of UK soils generally and those in Shipham are shown in Table 1.

The interdepartmental committee for the redevelopment of contaminated land (published 1987) gives a threshold level in the soil concentration below which no remedial action or clean up is required. The threshold for cadmium is 3-15µg/g air-dried soil. The concentration of cadmium in the 10cm of soil below a square metre of surface should not be more than 4.7µg/g. The estimated lowest-observed-adverse-effect-level (to produce acute gastrointestinal symptoms) for a single oral dose of ingested cadmium has recently been estimated at 43 µg/kg body weight (Nordberg, 1999). A 10kg child would need to eat the equivalent of an ounce of air-dried soil (contaminated at 15ug/g) to reach this dose.

There is considerable knowledge about the cadmium intake from food in the UK. Table 2 shows data from MAFF documents relating to the National Food Surveys and Total Diet Studies. Earlier determinations of cadmium may not have been as precise at these low levels compared with levels determined more recently since analytical methods have improved.

Three extensive dietary surveys were performed at Shipham in 1979. Analysis included zinc, lead, copper and mercury in addition to cadmium. Full details are given in the paper by Sherlock *et al.* 1983. Relevant data are summarised in Table 3.

### Other findings in Shipham

1. There was little or no correlation between the measured biochemical

parameters and either cadmium in soil or cadmium in the diet

2. The biochemical measures in urine were protein, low molecular weight proteins and beta-2-microglobulin. Biochemical measures in serum were beta-2-microglobulin and creatinine. All the biochemical measures were "within the normal range".

3. When the results for Shipham and Petherton residents were compared, confounding variables made it virtually impossible to find with any certainty meaningful differences between the two populations.

4. A higher body burden was found in women compared with men. It is known that absorption of cadmium from the gut rises with decreasing iron stores and it is likely that this is the explanation.

5. There was no correlation between blood pressure and cadmium exposure.

6. The health inventory for Shipham villagers showed no links with cadmium.

7. Admission to hospital, clinical effects and death certificates showed no correlation with cadmium exposure.

8. Four of the Shipham residents who were studied exceeded the provisional tolerable weekly intake of 400µg.

### **Long term follow up of mortality and cancer incidence among Shipham residents**

In view of the high soil concentrations of cadmium found in Shipham, a study of the long term health outcome of people who were resident in Shipham in 1939 was carried out, and compared with similar follow up of residents of the nearby village of Hutton. An analysis of 40-year follow-up of mortality was reported in 1982 (Inskip and Beral, 1982). A report of a further 18 years of mortality follow up of the original

1939 cohort, together with follow up of cancer incidence from 1971-1992, and a geographical study of mortality and cancer incidence, has recently been published (Elliott et al, 2000). Overall, mortality for Shipham was found to be lower than expected, and no clear evidence of health effects from possible exposure to cadmium in Shipham was found.

### **Conclusions with respect to the potential health risks in Shipham**

In Shipham therefore, even though the cadmium content of the soil was high, dietary intakes of cadmium by the villagers were generally less than double the national average and had no measurable effect on health. The cadmium in the soil was tightly bound up in minerals and not readily available for uptake by locally grown produce. In gardens, if vegetable leaves were yellow (due to zinc) the villagers did not eat them. Cadmium would be present in the leaves too.

Recommendations were made in 1979 that Shipham residents should be followed up but these studies have not shown any clear evidence of untoward effects and overall mortality is lower than expected.

In comparison with the long-term contamination in Shipham, ZnCdS was released on a limited number of occasions only. The extent of potential exposure from these limited releases is discussed in the following sections.

## The UK Zinc Cadmium

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### Introduction

Zinc cadmium sulphide is a hard, sintered, crystalline compound whose photosensitivity makes it useful for photographic purposes, and it is believed to be safe in this role (Patterson et al., 1994). Although it is insoluble in water and lipids, and only soluble in aqua regia, it is likely that it would become available in the soil with time, due to the activity of the sulphur-utilising bacteria in the soil, e.g. *Thiobacillus ferrooxidans* (Torma and Reddy, 1997). However, it might take some years for it to be released since the rate of utilisation is governed by, amongst other factors, the solubility of the compound. The immediate contamination of plant aerial material in the vicinity of the release sites is not considered to have been a serious problem as most of the releases were on airfields, which were not being used for the production of food.

The accumulation of cadmium in soil is currently governed by UK regulations only if it derives from sewage sludge (HMSO, 1989), however, this gives useful guidance on 'safe' levels that may be tolerated in soils. In the case of sewage sludge application, the maximum permitted concentration in the top 20 cm is 3 µg Cd/g soil DM, with a maximum deposition rate from sludge of 0.15 kg Cd/ha/10 years. The regulation only applies to land on which sewage sludge is being applied and it is not enforced for applications before the regulation came into force. The EU directive on which the UK regulation is based (EC, 1986) recommends the application of a more stringent maximum soil concentration (1 µg Cd/g soil DM) but this has not been adopted

in the UK, principally because the median soil concentrations in England and Wales are close to this level already (0.7 µg Cd/g soil DM, McGrath and Loveland, 1992). In addition the EU directive recommends that the cadmium concentration in the top 7.5 cm of the soil does not exceed the mean level in the top 20 cm to prevent livestock from consuming herbage contaminated with soil of high cadmium concentration.

The Ministry of Defence studies were conducted in the UK over a twelve year period, beginning in 1953, to ascertain whether long distance transport of biological agents is feasible, using ZnCdS as a tracer. The initial programme tested a number of disseminating and recording devices at ground level, followed by release of the tracer from aircraft in the later stages of the testing. Further studies investigated release from ships in the English Channel and the Irish Sea

### The land dissemination trials

The ground-disseminating trials were conducted from 1953-1964 and involved release of

between 0.35 and 9 kg of ZnCdS on each occasion from either a point source, for the testing of dissemination and recording equipment, and in the latter

stages of the programme from a moving vehicle, which simulated a stealth attack by the enemy. A total of approximately 51 trials were conducted from ground based sources, most at Porton and local airfields, with a total mass of ZnCdS liberated into the atmosphere of c. 250 kg. In the early programmes the fallout

was measured in some of the trials by placing Petri dishes in a grid around the point of emission and these programmes are summarised in Table 4.

The detailed investigation by the US National Research Council (NRC, 1997) considered that cadmium could be potentially both directly toxic (by inhalation) and indirectly toxic (through deposition on soil that subsequently became airborne during soil erosion and soil ingestion), but that soil contamination was not a problem for plants growing in the region because their roots penetrate below the top 1 cm where the cadmium would reside. However, the report does not acknowledge that cadmium may enter the deeper soil horizons by soil cultivation and that the splashing of plants with soil particles can directly lead to contamination. The American review modelled the possible inhalation of cadmium in sites prone to soil erosion and found the potential intake from airborne soil particles containing the zinc cadmium sulphide to be at maximum 0.2% of that inhaled directly, which they conclude was not a significant risk to people residing in affected areas. In most of Britain soil erosion is much less of a problem than in the US and this form of indirect exposure is therefore not believed to have been significant. We will now consider whether the dissemination of zinc cadmium sulphide by the UK Ministry of Defence resulted in a significant increase in UK soil cadmium concentrations.

### **Near distance fallout**

It is difficult to calculate the maximum soil concentration resulting from ZnCdS emissions and determine whether it is

above the legal limit for Cd in soil modified by the application of sewage sludge (3 µg/g DM in top 20 cm). The highest concentration reliably reported in the land-based trials where fallout was measured is '>20,000 µg per Petri dish' (it appears that it would not have been considerably above 20,000 µg as the concentrations in neighbouring Petri dishes were just below this level) (Titt, 1954c). It should be noted that there is some doubt surrounding the levels of cadmium reported in Field Report No. 353 (Titt, 1954 a,b), see Table 4. Calculations of maximum fallout are based on 20,000 µg /petri dish and 90 mm diameter Petri dishes. This concentration was observed only within the immediate vicinity of the dispenser in one trial at RAF Beaulieu, and equates to 110 µg/cm. Both an increase and, at more distant locations, a decrease in particle concentration in the Petri dishes were observed in all directions in front of the disseminating device, enabling a contour map to be created and proving that maximum concentrations had been recorded. A deposition rate of 110µg/cm<sup>2</sup> is considerably greater than the UK legal maximum rate of addition of Cd to soil from sewage sludge (0.15 kg/ha/10 years or 1.5 µg/cm<sup>2</sup>/10 years), but this limit does not apply to the situation reported here. Assuming a soil bulk density of 1.5 g/cm<sup>3</sup> (Brady, 1974) and a cadmium content of ZnCdS of 0.31 (based on a composition of 60% ZnS and 40% CdS (Collins,1981)), the top

20 cm of soil is predicted to contain a maximum additional 3.3 µg Cd/g DM (dry matter) as a result of the Beaulieu test. Added to the median concentration of Cd in English/Welsh soils (0.7 µg/g), the expected maximum concentration at the Beaulieu site is 4 µg/g DM, i.e.

above the legal limit for Cd in the top 20 cm of soil to which sewage sludge has been added (3 µg/g DM). If one assumes that all the Cd resides in the top 7.5 cm of soil, the concentration of 8.8 µg/g DM is considerably in excess of the recommended maximum level in this fraction. However, this is unlikely to lead to toxic effects in grazing livestock or people eating leafy vegetables grown on this site because the areas affected are so small (Table 4) and the cadmium would have become available in the soil over a number of years.

The greatest risk of ground contamination will have come from material washed off the dispenser in the field before it was returned to Headquarters. Operators were instructed that "gross contamination around the dispenser on the trailer will be brushed off and washed down before departing" (Collins and Bradley-Birt, 1954). The quantity of ZnCdS on the fan and dispenser wheel was collected and weighed and presumably discarded safely. This was typically between one third and one half of the total amount disseminated in each trial, i.e. about 300 g ZnCdS, and this amount was subtracted from the total dispensed to estimate fallout. The amount washed and brushed off the dispenser onto the ground will have been substantially less. There is likely to have been some soil contamination from material washed and brushed off the dispensers at the sites on the airfields and on the Porton Down range where the trials took place but these are not precisely identified in the reports. In the intervening 35-48 years it is expected that some of the ZnCdS will have been broken down by bacteria, and material that resisted breakdown for this period will be so insoluble as not to pose a health hazard.

A possible risk posed by the trials was to the MoD officials operating the disseminators, since the respirators may not have protected them fully against the extremely fine particles (median diameter 1.5 µm, density 4 g/cm<sup>3</sup>). However, of the four MoD personnel operating the disseminators in this series of experiments, two have survived and are in apparently good health, one died in 1994 of a heart attack, aged 73, and the final operator could not be definitively traced but is believed to still be alive. The building on the Porton Range where the ZnCdS was stored and prepared for use was identified as a possible source of contamination by MoD officials at the end of the programme and deliberately destroyed by burning (Titt, R. H., personal communication). Operators may also have been at risk from ultraviolet light exposure during the counting of ZnCdS particles, which could have damaged their eyes and any exposed skin (Moseley, 1994), but as this appears not to have happened it is not further considered.

1 RAF Beaulieu was taken over by the Forestry Commission in the late 1950's, but the central part of the airfield is still available for use by model aircraft enthusiasts.

### **Long distance fallout**

Long distance fallout resulted from both the ground-disseminating trials and from the air-disseminating trials which are discussed in detail in the next section. Dispersion tests from aircraft were conducted in the latter half of the Ministry of Defence programme of tests, with the objective of discovering whether long distance coverage of large areas of the country could be achieved. Drum

samplers on the ground, in aeroplanes and balloons detected the presence of ZnCdS particles, as well as the cascade impactors used to relate air measurements to fallout in Petri dishes referred to in the near-distance trials.

The land-based emissions were over periods from 10 minutes to one hour at rates of 30-70 g/minute. The emissions from aircraft were mainly delivered over BDE, Cardington and over the North Sea and the English Channel at 10-50 miles offshore at a height of c. 300 m at a rate of 0.5-1.5 kg/min, which was estimated to give a count of about 100 particles 250 miles away. Much larger quantities of ZnCdS were dispersed from the aircraft, typically about 100kg per trial, but ranging from around 5 to 140kg. Atmospheric mixing and fallout over sea would have reduced the rate of fallout over Britain by several orders of magnitude below that found in the near-distance measurements. If one assumes that the maximum amount of ZnCdS disseminated in any one trial (c. 140 kg) was distributed evenly across the whole country (240663 square km), the cadmium fallout rate would be only  $1.3 \times 10^{-5}$  mg/m<sup>2</sup>, i.e. several orders of magnitude less than the near distance fallout. Clearly this is not an accurate assessment of ground deposition rate, but further detailed study of soil contamination from the air and ship dissemination trials is not justified when the additional Cd load is several orders of magnitude below background levels in soil. On many occasions the maximum fallout rate on soil would have occurred at the coast when the aircraft was closest to the land (10 miles).

The extent to which people were exposed to ZnCdS particles by inhalation

as a result of these trials is considered in the next section.

### **Air and ship (long range) dissemination trials**

In addition to the land based trials, dissemination of ZnCdS by air and from ships took place over a period of about 8 years. These trials are reported in a series of Field Trial Programmes, Field Reports, Porton Notes, Porton Field Trial Reports and Porton Technical Papers. Not all the trials are recorded in any detail, while some trials appear to be reported more than once, and for others (for which field programmes were approved) no details of any trials are available (or even whether or not they took place). Information on a total of 42 trials with dissemination by air and nine trials from ships is available. Of the 42 recorded trials with dissemination by air, 29 took place over land (mainly the BDE Cardington, Bedfordshire), 11 at sea (four over the North Sea, six over the English Channel and one over the Irish Sea), and two over both land and

sea. The ship-based trials took place in the English Channel (eight trials) and Irish Sea (one). The aircraft dissemination trials that took place over the sea were done around 10-50 miles off the coast and at a height of around 300 m. The fall-out of fluorescent particles was then monitored as particle counts obtained from cascade impactors or specially designed drum impactors located at monitoring stations across the country. The trials showed that widespread dissemination over hundreds of miles occurred.

The first trial with dissemination by air was reported in September 1956 when

12 kg of zinc cadmium sulphide was released from a Lincoln 2 aircraft over Porton. The zinc cadmium sulphide powder was hand poured into a Venturi dispersing unit mounted below the aircraft. The last recorded trial with dissemination from an aircraft was carried out in March 1963, when 68 kg of zinc cadmium sulphide was released from a Devon aircraft using a mechanical feed dispenser along a 62 mile track 24 miles upwind and south-west of Norwich. Other programmes, dated 1960-64, proposed dissemination by air at an unstated location (Programme 23/60) and over Cardington (Programme 2/61, 24/62 and 10/63), Netheravon (Programme 14/63) and Norwich (Programme 2/64) but details of these trials are not available. The dissemination from ships occurred during October and November 1959 and January 1963.

### **Dissemination by air**

Available information on the trials with dissemination by air is summarised in Table 5. The table shows the report identification number, the date of the trial, the approximate amount of zinc cadmium sulphide released (where available), the location of the trial, the maximum particle count recorded and the sampling rate of the monitoring equipment (impactor) at that point, and the estimated (theoretical) inhaled dose at that point.

The amount of zinc cadmium sulphide disseminated was recorded, or could be calculated, for 35 of the 42 trials. The amount ranged from 5 to 139 kg (mean 70 kg, median 68 kg). Over all 35 of these trials, the total amount disseminated was 2,445.5 kg.

The highest particle count 2 (4,315) was found at Dorchester following a trial on 18 August 1959 over the English Channel; this involved the release of 127 kg of zinc cadmium sulphide from a Venturi operated dispenser mounted in a Valetta aircraft, with the flight path finishing close inshore at a point south of Swanage. Counts over one thousand were also recorded in trials over Cardington in August 1957 (particle count = 3,403) and November 1957 (1,070), and a trial over the North Sea and North of England (particle count = 1,591 at Silloth, which was at the start of the flight path).

The final column of Table 5 gives an estimate of the theoretical inhaled dose at the sampling point having maximum particle count, that is the dose that would have been received by a person stationed at that point during the passage of the particle cloud. To obtain this estimate, the following assumptions were made:

Volume of air inhaled of 16.6 litre/min. This is the figure used in the National Research Council (1997) report, and relates to breathing of an active adult; at rest, the alveolar ventilation (i.e., the volume of air available for gas exchange in the lungs) is around 5 litre/min.

Number of particles per gram =  $1.7 \times 10^{10}$  (Field Report 504; Porton Note 185).

Loss of 50% fluorescence of particles (Collins 1981). The loss of fluorescence occurred particularly during daylight and with more distant fallout.

Thus, for the trial on 18 August 1959, the estimated inhaled dose at Dorchester is  $[4,315 \text{ (particle count)} \times 2 \text{ (loss of$



fluorescence) x 16.6 (breathing rate)] / [20 (sampling rate) x 1.7 x 10<sup>10</sup> (number of particles per gram)] = 4.21 x 10<sup>-7</sup> g = 0.421 µg.

The cadmium concentration in air to cause lung damage in mammals is 100 µg cadmium / litre (Rusch et al., 1986) - many orders of magnitude higher.

2 Particle counts represent the number of particles integrated over time as the cloud passes the sampling point given the particular sampling rate in l/minute

### **Dissemination by ships**

Two sets of trials took place from ships as summarised in Table 6. The highest particle count (1,676) was recorded at Dorchester in a preliminary trial when 11 kg of zinc cadmium sulphide was discharged from the loading bays of a ship located 18 miles south of Portland Bill. The second set of trials conducted in the English Channel in January 1963 concerned the penetration of ships by aerosol, and no land based monitoring was done.

In total, from the nine trials involving dissemination from ships, 250 kg of zinc cadmium sulphide was released.

### **Estimation of total amount of zinc cadmium sulphide released during the long range trials**

It is not possible to give precise estimates of the total amount of zinc cadmium sulphide disseminated, as the amount released was not available for all the trials recorded, and some additional trials may have taken place during the

approved programmes for which data are not available. Nonetheless, some estimates can be made based on available data. From the 35 air-disseminated trials and nine ship-based trials with data, a total of c. 2,700 kg was released. Assuming the mean of 70 kg was released from each of the remaining seven trials, an estimated 3,200 kg in total was disseminated.

The programmes dated 1960-64 for which further data are not available are summarised in Table 7. A total of 28 trials was proposed, but three (at Netheravon airfield) were very small and are not considered further. Proposed releases of zinc cadmium sulphide from the remaining 25 trials amounts to 1,387 kg. Assuming that all these trials took place, this would have resulted in a total zinc cadmium sulphide disseminated during 8 years of c. 4,600kg, i.e. 1,426 kg cadmium (assuming cadmium content of zinc cadmium sulphide of 31%).

This should be compared with an estimated 15 tonnes cadmium released into the atmosphere from UK industry annually (Johnston and Jones, 1995). Thus, over an eight year period, the UK long range zinc cadmium trials added c. 1.2% to the atmospheric cadmium burden in the UK (assuming that cadmium industrial emissions were similar in the 1950's and 1960's to the 1990's).

### **Estimation of total dose of cadmium inhaled from the long range trials**

The tables show that the largest estimated dose received during any one trial was 0.42 µg of zinc cadmium sulphide, i.e., 0.13 µg cadmium.

We have assumed that there was a total of 76 UK trials of long range dissemination comprising the 51 trials reported in Tables 5 and 6 plus a further 25 trials from the programmes listed in Table 7 for which there seem to be no reports. We have made the (unrealistic) "worst case" assumption that at each trial, a person received the maximum estimated dose of Cd (0.13 µg) which was calculated from measurements at Dorchester on 18 August 1959 where the greatest contamination in any trial was found. Based on these assumptions the total dose to an individual over an eight year period would have been 9.9 µg Cd. This is the amount of cadmium that is inhaled in a normal urban environment in a period of between 12 and 100 days (or from smoking a total of 100 cigarettes).

Although insoluble CdS has a much lower bioavailability when inhaled than soluble cadmium compounds (Klimisch, 1993), the possibility of photodegradation of some of the ZnCdS to ZnCdSO<sub>4</sub> cannot be ruled out. Therefore a conservative renal accumulation of 35% of cadmium cleared by the lungs is assumed, with a lung clearance rate of 56% of inhaled cadmium (Klimisch, 1993), giving an additional renal burden of 1.9 µg Cadmium. Since the currently accepted critical limit of cadmium in the renal cortex is 200 µg/g (Rowels et al., 1981), any additional renal burden arising from the ZnCdS intake would have been negligible.

## Conclusions

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Between 1953-64 a series of trials were done involving the dispersion of Zinc Cadmium Sulphide particles. These included the land-based trials and those where the dispersion was done from ships or aircraft. These have been considered separately, since the land-based trials produce a different spectrum of possible harmful results over a very localised region only, compared to the dissemination into the atmosphere which involves much larger areas and bigger total amounts disseminated, but where the levels involved on the ground are much lower.

With regard to the land-based trials it is concluded that the dissemination of ZnCdS by the UK during the 'Cold War' did not significantly add to the cadmium concentrations in soil, except within a few metres of the point of dissemination of the material. Except immediately around the dispersal sites in two trials at RAF Beaulieu airfield, the predicted soil concentrations are within the range of cadmium concentrations normally observed in the UK and much lower than those found at Shipham. Risk to the public from soil contamination with cadmium from these and other ZnCdS dispersion trials is therefore considered to be negligible.

With regard to the long-range contamination trials, the dose potentially received by inhalation during even the most severe case was small (c. 0.13  $\mu\text{g}$  Cd). Over all the trials, our worst case estimate of personal dose received from these trials is c. 10  $\mu\text{g}$  over an eight year period. This is the amount of cadmium

that is inhaled in a normal urban environment in a period of between 12 and 100 days (or from smoking a total of 100 cigarettes).

We are also reassured by the fact that no increased incidence has been claimed for the diseases that are particularly associated with cadmium toxicity. The primary target of cadmium poisoning is the kidney. Although we have not been able to compare the incidence of kidney disease in more and less exposed areas, informal soundings from Dr M Macanovic, (consultant renal physician in Dorchester) do not suggest an unusual incidence of renal disease in Dorset where the highest ZnCdS particle numbers were recorded; and kidney disease has not given rise to any public concern in this connection. We were further reassured by discovering that the people largely involved in the dissemination of the cadmium sulphide from aircraft who will have been exposed to very much larger amounts than anybody on the ground suffered no ill effects.

Our conclusions therefore do mirror those in the much more comprehensive American study.

Although we fully understand the public unease that ensued when it was discovered, many years after the event, that large areas of Britain had been subjected to this form of experimentation, the existing evidence shows that no public health danger arose.

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## Annex Reports

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### Table 5

#### **Field Reports**

Titt RA The long distance travel of particulate clouds. *Field Report No. 504* (Programme 35/56), 5 May 1957

Wheeler C L. The long distance travel of particulate clouds. *Field Report No. 514* (Programme 1/57), 2 January 1958

Wheeler C L. The long distance travel of particulate clouds. *Field Report No. 515* (Programme 1/57), 5 February 1958

Titt R A. Determination of particle size in a cloud of FP produced by a Venturi dispenser mounted in a Valette aircraft. *Field Report No. 516* (Programme 19/57), 6 February 1958

Collins G F, Banfield J N. The penetration of built-up areas by aerosols at night. *Porton Field Trial Report No. 610* (Programme 2/63), 7 May 1964 (secret)

#### **Porton Notes**

Titt R A, Laird A. Long distance travel of particulate clouds. Porton Note No. 68, 23 January 1959 (restricted)

Thompson N. The vertical diffusion of particulate clouds over medium distances of travel: preliminary results from trials carried out in 1959. Porton Note No. 134 (restricted)

Collins G F, Laird A R. Long distance travel of particulate clouds. Porton Note No 138 (Programme 10/58), 29 February 1960 (restricted)

Collins G F, Laird A R. Long distance travel of particulate clouds. Porton Note No 139 (Programme 10/58), 26 February 1960 (restricted)

Titt R A. Long distance travel of particulate clouds. Porton Note 145 (Programme 10/58), 24 March 1960 (restricted)

Thompson N. The vertical diffusion of particulate clouds over medium distances of travel: results from trails carried out in 1960. Porton Note No. 183, 15 December 1960 (restricted)

Collins G F, Laird A R. Long distance travel of particulate clouds. Porton Note No. 185 (Programme 10/58), 26 January 1961 (restricted)

Laird A R, Titt R A. The long distance travel of particulate clouds. Porton Note No. 186 (Programme 10/58), 26 January 1961

Laird A R, Titt R A. The long distance travel of particulate clouds. Porton Note No. 187 (Programme 10/58), 1 March 1961

Laird A R, Titt R A. The long distance travel of particulate clouds. Porton Note No. 188 (Programme 10/58), 30 January 1961 (restricted)

Collins G F. Experimental dispensers for fluorescent powder. Porton Note No. 253 (restricted)

#### Programme Reports

Titt R A, Bradshaw A E, Wheeler C L. The travel of aerosols by night and the influence of topography upon the dosage including penetration into built-up areas. Programme No. 17/60, 10 August 1960

**Porton Technical Papers**

Jones J I P, Butler H E. Some measurements of the vertical distribution of particles at about 50 miles from a source. Porton Technical Paper No. 633, 20 March 1958

Titt R A. A preliminary study of the vulnerability of ships to BW attacks at sea. Porton Technical Paper No. 764, 17 March 1961

**Table 6****Porton Notes**

Titt R A. Large area coverage by aerosol clouds generated at sea. *Porton Note No. 146* (Programme 6/59), 24 March 1960 (restricted)

Collins G F, Laird A R, Titt R A. Large area covered by aerosol clouds generated at sea. *Porton Note No. 203*, 22 March 1961

**Porton Technical Papers**

Collins G F, Banfield J N. The vulnerability of ships at sea to BW aerosol attack. Porton Technical Paper No. 893, 7 February 1964

**Table 7****Programme Reports**

Trouern-Trend K, Bradshaw A E, Wheeler C L. Travel of particulate clouds: release of FP above friction layer. Programme No. 23/60, 7 November 1960 (restricted)

Thompson N, Barker A, Wheeler C L. The vertical diffusion of particulate clouds over medium distances of travel. Programme No. 2/61, 20 January 1961 (restricted)

Farley G G. Check of sampling and assessment technique in trials employing FP tracer. Programme No. 24/62, 15 January 1964 (restricted)

Collins G F, Bradshaw A E, Musty J W G. Comparison of FP dosages measured by drum impactors and millipore filters after different distances of cloud travel. Programme No. 10/63, 17 September 1963 (restricted)

Banfield J N, Bradshaw A E, Musty J W G. The penetration of built-up areas by aerosols at night. Programme 2/64, 1 January 1964

## Tables

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**Table 1. Cadmium content of soil**

Soil type	Cadmium content ( $\mu\text{g/g}$ )	
	Median	Range
UK soils generally		Rarely exceed $2\mu\text{g/g}$ with a range of 2-8 $\mu\text{g/g}$ in top soil
Agricultural soils in England and Wales	<1	0.08 -10
Garden soil in Shipham	91	2 - 360
Agricultural soil in Shipham		30 - 800

**Table 2. Cadmium intake from food in the UK**

Year	$\mu\text{g Cd/day}$	$\mu\text{g/kg body weight/week}$	Total weekly intake $\mu\text{g}(60\text{kg /person})$
1973		4	Not more than 250
1974-1984	Less than 20	2.3	Less than 140
1981	Probably 10-17		70-119
1982	9-18		63-126
1983	11-18		77-126
1984	12-19		84-133
1985	12-15		84-126
1986	10-17		70-119
1987	11-18		77-126
1988	11-19		77-133
1991	12-18		84-126
1989-1993	7		49

Smoking adds 1-4 $\mu\text{g/day}$  of Cd, assuming 20 cigarettes per day. The NRC figure is 2-4 $\mu\text{g/day}$ .

The intake figures may be compared with the Provisional Tolerable Weekly Intake (PTWI) figures for a 60kg person:

In 1972 between 400 $\mu\text{g}$  and 500 $\mu\text{g}$  /person

In 1989 420 $\mu\text{g}$  /person

In 1993 420 $\mu\text{g}$  /person



**Table 3. Dietary intake of cadmium in Shipham and nationally**

	<b>Average µg/week/person</b>	<b>Range</b>
From diary estimates kept by 75 families in Shipham during 4 weeks in May and September 1979	250	130 - 520
Duplicate diet estimate, based on duplicates of a simple week's diet, provided by 65 Shipham people in September 1979	200	60 - 1080
National average estimate (1978 data)	140	90 - 180

**Table 4. Land-based dissemination of ZnCdS by the UK Ministry of Defence sampled by fallout impactors at near distance (<300 m) and cascade impactors at 25 and 50 miles.**

<b>Field report</b>	<b>Material disseminated (g)</b>	<b>Material recovered from disseminator (g)</b>	<b>Location</b>	<b>Additional cadmium load to soil (ug Cd/g soil DM)</b>	<b>Area above 'legal' maximum concentration**</b>
<i>Programme 1/54 Assessment of 'fallout' of fluorescent powder emitted from the 'Stanford' type aerosol generator</i>					
353	367	302	SE corner of Beaulieu airfield (no further details)	2.5 (180 <sup>+</sup> )	10 x 2.5 (138 x 25 <sup>+</sup> ) m
405 <sup>++</sup>	1) 736 2) 489	5 110	Beaulieu airfield	0.1 1.6	None None
<i>Programme 17/53 The long distance travel of particulate clouds</i>					
370	3462	507	RAF Beaulieu airfield (grid reference given)	3.3	4 x 2 m
371	2291	1032	RAF Beaulieu airfield (grid reference given)	No near distance sampling was conducted, but only 54 and 27% of emissions in Field Report No 370 were reported at 25 and 50 miles from the source, respectively.	Probably one

372	3106	409	Porton	52 and 40% of levels reported in Field Report 370 (fewer sampling points)	Probably none
373	3031	343	RAF Yatesbury (grid reference given)	40 and 40 % of levels reported in Field Report 370 (fewer sampling points)	Probably none
382	2227	709	A35, A351 and B3075 near Blandford, Salisbury++	56 and 39% of levels reported in Field Report 370 at 25 and 50 miles from the source, respectively.	Probably none
388	2959	355	RAF Hullavington, Nr Chippenham	Samplers inaccurately placed and were missed by the cloud of ZnCdS	Probably none
392	2345	337	4 mile stretch of minor roads near Frome, Somerset	30 and 15%+ of levels reported in Field Report 370 at 25 and 50 miles from the source, respectively.	Probably none

\*\* Assuming critical addition is c.  $85 \mu\text{g}/\text{cm}^2$ , a value equating roughly to +2.3 mg/kg soil DM, assuming baseline soil concentration is 0.7 mg/kgDM

†† The field report (Titt, R.A. 1954a. Assessment of 'fallout' of fluorescent powder emitted from the 'Stanford' type aerosol generator. Field Report No 353, Ministry of Defence, Porton) gives the units for the contour diagram of fallout as mg/m<sup>2</sup>. However, an amendment dated 2/4/54 (Titt, R.A. 1954b. Assessment of 'fallout' of fluorescent powder emitted from the 'Stanford' type aerosol generator, Amendment 1 to Field Report No 353, Ministry of Defence, Porton) states that the units should be micrograms/Petri dish. If the units are assumed to be micrograms/Petri dish it can be calculated from the contour map of fallout that approximately 20-30 kg of material should have been dispersed from the dispenser; the same calculation produces a quantity of c. 150-250g if it is assumed that the units are, as originally stated, mg/m<sup>2</sup>. The report records that

367g was disseminated and it therefore seems likely that the units were  $\text{mg/m}^2$ , with the difference in the amount disseminated and that recorded on the contour map being due to photodegradation, some material falling outside the recorded area and experimental error. The reason for the amendment is unknown. The higher values shown are for the assumption that the units are  $\mu\text{g}/\text{Petri dish}$ .

<sup>+</sup> 30 and 80 miles sampling points rather than 25 and 50 miles from source.

<sup>++</sup> Dispersion over distance from vehicle to simulate 'stealth attack' by enemy.

†† Comparison between two disseminators

**Table 5. Trials of dissemination from aircraft, and estimated theoretical inhaled dose of ZnCdS at sampling point with maximum particle count**

Report	Date of Trial	Approx. amount released (kg)	Location	Max particle count	Sampling rate (l/min)	Estimated inhaled dose ( $\mu\text{g}$ ) <sup>†</sup>
FR 504	11.9.56	12	Porton	351	17.5 <sup>†</sup>	0.039
	17.9.56	111	North Sea	93	17.5	0.010
FR 514	25.4.57	138	North Sea	337	22	0.030
FR 516	28.8.57	5	Cardington	3403	20	0.332
FR 515	13.11.57	117	North Sea	112	10	0.022
PTP 633	14.11.57	**	Cardington	99*	20	0.010
	15.11.57	**	Cardington	1070*	20	0.104
PN 68	22.7.58	98	Irish Sea	415	20	0.041
PN 138	18/19.9.58	116	English	674	20.5	0.064
PN 139	28.10.58	120	English	669	20.5	0.064
PN 145	11.12.58	83	North Sea	144	20.5	0.014
PN 185	14.4.59	123	English Channel	182	20.5	0.017
PN 186	26.5.59	139	North Sea & North East England	1591	21.5	0.145
PN 187	7.7.59	139	English Channel & Cornwall	123	20	0.012
PN 188	18.8.59	127	English Channel	4315	20	0.421
PN 253	19.3.58	**	Cardington		**	**
	4.3.59	**	Cardington		**	**
	7.5.59	**	Cardington		**	**
	9.9.60	**	Cardington		**	**
PN 134	29.5.59	45	Cardington	96*	20	0.009
	3.6.59	45	Cardington	44*	20	0.004
	25.8.59	45	Cardington	17*	20	0.002
	15.9.59	45	Cardington	129*	20	0.013
	16.9.59	45	Cardington	34*	20	0.003
	17.9.59	45	Cardington			
	23.9.59 (1st)	45	Cardington	278*	20	0.027

	23.9.59 (2nd)	45	Cardington	271*	20	0.026
PN 183	21.4.60	17.5	Cardington	209*	20	0.020
	25.5.60	17.5	Cardington	143*	20	0.014
	26.5.60 (1st)	17.5	Cardington	285*	20	0.028
	26.5.60 (2nd)	17.5	Cardington	315*	20	0.031
	26.5.60 (3rd)	17.5	Cardington	157*	20	0.015
	17.6.60	91	Cardington	387	20	0.038
	29.6.60	15.11.57	Cardington	30	20	0.003
	30.6.60	91	Cardington	87	20	0.008
	1.7.60	91	Cardington	36	20	0.004
PTP 764	27.10.60	74	English Channel	2§	21.3	0.000
	28.10.60	74	English Channel	91§	19.8	0.009
Prog. 17/60/PF TR 610#	**	**	Salisbury	**	**	
PFTR 610#	28.3.63	68	Norwich	377	10	0.074

§ Land based sampling (samples were also collected at sea)

\* At height 2m

\*\* Not given

# PTFR 610 missing, but reported on in Carter (1999)

FR Field Report

PN Porton Note

PTP Porton Technical Paper

PFTR Porton Field Trial Report

**Table 6. Trials of dissemination from ships, and estimated theoretical inhaled dose of ZnCdS at land-based sampling point with maximum particle count**

Report	Date of Trial	Approx. amount released (kg)	Location	Max particle count	Sampling rate (l/min)	Estimated inhaled dose ( $\mu\text{g}$ )#
PN 146/203	7.10.59	11	English Channel	1676	20.4	0.160
	7/8.11.59	93	English Channel	40	20.2	0.004
	9/10.11.5	95	Irish Sea	115	21.4	0.010
PTP 893	28.1.63	7.6	English Channel	§		
	28.1.63	8.6	English Channel	§		
	29.1.63	9.5	English Channel	§		

	29.1.63	4.8	English Channel	§		
	30.1.63	10.2	English Channel	§		
	30.1.63	10.4	English Channel	§		

‡ Assuming respiratory rate of 16.6 l/min (NRC 1997),  $1.7 \times 10^{10}$  particles/g (FR 504; PN 185) and loss of 50% of fluorescence (Collins 1981) .

§ Land based sampling not done

PN Porton Note

PTP Porton Technical Paper

**Table 7. Approved programmes of trials § of dissemination of ZnCdS from aircraft, 1960-64, where reports of the trials (and indication as to whether or not they took place) are unavailable**

Programme number	Proposed location	Numbers of trials proposed	ZnCdS per trial (kg)	Total amount ZnCdS (kg)
23/60	Not stated	3	91	273
2/61	Cardington	4	68	272
	Cardington	6	14	84
24/62	Cardington	6	45	270
10/63	Cardington	2	136	272
2/64	Norwich	4‡	54	216
			<b>TOTAL</b>	<b>1,387</b>



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