

**DIOXIN SOURCES, AIR TRANSPORT AND  
CONTAMINATION IN DAIRY FEED CROPS AND MILK**

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## SUMMARY

Exposure of the U.S. population to dioxin is a serious and unresolved problem. It is serious because the amounts of dioxin in the bodies of representative Americans result from a level of exposure that causes concern about an increased incidence of cancer and birth defects. It is unresolved because critical gaps in what is known about human exposure to dioxin have hindered action to reduce or eliminate it.

A great deal is known about the dioxin problem. It is known that nearly all of the exposure to dioxin is due to its presence in food, chiefly milk, dairy products and beef; that these foods are contaminated by the dioxin present in cattle feed, which in turn is absorbed from the air as the feed crops grow; that dioxin is carried through the air and can contaminate farms even 1000 miles from the sources that emit it; that preventive measures to eliminate the entry of dioxin into the air must be directed at these sources, such as incinerators, smelters, and backyard trash burners.

However, there has been a major gap in what we know about this chain of events. We have not known enough about the link between the emitting sources and the ecological receptors, such as dairy farms, to actually identify those sources that are chiefly responsible for the contaminated crops and human food. Such knowledge is a prerequisite for effective preventive action. At the same time, little has been learned so far about the possible effect of different dairy farm practices on the dioxin content of milk, knowledge that might permit actions, on-farm, to reduce it.

These considerations defined the purpose of this study: to rank the numerous U.S. and Canadian dioxin sources with respect to their contributions to the concentrations of airborne dioxin at individual dairy farms, and to explore the effect of dairy feed practices on the dioxin content of milk.

For this purpose, four dairy farms in Vermont and four in Wisconsin were selected for study. Of these farms, three (VT-A, VT-C and WI-A) were intensive grazing (pasture) farms; their cows received 85-95% (wet weight) of their diet from the farms' pastures and the rest from grain supplements. The remaining five farms were conventional confinement farms, where the cows were fed Total Mixed Ration prepared from a variety of components, including locally grown or purchased silage and hay, grain and

supplements. The eight farms maintained a total of 1,092 cows, averaging 137 per farm. At two Vermont farms and two in Wisconsin, air was continuously sampled for dioxin analysis over a one-month period in August/September 1996. During that same period, at each farm the cows' diet and the herds' milk output were also sampled for dioxin analysis.

The contribution of U.S. and Canadian sources to the total concentrations of airborne dioxin at the farms during the test period was estimated by means of the HYSPLIT/TRANSCO computer model, which traced emitted dioxin from each of the numerous sources to each of the eight test farms. The overall dioxin inventory includes a total of 24,644 sources: 5,710 individually identified (e.g., the municipal waste incinerator at Bridgeport, CT), and 18,934 area-based (e.g., all the backyard trash burners in Calumet County, WI). The inventory, which provides estimates of the annual amount of dioxin emitted into the air from each of these sources, is classified into 20 different types of sources. However, only five source-types account for 86% of the total emissions: municipal waste incinerators, medical waste incinerators, cement kilns (not burning hazardous waste), secondary copper smelters, and iron-sintering plants, in descending order of emissions.

The model was evaluated by comparing the measured concentrations of airborne dioxin with model-generated concentration estimates at the four farm sampling sites and at a remote Connecticut mountain site (three month-long measurements had been made in 1996 by a consultant for the State). The results showed that the model produces reasonably accurate estimates of the measured concentrations at the Vermont and Connecticut test sites. At the two Wisconsin test sites the model significantly underestimated the measured airborne dioxin concentration, apparently because of a defect in the inventory characterization of a local source or unusual local wind conditions.

The model was used to estimate the contribution that each of the numerous sources made to the airborne dioxin concentrations at the eight farms during the month-long test period. Initially these data were analyzed with respect to several features that characterize the dioxin sources: distance from the receptor; geographic orientation

relative to the receptor; and source-type. The model-generated data were also used to rank the entire list of 24,644 sources with respect to their contributions to the concentration of airborne dioxin at the test farms. Despite the very large number of sources that contribute to the overall emissions of dioxin, very few sources are responsible for most of the airborne dioxin at any given farm. Thus, the 1,000 highest-ranked sources, only about 4% of the total number, accounted for 90%-98% of the total dioxin concentration at the test farms.

At the Vermont receptors, only the eight or nine highest-ranked sources (less than 0.1% of the total number) account for about 60% of the total airborne dioxin concentration. At VT-C (central Vermont), for example, these sources include six municipal waste incinerators, three in New York, one each in Illinois and Ohio, and one in Quebec, Canada; a secondary copper smelter also in Quebec; and backyard burners in a county in northern New York.

At WI-A (southeastern Wisconsin), a region more heavily exposed to dioxin sources than Vermont, the 43 highest-ranked sources contributed 60% of the total airborne dioxin at the farm. They included not only a number of municipal waste incinerators, an equal number of cement kilns (not burning hazardous waste), mostly in Nebraska, Kansas, Missouri and Illinois, but one in Alberta, Canada, as well; eight iron sintering plants in Indiana, Ohio and Maryland; and three secondary copper smelters in Illinois, Ohio and Maryland. Thus, such data specify, for each test farm, the individual sources to which preventive action can be most effectively directed.

At the three intensive grazing farms, pastures provided nearly all of the cows' diet, and the dioxin content of the pasture vegetation ingested during the month-long test period was therefore derived concurrently from the air. In contrast, the dioxin content of the Total Mixed Ration fed to the cows at the confinement farms was expected to bear no relation to the airborne dioxin concentration at those farms during the test period, because the component crops (e.g., corn for silage) were often purchased rather than grown on-farm, and therefore not exposed to the local airborne dioxin during the test period. Accordingly, only the dioxin concentrations of the pasture-based diets were expected to reflect the local concentration of airborne dioxin. A plot of

the dioxin concentration in these locally grown diets against *the measured* concurrent dioxin concentration in the air, although necessarily limited to the three pasture farms, indicated that the diet concentrations are generally proportional to the air concentrations. Thus, the approximately three-fold range in the measured concentration of airborne dioxin at the three pasture farms (two in Vermont and one in Wisconsin) is reflected in a comparable range in the diet concentrations as well.

By comparing the dioxin content of the different diets at the eight test farms with the dioxin content of the milk produced concurrently, it was possible to estimate the carryover rate — i.e., the percentage of the dietary dioxin that appears in the milk. The values, which range from 3% to 51%, bear no simple relationship to the amount of dioxin ingested in the diet since other, undetermined, factors may influence the rate as well. In particular, dioxin accumulated in the body fat during the life of the cow may be mobilized and add to the dietary dioxin that appears in the milk. In turn, the rate of mobilization is influenced by various metabolic factors, especially high rates of milk output, which may tend to increase fat mobilization and hence add non-dietary dioxin to the milk, thereby increasing the apparent carryover rate. The average carryover rate for all eight farms is 17%, close to the range of values (20-25%) reported in the recent literature, generally under relatively controlled concentrations.

Certain useful generalizations about the dioxin problem and recommendations for remedying it can be drawn from the results of this study:

- The level of overall exposure of dairy farms to airborne dioxin and the geographic distribution of the sources responsible for most of this exposure appear to be characteristic of regions such as those typified by Vermont and Wisconsin. In Vermont, the farms are exposed to dioxin from relatively few major sources, and airborne dioxin levels are relatively low; in Wisconsin, the farms' dioxin exposure is due to more numerous major sources (which are more common in the region), and airborne dioxin levels are relatively high.
- There is a reasonably linear proportionality between the concentrations of dioxin in the air and in the vegetation grown concurrently. Consequently, if the presence of airborne dioxin at a dairy farm is eliminated, within the next growing season the

dioxin content of the feed crops grown on-farm will fall to zero. The cows bred at that farm and fed farm-grown crops would then produce dioxin-free milk, within the time required to replace older milk cows by new ones — approximately three to four years.

- Despite the very large numbers of sources of various types in the total dioxin inventory, relatively few, readily identified, sources are responsible for the major part of the dioxin that reaches a given farm. This greatly facilitates designing preventive action.
- Studies of the effect of diet composition, especially between grazing and confinement farms, on the dioxin content of milk could identify farm practices that might minimize the degree to which dietary and body fat dioxin appear in the milk, for example by eliminating the feeding of fats and oils.
- In sum, the results of this study support the development of a practical policy to prevent the production of dioxin at the source and thereby eliminate the exposure of dairy farms (and other important receptors as well). By identifying the distinctive array of sources which — if replaced by dioxin-free alternatives — could virtually eliminate exposure, the study provides a practical physical basis for remedial policy. By specifying the geographic locations of these major sources, the study also defines the regulatory and political features of such policy.

Economically feasible dioxin-free alternatives exist for most of the major sources that are responsible for the airborne dioxin that contaminates the milk produced at the test farms in Vermont and Wisconsin: for example, substituting mandatory recycling and composting of municipal waste for incinerating it; substituting autoclaving and landfill disposal for medical waste incineration; and changes in iron sintering technology to eliminate chlorine-bearing materials from steel plant residues. The effort to implement such preventive measures involves the interests of dairy farmers, the dairy product industry, regulatory agencies, the operators of dioxin sources, community environmental groups, and the general public. There is now an opportunity for coalitions that bring all these forces to bear on the common goal of producing dioxin-free milk.

## I. INTRODUCTION

This project was concerned with a serious environmental issue: the exposure of the general U.S. population to potentially harmful levels of dioxin, chiefly through the ingestion of milk, dairy products and beef. The most recent evaluation of the problem by the U.S. EPA points out that exposure to this highly toxic pollutant — as evident from the levels found in body fat and mothers' milk — is sufficient to cause concern about the occurrence of an increased incidence of cancer and disrupted fetal development. Milk and dairy products alone, essential elements of the diet, account for more than one-third of the total intake of dioxin by the general population. This project was designed to help meet the need for a rational analysis of the environmental processes that mediate the contamination of milk by dioxin and for economically constructive action to eliminate this hazard.

Such an analysis requires that the sources that introduce dioxin into the environment are identified; that the passage of dioxin through the environmental pathways leading to the agricultural receptors that are exposed to dioxin — dairy farms — is characterized; and that the processes that carry dioxin from the contaminated feed crops to milk are defined. Earlier studies have indicated that the preponderant exposure of humans through food is the result of the transport of dioxin from the numerous sources that emit it — chiefly combustion processes, such as incineration — through the air to agricultural crops. The analysis of such source-receptor relationships is critical, because it is now widely recognized that the only lasting, effective remedy to human exposure is to prevent the generation of dioxin at the source. In turn this requires that the sources be ranked with respect to their impact on the receptors, such as dairy farms, so that action can be directed to those sources that are chiefly responsible for the dioxin content of the milk that the farms produce.

To meet these requirements, the project has made use of a computer model that we developed originally to estimate deposition of airborne dioxin on the Great Lakes<sup>1</sup>.

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<sup>1</sup>Cohen, M., Commoner, B., *et al.*, 1995: Quantitative Estimation of the Entry of Dioxins, Furans and Hexachlorobenzene into the Great Lakes from Airborne and Waterborne Sources. Report to the Joyce Foundation.

The model is capable of tracking dioxin from each of numerous geographically localized sources, through the air (where it is subject to diffusion, destruction and deposition en route), to receptors located anywhere else in the United States and most of Canada. The model estimates the total airborne concentration of dioxin that reaches any given receptor and estimates the contributions of each of the numerous sources to that total.

As an initial effort to demonstrate the applicability of this model to the source-receptor relations that mediate human exposure to dioxin, the project was designed to trace airborne dioxin from the known sources in the United States and Canada to eight dairy farms in Vermont and Wisconsin. Two of these farms, VT-A and VT-B, are located in northern Vermont, and two, VT-C and VT-D in central Vermont. Two of the farms are located in southeastern Wisconsin (WI-A and WI-B), one (WI-C) in central Wisconsin, and one (WI-D) in western Wisconsin. In all but the central Wisconsin region, airborne dioxin was collected continuously over a one-month period (August/September 1996; see Table 1) and analyzed with respect to the concentrations of the various toxic dioxin and furan congeners.

Samples of the dairy feed crops and milk produced during the test period were also collected for dioxin analysis at each of the eight farms, with the aim of delineating the passage of dioxin from the air, through feed crops to the milk produced for human consumption.

**Table 1: Air Sampling Locations and Periods**

Region	Location	Date (1996)	
		Start	Stop
Northern Vermont	VT-A	Aug. 1	Aug. 28
Central Vermont	VT-C	Aug. 1	Aug. 28
Southeastern Wisconsin	WI-A	Aug. 22	Sept. 17
Western Wisconsin	WI-D	Aug. 20	Sept. 18

## II. PROCEDURES

### A. Operation of the Dioxin Air Transport Model

#### Background

The amount of dioxin that reaches an ecological receptor depends on the amount that the sources emit into the air and how much of that is lost (destroyed or deposited) in transit or goes somewhere else. As soon as the dioxin leaves the source, some of it — the portion chiefly attached to relatively large particles — settles out on the ground nearby. However, only about 1-10% of the emitted dioxin is deposited within 30 miles of the source. The rest, in the form of vapor or attached to small particles, moves with the wind and spreads over an ever-increasing area. Meanwhile, subject to gravity, the vertical movement of the air, diffusion, and precipitation, some of the dioxin falls to the ground, reducing the amount that is still airborne and able to reach a distant receptor. At the same time, there are destructive processes at work: sunlight can destroy dioxin depending on whether it is in the form of vapor and therefore exposed to the ultraviolet radiation, or attached to solid particles, and thereby shielded from it. What is more, whether the dioxin is attached to protective particles or in the form of vapor depends on the temperature; there is a higher proportion of vapor in warm air. Finally, when the dioxin reaches the receptor, the amount that comes down depends on the local weather conditions at the time and the nature of the receptor itself (e.g., the prevalence of vegetation). The dioxin air transport model takes all of these processes into account.

#### Model Operation

The model employed in this project is based on one developed by the National Oceanic and Atmospheric Administration called HYSPLIT, after Hybrid Single Particle Lagrangian Integrated Trajectory. It incorporates detailed weather data for the United States, Canada and northern Mexico for a grid of 924 points 183 km apart at six levels up to 3,000 meters, recorded at two-hour intervals for every year since 1988. In this study weather data for 1996 were used. The computer model starts with a "puff" containing a fixed amount of material, emitted at set intervals into the air from a source

at a known geographical location. It then tracks each puff as it spreads, moves with the weather, and the material in it is destroyed in transit, or is deposited. We have modified HYSPLIT to incorporate the behavior of the 17 molecular forms of toxic dioxins and furans (congeners) and eight additional non-toxic congener groups, in particular with respect to their distribution between the vapor and particulate-bound phases.

The various congeners differ a great deal in their relative toxicity. In order to estimate the toxicity of the entire group, a widely accepted procedure is used, in which the toxicity of each congener is expressed as a fraction — the Toxic Equivalence Factor (TEF) — of the toxicity of the congener 2,3,7,8-tetrachloro-dibenzo-p-dioxin (TCDD), which has the highest level of toxicity. The sum of the TEF values for all 17 toxic congeners is the Toxic Equivalence Quotient (TEQ). The amount of dioxin is then given as "picograms (pg) TEQ" — thus expressing the amount in terms of the overall toxicity of the entire group of dioxin and furan congeners that we refer to as "dioxin," or as "PCDD/F."

In a typical computer run, the model tracked the movement of a series of puffs emitted from each source at regular intervals over a time period selected for simulation. It computed the position of each of these numerous puffs and the amounts of the congeners it contained. The model finally estimated the amounts of the congeners emitted from each of the sources that were deposited at each receptor.

Initially model runs were carried out for a series of 28 standard (hypothetical) sources (suitably distributed, geographically), assumed to emit dioxin at a standard rate of 1 gram per hour. The model tracked the emitted dioxin to each of the receptors, that is, the dairy farms. This procedure yielded, for each standard source-receptor combination, the concentration of airborne dioxin at the receptor resulting from 1 gram unit-emissions at the standard source locations over the test period. In effect, these data established, for each standard source-receptor combination, an Air Transport Coefficient (ATC) — i.e., the fraction of the dioxin emitted by the source that occurs in the airborne dioxin at the receptor. For these initial 28 standard sources, simulations were performed for four different congeners (2,3,7,8-TCDF, 2,3,7,8 TCDD, 2,3,4,7,8-

PeCDF, and OCDD), using a time step of one hour with a seven gram puff once every seven hours. A total of 112 such runs were carried out, with each run requiring on the order of three to eight hours on 200-300 MHz personal computers. Runs of this type were also carried out with a set of nine close-in standard source locations for each concentration evaluation data point (a total of 63 additional standard points) involving 252 computer simulations. Finally, an additional 288 HYSPLIT runs were carried out to simulate the effect of the potentially largest sources of dioxin to which the test farms might be exposed. In sum, a total of 652 HYSPLIT simulations, requiring about 3000 hours of computer time, were carried out.

An interpolation procedure (TRANSCO) has been developed that estimates the air transport coefficient for each actual source-receptor combination. The procedure is based on: (a) the relative distances between the actual source and the four nearest standard sources; and (b) the relation between the angular orientations of the actual source and the four standard sources to the receptor. TRANSCO also estimates the impact of congeners not explicitly modeled by means of an interpolation procedure based on their vapor/particle partitioning characteristics relative to those of the modeled congeners. The computed air transport coefficients of the actual sources, multiplied by their actual emission rates, yields the concentration of dioxin and/or the amounts deposited at the receptor sites by each actual source.

#### **B. Farms Selected**

Table 2 below summarizes the relevant features of the eight farms studied. All the farms produced milk for the liquid milk market 12 months of the year; none were strictly "seasonal" in milking, a practice in which cows are not milked for approximately two months, normally in winter, so that the entire herd comes into lactation at approximately the same time. However, Farms VT-A and VT-C were somewhat seasonal in milk production. Farms, VT-A, B, C and WI-A, B, C were relatively remote from localized sources of dioxin emissions, such as incinerators. However, VT-D and WI-D were closer to potentially higher levels of local dioxin emissions.

Table 2: Summary of Farms in Study

State	Farm	Descriptive Location in State	Proximity to Other Study Farms	Type	# of Cows Producing the Milk Sampled	Main Forage Feeds Used in addition to other feeds etc.	Geographical Source of These Forage Feeds	Year Forage Grown	Nearby Pollution Sources
WI	A	South-eastern	WI farms A and B were about 3 miles apart	Intensive Pasture Management/ Rotational Grazing	86	pasture	on study farm	1996	relatively major road with heavy traffic  less than 100 miles from large urban centers (Chicago and Milwaukee)
WI	B	South-eastern		Conventional Confinement	53	hay	on study farm	1996	
WI	C	Central		Conventional Confinement	175	haylage	in farm region (a)	≤ 1995	
						hay	in farm region (b)		
WI	D	Western		Conventional Confinement	240	haylage	on study farm	1996	near urban area that includes a waste incinerator; near Mississippi River (with heavy boat traffic)
						corn silage	on study farm	1995	
VT	A	Northern	VT farms A and B were adjacent to one another	Intensive Pasture Management/ Rotational Grazing	140	pasture	on study farm	1996	less than 100 miles from large urban area (Montreal)
VT	B	Northern		Conventional Confinement		corn silage	in farm region (c)	1995	
						haylage	in farm region (c)	1995	
						hay	in farm region (c)	1995	
VT	C	Central		Intensive Pasture Management	95	pasture	on study farm	1996	
						haylage	on study farm	1996(d)	
VT	D	Central		Conventional Confinement	28	haylage	near VT Farm C	1995	near an interstate interchange in an urban area, with relatively heavy traffic
						corn silage	near VT Farm C	1995	

Notes: (a) 75% of haylage from hay grown on farm; 25% purchased; haylage reported to be at least a year old at time of study (i.e., ≤ 1995)  
 (b) hay purchased within 10 miles of farm 95% of the time  
 (c) farmer utilized may fields within 3-4 miles of farm  
 (d) 75% from 1996; 25% from 1995

Two farms in Vermont (VT-A and VT-C) and one in Wisconsin (WI-A) practiced intensive pasture management. This means that the primary feed in spring, summer and fall was forage, grazed by the cows from small, separately fenced pasture lots through which the cows were rotated during the grazing months. Pastured cows returned to the barn twice a day to be milked and to receive moderate to small amounts of high energy concentrates (generally grains). The confinement farms held their cows in the barn, with only short periods of time outside in exercise lots; these cows were fed and milked twice a day, in the barn. Feed consisted of a composite diet of Total Mixed Rations (TMR); grazing was not part of their diet. Pasture and confinement farms were chosen adjacent to each other where possible. Thus, in Wisconsin, farm A (pasture) and farm B (confinement) were only three miles apart. In Vermont, farms A (pasture) and B (confinement) adjoined each other, and farms C (pasture) and D (confinement) were approximately 20 miles apart.

As far as feasible, farms were selected that grew most of their own forage and feed and bred and raised their own replacement heifers (this term is used for a female cow that has not yet been bred and is not producing milk, although it may also be used for a cow during the first lactation). Finally, farms were selected to be as close as possible to a national weather station so that comparisons could be made with the data from on-site weather stations.

### **C. Field Sampling**

A common sampling protocol, which is summarized in Table 3, was followed at all farms, except for minor variations due to uncontrollable events (e.g., weather). The following types of samples were collected:

#### Air

Instrumentation (in keeping with EPA method TO-9) was provided and set up by the ENSR Laboratories at Farms A and C in Vermont and Farms A and D in Wisconsin where month-long air samples were collected for dioxin analysis (see Table 1). A duplicate measuring station was operated at VT-C. Meteorological stations were also installed at VT-A and VT-C to continuously record precipitation, wind direction, wind

Table 3: Overall Summary of Sampling Conducted During Study

Material	General Characteristics of Sampling Programs*					Was Sampling Program Conducted at Farm? (Y=yes)									
	Sample Type	Sampling Frequency	Sample Duration and/or Compositing	Field Blanks Collected and Analyzed?	Duplicate Samples?	State--	Wisconsin				Vermont				
						Farm--	A	B	C	D	A	B	C	D	
						Location--	S	S	C	W	N	N	C	C	
						Type--	g	c	c	c	g	c	g	c	
Milk	individual samples from stirred bulk tank before milk tanker collection	once every 1-3 days, depending on milk tanker collection frequency	individual samples collected for one month and then combined into one composite sample	WI: yes (f) VT: yes	at one farm in each state		Y	Y	Y	Y	Y	Y	Y	Y	Y
Total Feed	individual grab samples collected from mixed material	once every week	individual samples collected for one month and then combined into one composite sample	WI: yes (f) VT: yes	VT one farm		-	-	Y	Y	-	Y	-	Y	
Feed Components (including pasture at grazing farms)	individual grab samples collected from mixed material	once every week	individual samples collected for one month and then combined into one composite sample	-	duplicate pasture samples at 1 farm per state		Y	Y	e	Y	Y	-	Y	Y	
Air (in addition, weather station data were collected at one site per state)	continuous air sample drawn through glass fiber filter and polyurethane foam	one continuous monitoring sample taken throughout study period	each sample was approximately one month in duration; filter and foam samples combined into one sample per site	-	one co-located air sample collected in Vermont		Y	a	-	Y	Y	b	Y	-	
Pasture (air-to-crop)	all vegetation clipped at end of air sampling from pasture that had been grazed just before air sampling started	one sample collected at end of air sampling	~2 liters of material from each of ~10 random areas in pasture combined into one sample	-	-		Y	-	-	-	Y	-	-	-	
Corn (air-to-crop)	whole corn plants cut from field	one sample at start and end of air sampling	~ 10 whole corn plants collected from different areas in field and combined into one sample	-	-		-	-	-	d	-	Y	-	-	
Alfalfa (air-to-crop)	whole alfalfa plants clipped from field	one sample at start and end of air sampling	~ 2 liters of material collected from each of ~10 random areas combined into one sample	-	-		-	-	-	Y	-	Y	-	-	

Notes and Code Explanations:

- \* Nominal features of programs (there were minor variations among farms)
- a Wisc. Farm B was ~1 mile from Farm A so air sample somewhat relevant
- b In VT, Farm B was adjacent to Farm A so air sample very relevant
- c Conventional feed farm

- d Representative ending sample not collected, as crop was cut prematurely
- e Samples collected but not analyzed
- f Samples collected but lost at laboratory
- g Rotational Grazing Farm

speed, and temperature as one-hour averages for the duration of the sampling period. Instrumentation was monitored on a regular basis several times a week. In addition, a sling psychrometer was used to measure humidity at times of data collection. At the Wisconsin sites, wind speed, direction and temperature were measured at intervals with a portable instrument set.

#### Crop Samples

As noted earlier, the dairy herds on the test farms were maintained on two different types of diet. On the three pasture farms (VT-A and VT-C and WI-A) herds were rotated through a series of small pasture lots, generally given a fresh paddock after each milking, or more frequently depending on pasture growth. These cows received 95% (by weight) of their diet from pasture, and the remainder in a mixture of different types of grains (grain concentrate) and some minerals. The herds on the confinement farms (VT-B and D; WI-B, C and D) were fed Total Mixed Rations (TMR), a mixture of feeds generally composed of corn, alfalfa, or grass silage, grains, animal protein and fat, cottonseed, and minerals. The exact proportions varied from farm to farm (see Table 5). The two types of diet differed with respect to their exposure to dioxin, which is absorbed by the feed crops from the air during growth. Thus, on the pasture farms, where the feed crop is consumed soon after it grows, the dioxin it contains was received from the air, at the farm, during the current growing season. In the confinement farms the components of the TMR were grown in different places (some at the farms themselves and others in the Midwest or Canada), generally in the preceding growing season (1995); hence, the feed component's dioxin content bears no relation to the airborne dioxin concentration at the farm during the sampling period (August/ September 1996).

All of the dioxin analyses of the vegetation and milk samples took place at the Midwest Research Institute (MRI) in Kansas following U.S. EPA protocol number 8290. Feed samples were stored in laboratory pre-cleaned 2-liter glass bottles; field blanks consisted of identical bottles exposed at time of mixing to ambient air. The samples that were taken for analysis are described below.

### Concurrent Diet and Milk Samples

These samples were intended to provide an estimate of the dioxin intake of the cows during the one-month test period, for comparison with the amount excreted in the milk during that same period.

*Total Mixed Ration:* Grab samples were taken from the TMR mixture prepared by the farmer, weekly, over the one-month test period. The four weekly samples were mixed at the end of the test period and a two-liter aliquot taken for dioxin analysis. This procedure applied to the confinement farms (VT-B and D; WI-B, C and D). (At three of these farms (VT-D, WI-B and D) samples of the individual TMR components were also taken for separate dioxin analysis.)

*Pasture:* Samples were collected by hand (carefully cleaned), to simulate the actions of a cow in feeding, from 10 randomly selected areas of the farm pastures. Such samples were taken weekly during the one-month test period from pastures about to be grazed, then combined and a two-liter aliquot taken for dioxin analysis. Samples of the grain supplement provided to the cows during the test period (the only other diet source at the pasture farms) were taken for dioxin analysis weekly; these were combined at the end of the test period and a two-liter aliquot taken for dioxin analysis.

*Milk:* At each farm samples were taken from the stirred bulk tank, continuously for the entire month before every collection of liquid milk by the bulk milk tanker. Milk sampling took place every 1-3 days depending on the tanker pick-up schedule. Samples were collected using a site-specific cleaned stainless steel milk ladle, and stored in 125 ml pre-cleaned Teflon bottles. Each sample was stored, frozen, until the end of the test period; they were then thawed, and aliquots of each of the samples were mixed to produce a one liter composite sample of the entire milk production during the test period. One duplicate sample and one blank (empty bottle opened during milk sampling) were collected at one farm in each state. The samples were shipped, frozen, in pre-cleaned bottles for analysis at the MRI laboratory. The samples were analyzed for 17 dioxin congeners; the fat content of the milk samples was also determined.

### Air-to-Crop Samples

To compare the dioxin content of diet components grown on-farm during the test period with the dioxin concentration in the air at the farm during that same period, the following samples were taken:

*Pasture:* A representative pasture that had just been grazed was fenced off at the start of the test period and allowed to grow, untouched, during the next month. Twenty-two (22) liters of vegetation were clipped from a total of eleven randomly selected sections of the pasture, and stored in two-liter pre-cleaned glass jars. The major pasture components, grass, clover, forbs (small herbaceous plants), were then hand-separated, densified in a stainless steel Hobart food cutter, to provide one two-liter sample of each pasture component for dioxin analysis. These samples, six liters total from each farm, contained the dioxin taken up from the air by the vegetation produced during the test period. Such samples were collected at VT-A and WI-A.

*Alfalfa:* Plants were clipped in the field at random sites. At WI-D, two liters of material were collected immediately before the study began, and again on the last day, thus capturing the alfalfa crop produced during the one-month test period. (In Vermont persistent rainy weather prevented alfalfa sampling at the end of the test period.)

### Farm site data

On each farm site-specific observations were made concerning a range of farm management procedures that may have some bearing on the presence of some of the dioxin found on farms. A checklist of observational items is presented in Appendix A.

## **III. RESULTS**

### **A. The Sources of Airborne Dioxin**

Dioxin is produced and introduced into the environment by a considerable number of industrial processes, such as: the manufacture of chlorinated organic chemicals; the production of pulp and paper; the operation of internal combustion engines, electric power plants, and various kinds of incinerators and furnaces. Only a few of these processes — especially paper and pulp production and some chemical

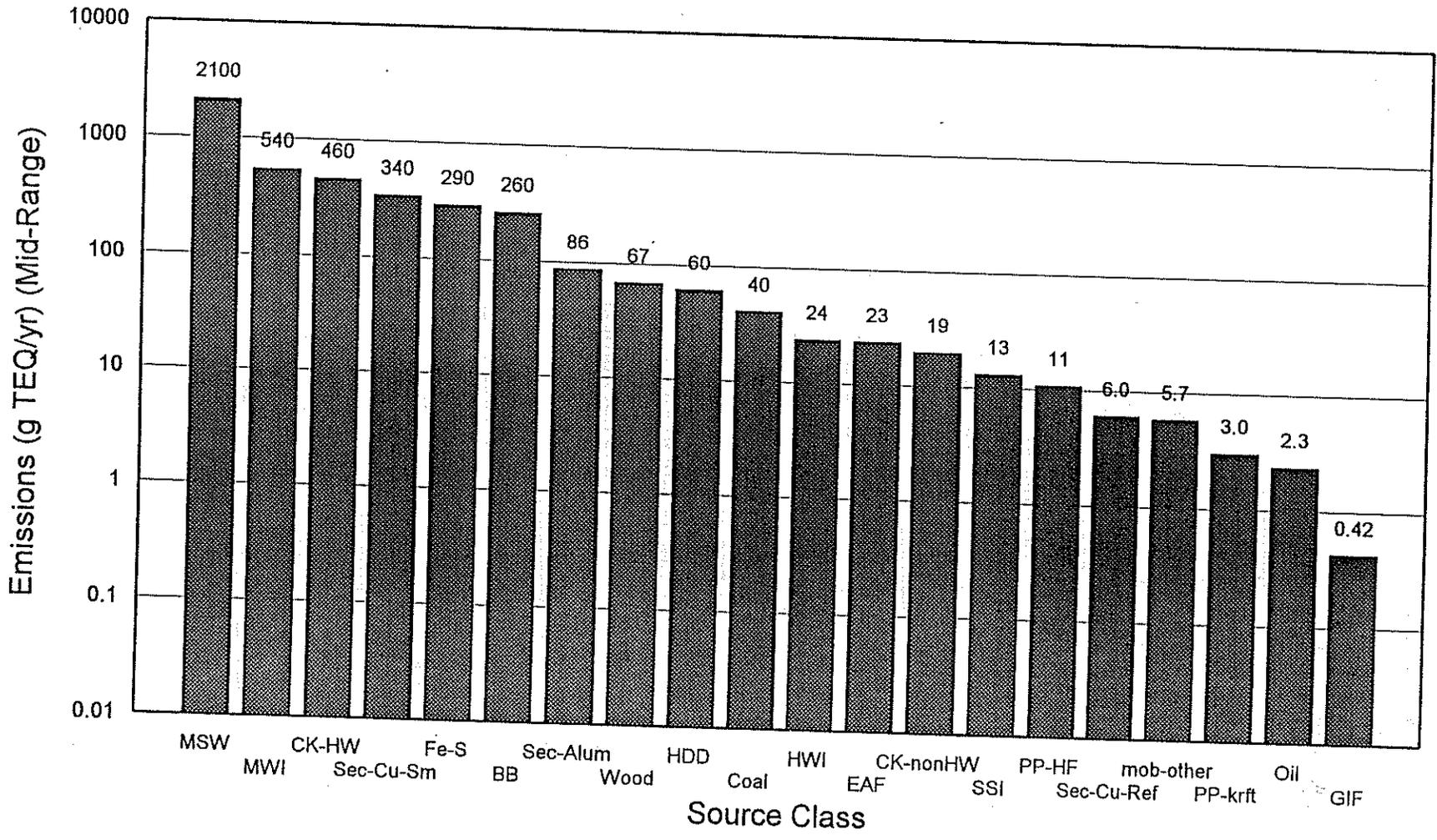
manufacturing — disperse dioxin into surface water; the preponderant route of entry into the environment is the air, largely as a result of combustion processes.

As noted earlier, a basic purpose of this project was to identify the sources of airborne dioxin that are chiefly responsible for the amount deposited on dairy farms, so that action can be taken to prevent emission at the source. As an initial step, therefore, it was necessary to identify all of the sources of airborne dioxin in the United States and, insofar as possible, in Canada as well; determine their geographic location; and estimate the amounts that they emit into the air. This inventory, entered into the air transport model, leads to estimates of the amount of dioxin that each of the sources deposits on the test farms.

For the purposes of this project we reviewed our earlier (1993) inventory of dioxin sources and brought it up to date as of July 1997. The assembly of an inventory is a complex process, subject to a number of uncertainties (see Appendix B for details). Figure 1 summarizes the overall result, which is based on the most probable estimate (i.e., the midpoint of the minimum and maximum values) of annual emissions from each of 20 types of sources. The total annual emissions of dioxin from sources in the United States and Canada is 4,350 grams TEQ, of which 3,890 grams TEQ is generated in the United States and 460 grams TEQ in Canada. Given the uncertainties inherent in such inventories (see Appendix B), the actual values may be several times greater or smaller than the mid-point.

Of the 20 source types that comprise the dioxin inventory, 15 consist of individually identified facilities at specified locations. The five remaining source types cannot be localized because they were mobile (for example, diesel trucks) or too numerous to be individually identified (for example, backyard trash burners). In these cases, emission estimates were based on the number of such sources in each county, with their collective location represented by the county centroid. (In Canada, such sources were localized by metropolitan area and province.) These collective estimates were based on indirect data, such as the density of truck traffic or the amounts of residential trash produced by rural households, together with the appropriate emission

**Figure 1 Summary of Dioxin Emissions Inventory Used in Analysis  
United States + Canada (i.e., Total Inventory)**



see separate table for descriptions of source class abbreviations  
 numbers above bars are estimated midrange emissions (g TEQ/yr), rounded to two significant figures

Figure 1 (continued)

Abbreviations for Source Classes Used in Emissions Inventory Graphs

Source Class Code Number	Source Type Code	Source Class Name	Abbreviation
1		municipal solid waste incinerator	MSW
2		medical waste incinerator	MWI
12	3	cement kilns burning hazardous waste	CK-HW
3		secondary copper smelters	Sec-Cu-Sm
13		iron sintering	Fe-S
20		backyard waste burning	BB
14		secondary aluminum smelters	Sec-Alum
7		wood combustion	Wood
8	1	mobile sources, heavy duty diesel	HDD
6		coal combustion	Coal
10		hazardous waste incinerators*	HWI
15		electric arc furnaces	EAF
12	4	cement kilns not burning hazardous waste	CK-nonHW
5		sewage sludge incinerator	SSI
21		hog fuel/sludge combustion	PP-HF
4		secondary copper refiners	Sec-Cu-Ref
8	2 & 3	mobile sources, other	mob-other
17		kraft black liquor recovery boilers	PP-krft
19		residential oil combustion	Oil
16		grey iron foundries	GIF

\*Including cement kilns that burn hazardous waste.

factors. Thus, the dioxin emissions from such an area-based source would be represented, for example, by the total emissions from the diesel trucks operating in a given U.S. county or Canadian metropolitan area.

The total dioxin inventory consists of 24,644 sources: 5,710 individually identified sources and 18,934 area-based sources. The latter represent the total emissions of all the sources of a given type in each U.S. county and, in some cases, municipal areas as well. As can be seen from Figure 1, 3,730 grams TEQ, or 86% of the total dioxin, was emitted by only five of the 20 types of sources, and only two of these dominant sources, municipal waste incinerators and medical waste incinerators, account for nearly two-thirds of the total emissions.

In sum, the inventory provides the following information about the sources: an estimate of the amount of dioxin emitted annually by all of the sources of a given type; for individually identified sources, such as incinerators, the geographic location (latitude and longitude) and estimated annual dioxin emissions from each of them; for mobile or very numerous sources, emissions estimated by county and the location given by the county centroid in the United States; in Canada, source emissions estimated by metropolitan area and province. The model uses these data to estimate the amount of dioxin that each of the numerous sources contributes to the concentration of dioxin at each of the eight test farms.

#### **B. Evaluation of the Model**

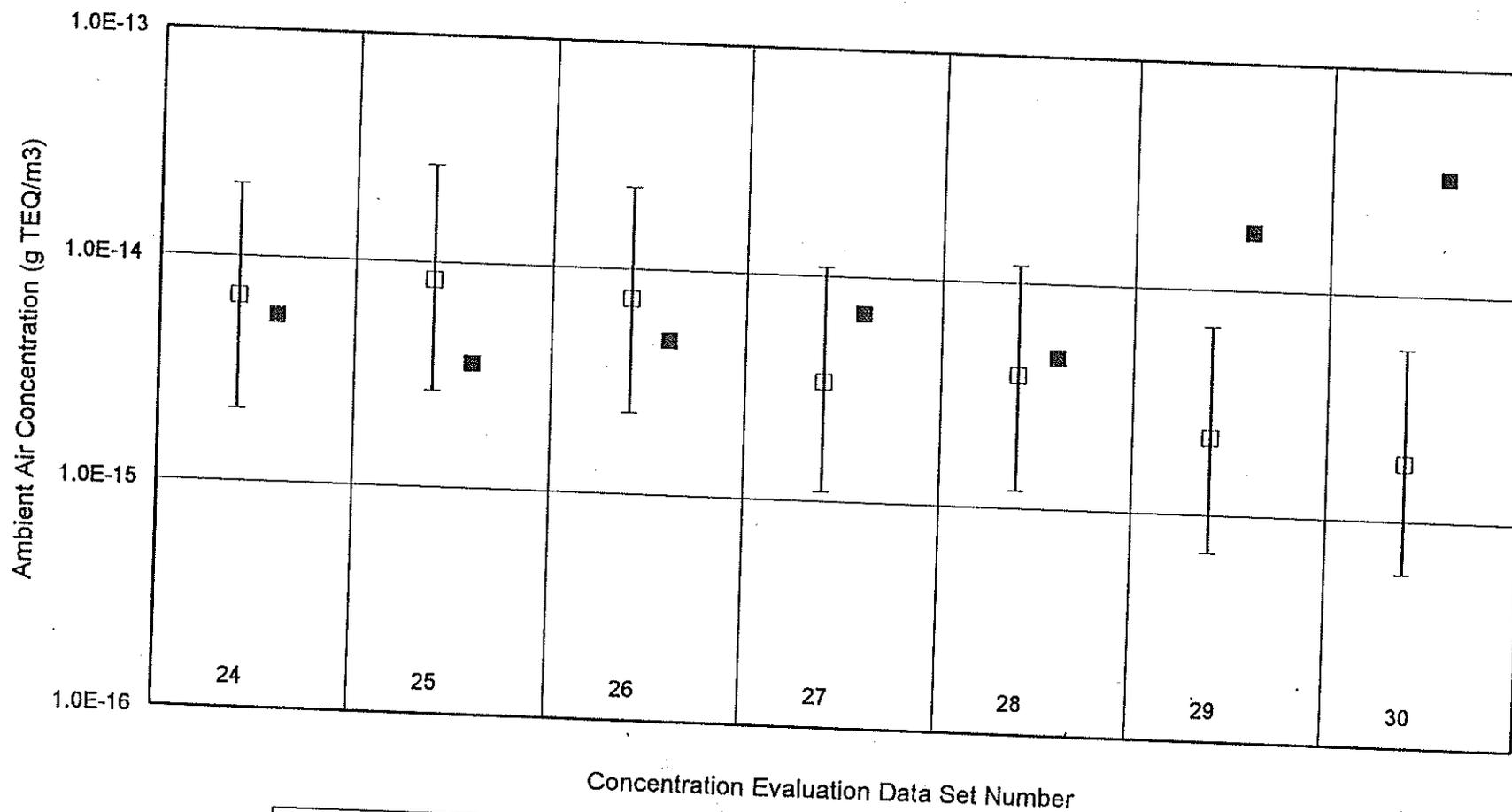
For the purposes of this project, certain modifications were made in the air transport model developed for the earlier (1995) CBNS study of the deposition of airborne dioxin in the Great Lakes. These were: (a) improved treatment of the destructive processes that affect airborne dioxin; (b) characterization of the receptors (i.e., dairy farms) with respect to crop-specific deposition of airborne dioxin. The earlier model had been evaluated by comparing the predicted concentrations of airborne dioxin with actual measurements at a site in Ontario, Canada, over a one-year period (1993). Although there were considerable differences between the weekly values of predicted and measured concentrations, yearly averages of the predicted and measured values agreed quite well.

In the present study, a total of seven comparisons of predicted and measured dioxin concentrations were made. Four of these comparisons were based on air samples collected at farms VT-A, VT-C, WI-A and WI-D during the month-long test periods shown in Table 1. The air samples were collected continuously over the test period by an experienced consultant, ENSR, and analyzed by an EPA-authorized laboratory, Alta Analytical Laboratory, El Dorado, California. Three additional comparisons were based on dioxin analyses of month-long air samples taken by ENSR for the Connecticut Department of Environmental Protection at a remote site on Mohawk Mountain in northwest Connecticut in 1996 (May 13-June 13; August 15-September 12; October 23-November 22). These measurements were compared with our modeled predictions for that site.

The results of these seven comparisons are shown in Figure 2, summarized for all of the separate dioxin and furan congeners, as grams TEQ per cubic meter of sampled air. Five of the seven predicted concentrations are in satisfactory agreement with the measured values. However, at both Wisconsin test sites, the predicted concentrations are significantly less than the measured values; these comparisons are therefore unsatisfactory. The comparisons based on the separate congeners generally lead to the same conclusion; the unacceptable differences between predicted and measured values occur only in the samples from the two Wisconsin test sites.

There are several possible explanations for the failure of the model to satisfactorily predict the measured values at the Wisconsin test sites. First, the characteristic complexity of wind patterns in the Midwest, which involves localized diurnal variations in vertical mixing, may be a factor. This phenomenon does not occur significantly in the Northeast. Second, errors in the emissions inventory may have underestimated the dioxin emissions that affected the Wisconsin sites. These factors are particularly important at WI-D, where the difference between measured and modeled values is greatest. This farm is on a Mississippi River bluff only a few kilometers downwind from a municipal waste incinerator located on an island. As a result, a very localized wind channeled by the river bluff may carry dioxin very efficiently to WI-D, but will not be reflected in the model's relatively coarse meteorological data,

Figure 2 Comparison of Model Predictions with Ambient Measurements at Month-Long Sample Sites, Total Dioxin (TEQ)



■ meas (mid-range) — meas (hi/low) □ model (mid-range) — model (hi/low)

1-month samples: Mohawk Mtn CT (24-26); Northern VT (27); Southern VT (28); Southeast WI (29); Western WI (30)

which is based on a 183 km grid. In the dioxin inventory, the incinerator in question was assigned a very low emission factor, apparently based on a single dioxin test in 1988, soon after it was built, and may not accurately reflect its actual operating condition in 1996.

A fourth possibility — that the discrepant results at the Wisconsin test sites are due to faults in the model itself — appears to be unlikely. Such built-in faults in the model would likely affect the results at all sites rather than only the two in Wisconsin. In addition, all the comparisons reported in Figure 2 were made after the original model had been extensively modified. Yet the comparisons that were made at the same sites before the model was modified produced essentially identical results. This leads to the conclusion that the model itself is relatively robust — that is, it is not overly sensitive to the influence of the built-in methodological assumptions and, by analogy, to any errors that might affect these methodological factors. Finally, as noted in section C below, the results of analyses of the model's data with respect to the effects of source/receptor distance, geographic orientation and source type are notably consistent with the actual mapped position of the sources relative to the receptors at all sites, including those in Wisconsin. Again this suggests that the model itself performs accurately, and that the discrepancies noted at the Wisconsin test sites are due to an apparent discrepancy in the dioxin emission estimate for a large local source or to localized meteorological effects.

In sum, the model is capable of reasonably accurate estimates of the transport of the airborne dioxin emitted by the sources to receptors in the general region of Vermont and Connecticut. In the region represented by Wisconsin, however, the model seriously underestimates the amounts of dioxin that reach the receptors. As noted below, only the actual measurements of airborne dioxin are used as indicators of the level of exposure of farm crops and milk.

## C. The Contribution of the Different Sources to the Airborne Dioxin That Reaches the Test Farms

### Analytical Procedures

The HYSPLIT/TRANSCO model was used to evaluate the degree to which the different sources of dioxin contribute to the amount that reaches the test farms VT-A, VT-B, WI-A and WI-D in Vermont and Wisconsin. The model generated the following basic data: The concentration of dioxin in the air (as pg TEQ per cubic meter of air) at each test farm during the month-long test period; and the dioxin contributed to this concentration by each point source (for example, a municipal waste incinerator in Bridgeport, CT) and each area source (for example, diesel trucks operating in Nassau County, NY).

These data reflect the influence of several key factors that govern the relationship between sources of dioxin and a receptor: the amount of dioxin emitted by the various sources; their linear distances from the receptor (the amount that reaches a receptor falls with increased distance from the source); and the geographic orientation of the sources relative to the receptor (dioxin air transport will be more efficient in the direction of the prevailing winds). The influence of each of these factors on the transport of dioxin between the sources and the receptors was derived from the basic model-generated data by means of several analytical procedures:

*Source-receptor distance:* For a given receptor, e.g., a test farm, the total inventory of sources was segregated into a series of concentric rings with respect to their increasing distance from the receptor. It was then possible to estimate, for the sources located within each distance range, the total amount of dioxin they emitted and their contribution to the concentration of the airborne dioxin at the receptor. These data were expressed as the percentage of the dioxin emitted by all sources that is due to those within each distance range; and, similarly, as the percentage of the total concentration of dioxin at the receptor that originates from the sources in each distance range.

*Source-receptor geographic orientation:* The total inventory of sources was

divided into four quadrants (NE, SE, SW, NW) centered at the location of a given receptor. The percentages of the total emissions and of the total dioxin concentration at the receptor that are due to the sources in each quadrant were then estimated.

*Source type-receptor relation:* The total inventory of sources was segregated according to source type, based on the classification shown in Figure 1. For a given receptor, the percentages of the total dioxin emissions and of the total dioxin concentration at the receptor arising from each of the source types was estimated.

*Ranking of individual source contributions to dioxin concentration at receptors:* Since the model estimates the fraction of the total concentration of dioxin at the receptor that originates from each of the numerous sources, these data can be used to rank the individual sources in this respect. In order to visualize the effect of the most important individual sources, it is useful to limit this plot to the 1000 highest-ranked sources (see below).

By analyzing the foregoing data, it was possible to characterize the sources that are chiefly responsible for the dioxin that reaches the test farm sites.

#### Northern Vermont

The receptor is farm VT-A. The relevant data are shown in Figures 3.A, 3.B, 3.C and 3.D. Figure 3.A describes the effect of distance on the source-receptor relationship; most (72%) of the dioxin is emitted from sources that are 400-2,000 kilometers from the Northern Vermont test site. However, most (74%) of the airborne dioxin that reaches the test site comes from sources that are closer to the receptor, 200-1,000 km away; about 7% comes from sources in the 40-200 km range, with the rest scattered among several other ranges.

Within each distance range, the relation between the amount of dioxin emitted and the concentration at the test site indicates the efficiency of air transport between the sources and the receptor. For example, although the sources in the 200-400 km range produce only about 8% of the total dioxin emissions, they account for 26% of the dioxin concentration at the receptor. In contrast, while the sources in the 1,000-2,000 km range produce 36% of the total emissions, they account for only 16% of the

Figure 3A. Dioxin (TEQ) Emissions and Air Concentration Impact as a Function of the Distance of Sources from the Northern Vermont Sampling Site

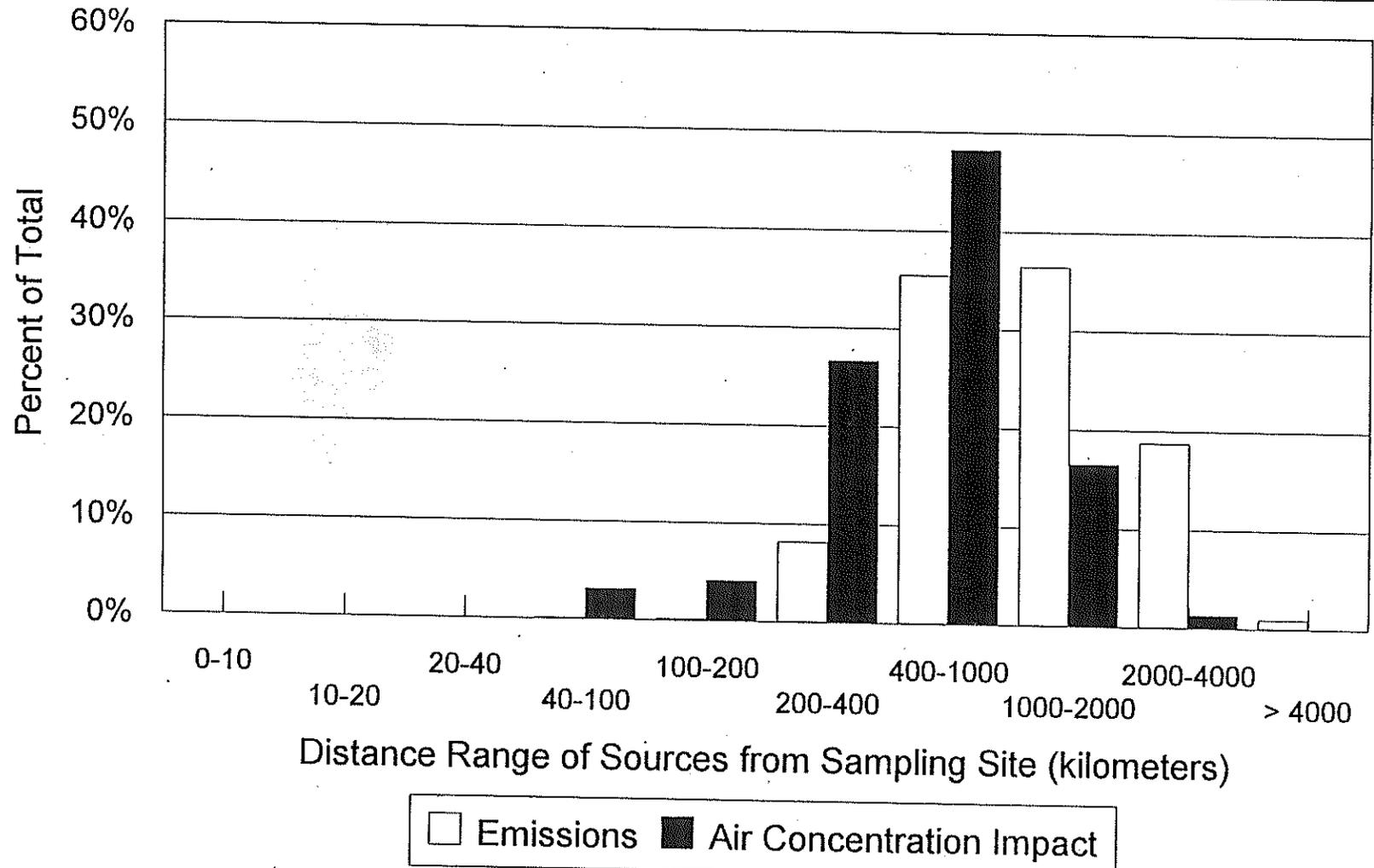


Figure 3B. Dioxin (TEQ) Emissions and Air Concentration Impact from Different Directional Orientations for the Northern Vermont Sampling Site

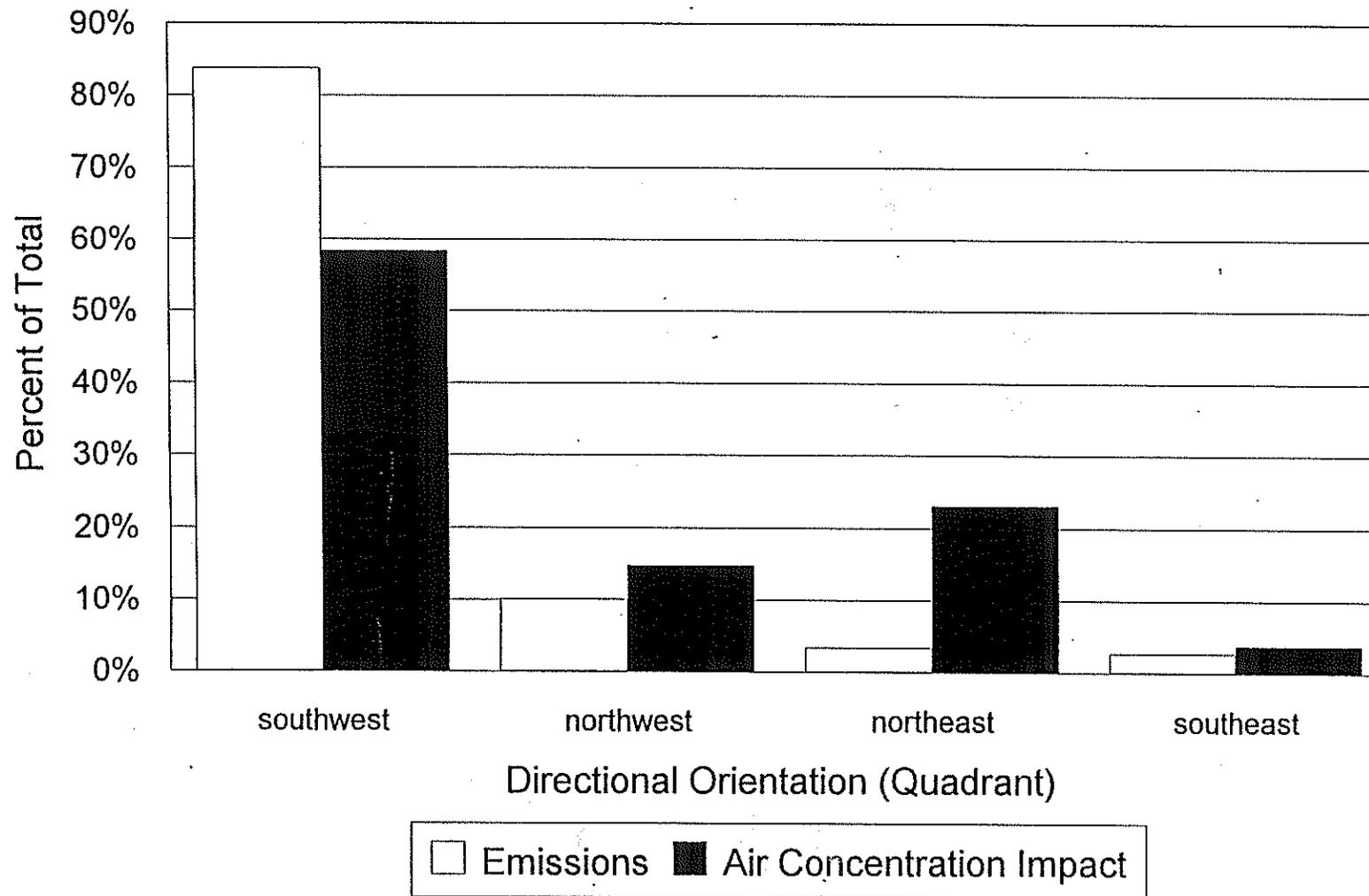


Figure 3C. Dioxin (TEQ) Emissions and Air Concentration Impact of Different Source Types for the Northern Vermont Sampling Site

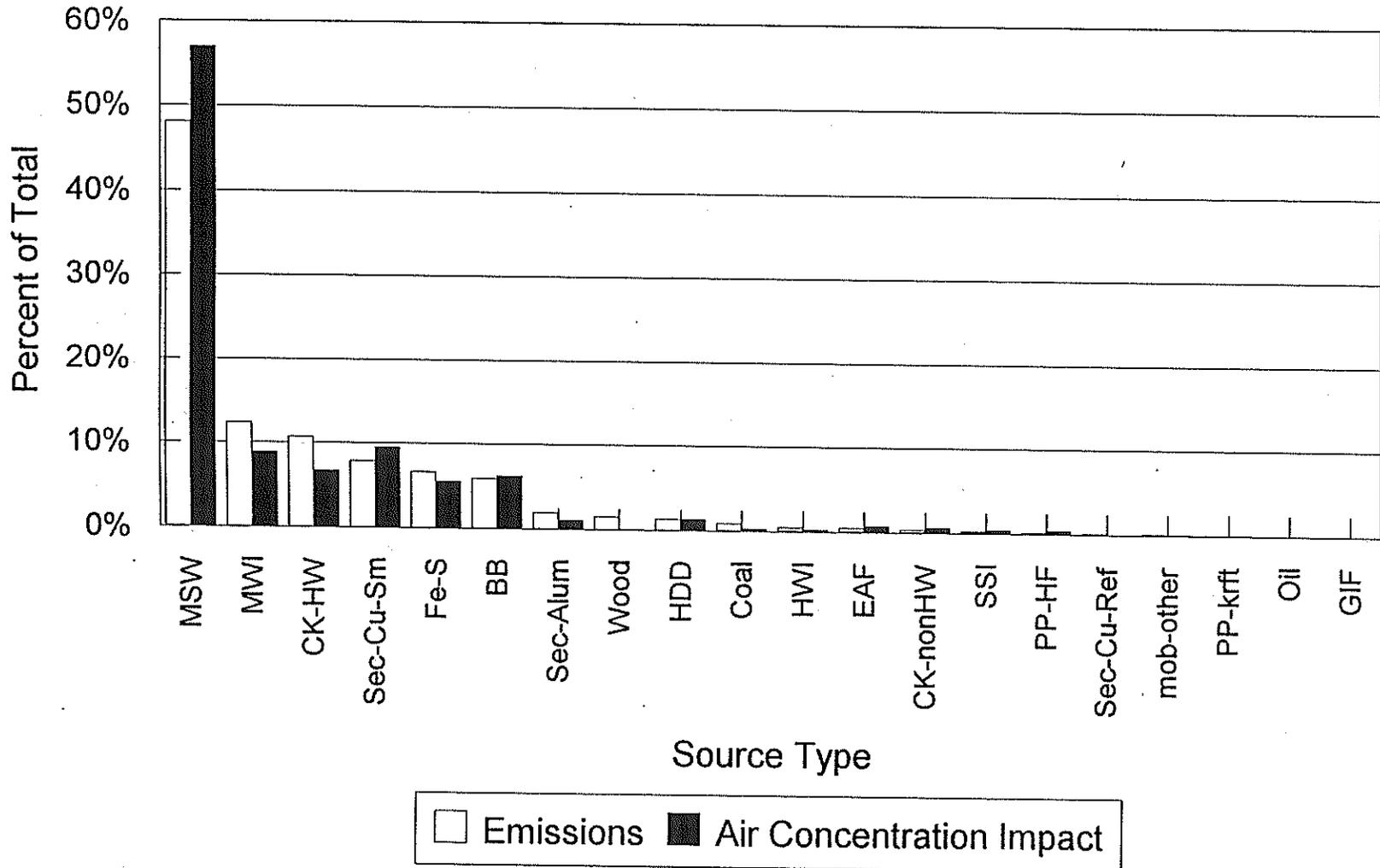
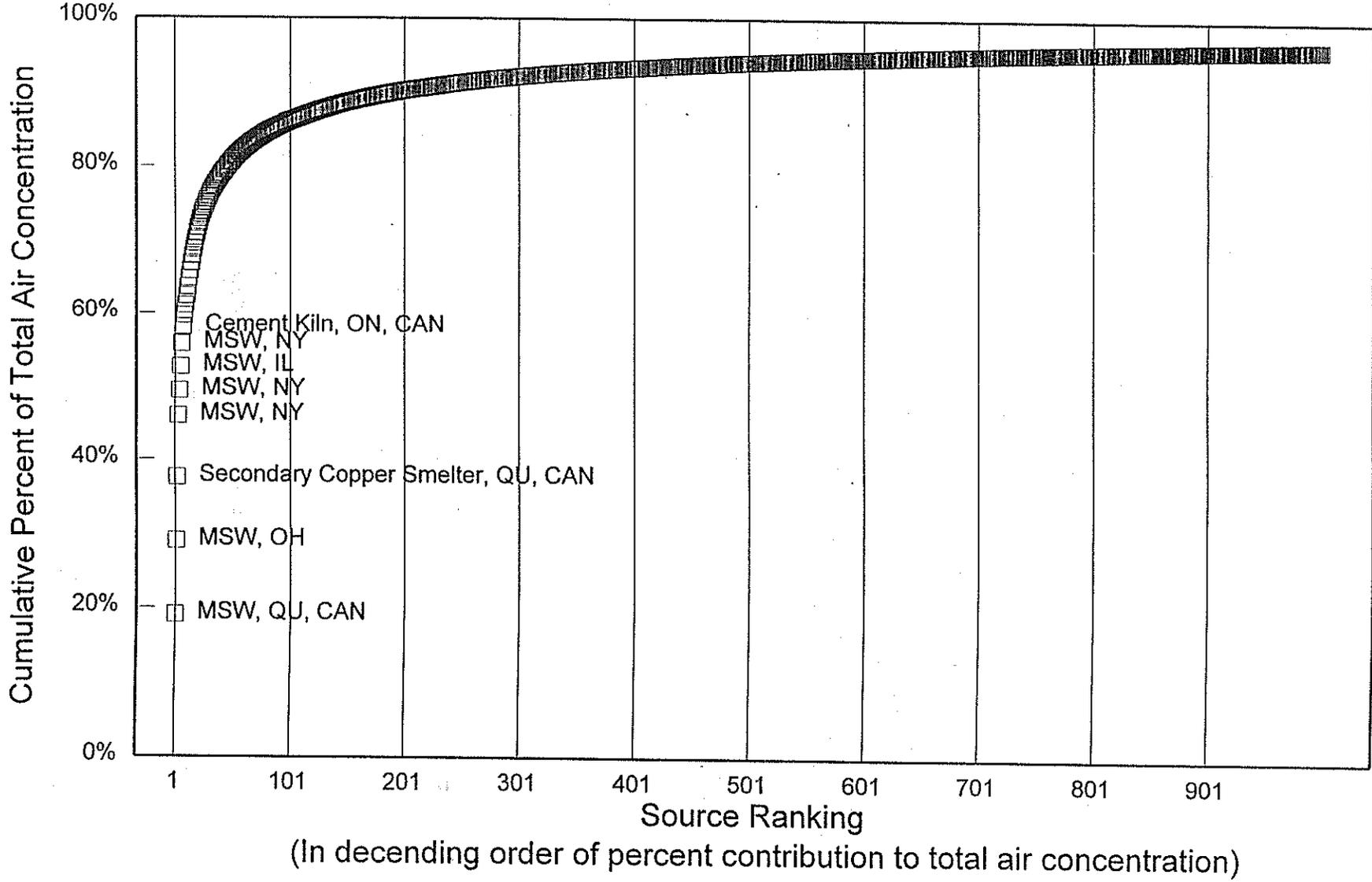


Figure 3-D Contribution of 1000 Highest-Ranked Sources to Air Concentration of Dioxin (TEQ) at Farm VT-A (Northern Vermont)



airborne dioxin concentration at the test site. Thus, the efficiency with which emitted dioxin is transported to the test site receptor decreases as the source-to-receptor distance increases. This results from the processes that occur during air transport that prevent dioxin from reaching the receptor: diffusion, destruction and deposition en route. That sources as far as 1,500 km from VT-A contribute significantly to the farm's concentration of airborne dioxin confirms our earlier observations of appreciable dioxin transport to the Great Lakes from sources as distant as Texas.

Figure 3.B illustrates the effect of geographic orientation on the efficiency of dioxin transport between the sources and the receptor. This shows that airborne dioxin is more efficiently transported to the receptor from those sources that are in the NW and NE quadrants than those in the southern quadrants. Since VT-A is located in the northeast corner of the United States, most (84%) of the source emissions lie to the southwest. However, the emissions from the sources in the SW sector are not efficiently transported to the receptor, for they account for only 58% of the airborne dioxin concentration there. In contrast, the emissions from sources in the NW and NE sectors, which together represent only 13% of the total emissions, are transported much more efficiently, contributing 38% of the airborne dioxin at the receptor. These results reflect the influence of wind direction, which is likely to come from the north of this receptor during August.

Figure 3.C characterizes the source/receptor relationship with respect to source type. The relative emission values of the 20 source types reflect the pattern evident in the total inventory of sources in which emissions from municipal waste incinerators are dominant (see Figure 1). Most of the airborne dioxin concentration at the receptor appears to come from municipal waste incinerators and is efficiently transported; thus, their contribution to airborne dioxin at VT-A (58%) is greater than their contribution to total emissions (48%). The only other source type that is efficiently transported to this receptor is secondary copper smelters, which account for 10% of the receptor's airborne dioxin concentration, but for only 8% of the emissions.

Figure 3.D shows the cumulative effect of the 1,000 highest-ranked sources on

the concentration of airborne dioxin at VT-A. Although these sources represent only 4% of the total inventory of 24,644 sources, they account for 98% of the airborne dioxin concentration at the receptor. The eight highest-ranked sources, which are identified in Figure 3.D, together account for more than half of the total dioxin concentration. The highest-ranked individual source — which accounts for about 20% of the total concentration — is a municipal waste incinerator in Quebec, Canada; a similar incinerator in Ohio accounts for an additional 11%; a secondary copper smelter in Quebec adds 10% more; and four municipal waste incinerators in New York and Illinois and a cement kiln in Ontario, Canada, bring the eight-source contribution to nearly 60% of the total airborne dioxin concentration at the receptor. That, according to Figure 3.D, a single secondary copper smelter contributes 10% of the total dioxin concentration at the receptor is confirmed by Figure 3.C, which shows that the contribution of all secondary copper smelters, as a class, is also 10%.<sup>2</sup>

Finally, maps of the location of the major sources help to further refine their relationship to the receptor. Figure 7, which maps the location of secondary copper smelters in the United States and Canada, confirms that, as expected, there is a single secondary copper smelter, in Quebec, Canada, it lies northwest of the VT-A receptor, about 800 km from it. The impact of this source should also be reflected in the data regarding sources in the NW sector. As shown in Figure 3.B, this sector accounts for 15% of the airborne dioxin concentration at the receptor, so that the additional 5% must be due to some sources other than the smelter. As indicated by Figures 8 and 9, the NW sector also includes a municipal waste incinerator and a number of medical waste incinerators that are sufficient to account for the additional 5%.

These maps also help to explain an observation evident in Figure 3.B: the

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<sup>2</sup> However, it is important to note that the quantitative impact of any such single source is subject to considerable uncertainty, particularly because of the wide range in the possible emission rates computed from emission factors rather than direct measurements of dioxin emissions. Given these uncertainties, such individual modeled estimates need to be evaluated against direct measurement of the source's actual emissions of dioxin. In effect, ranking the individual sources identifies those sources that merit direct analysis of their dioxin emissions to confirm their high estimated impact on the receptor.

Figure 7 Location of Secondary Copper Smelters



Inventory : July 1997

Total Number: 8

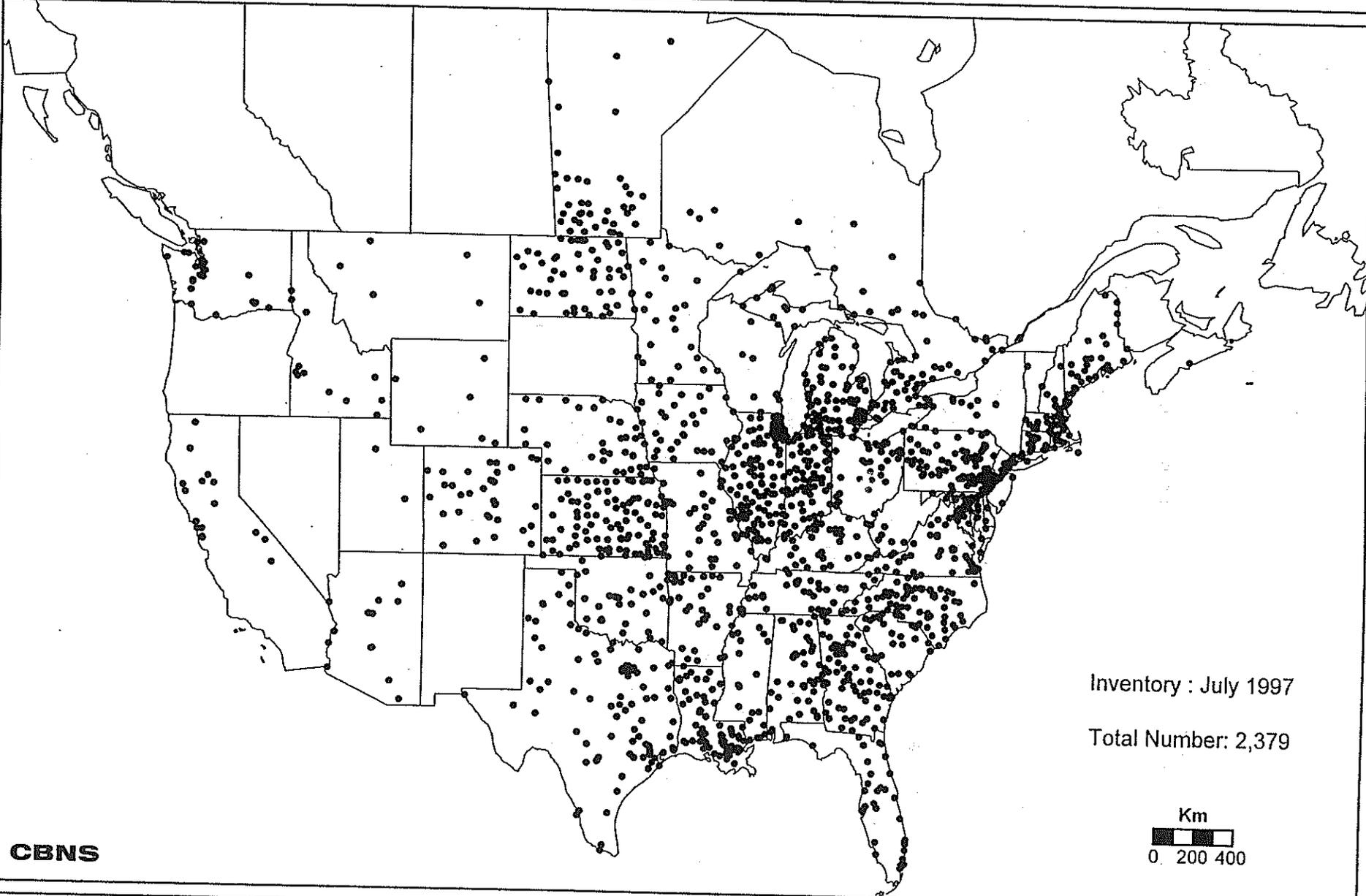
Km  
0 200 400

CBNS

Figure 8 Location of Municipal Solid Waste Incinerators



Figure 9 Location of Medical Waste Incinerators



relatively low transport efficiency exhibited by sources in the SW sector. As Figures 8 and 9 show, while there are numerous incinerators southwest of VT-A, most of them are at a considerable distance from this receptor, and are therefore subject to large losses en route. In contrast, Figure 3.B shows that sources in the NE sector are transported to the receptor with a notably higher efficiency; as Figures 8 and 9 show, there are a number of relatively close municipal and medical waste incinerators in this sector.

#### Central Vermont

The receptor is farm VT-C. Figures 4.A, 4.B, 4.C and 4.D describe the influence of the several source characteristics on the emissions and airborne dioxin concentration at the receptor. Figure 4.A shows that two distance ranges, 100-200 km and 400-1,000 km, account for most (82%) of the airborne dioxin at the receptor, although the dominant emissions are from sources in the 400-1,000 and 1,000-2,000 km ranges. Figure 4.B shows that 80% of the airborne dioxin at the receptor comes from sources in the SW and NW quadrants; again, as in the case of VT-A, air transport from sources in the SW quadrant is relatively inefficient compared to sources in the other sectors. Figure 4.C shows that the source type that dominates the airborne dioxin at the receptor (nearly 56% of the total) is municipal waste incinerators.

In common with VT-A, air transport to VT-C from municipal waste incinerators is relatively efficient; the same is true of emissions from secondary copper smelters and backyard trash burners. Figure 4.D is a plot of the cumulative contributions of the 1,000 highest-ranked sources to the airborne dioxin concentration at the receptor. With certain notable exceptions, the results are similar to those characteristic of VT-A. Only eight individual sources account for nearly 60% of the total concentration; the highest-ranked source is a municipal waste incinerator in Quebec; the secondary copper smelter in Quebec contributes 9% to the total concentration, slightly less than the effect on VT-A. A major difference between the two Vermont sites is that at VT-C, backyard waste burning appears among the eight high-ranked sources, accounting for 10% of the total airborne dioxin concentration at the receptor, as compared with 6% at

Figure 4A. Dioxin (TEQ) Emissions and Air Concentration Impact as a Function of the Distance of Sources from the Central Vermont Sampling Site

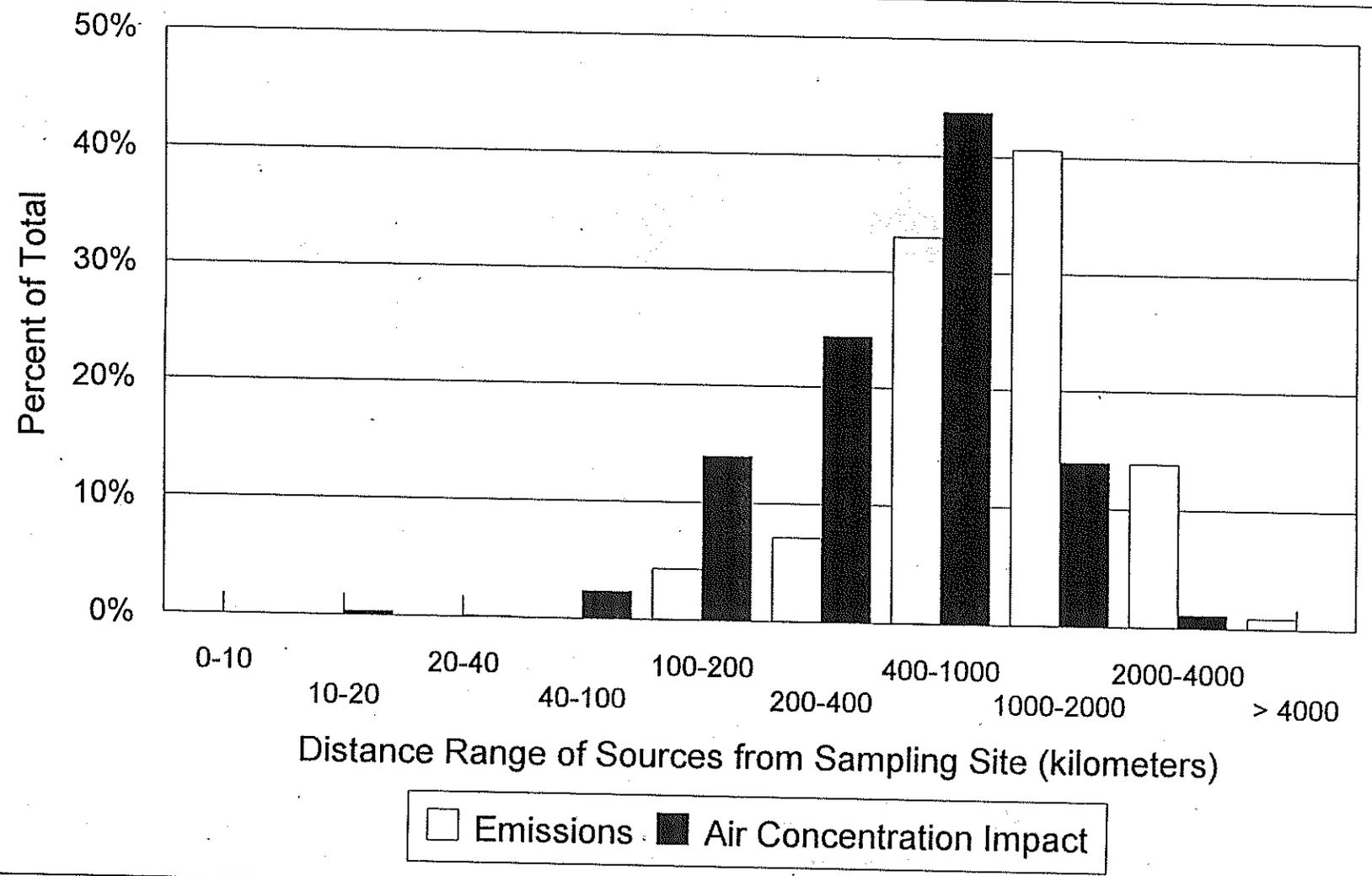


Figure 4B. Dioxin (TEQ) Emissions and Air Concentration Impact from Different Directional Orientations for the Central Vermont Sampling Site

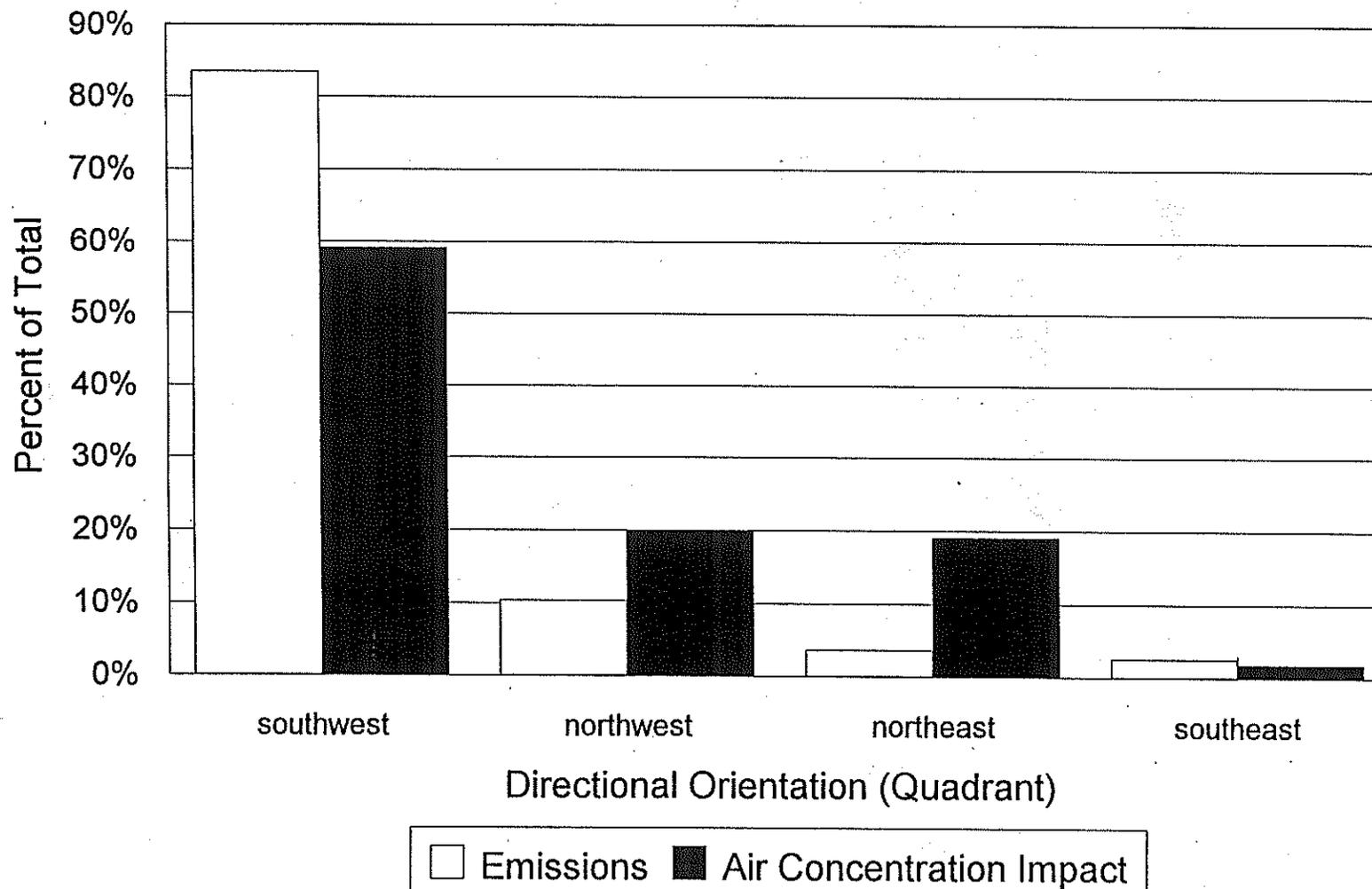


Figure 4C. Dioxin (TEQ) Emissions and Air Concentration Impact of Different Source Types for the Central Vermont Sampling Site

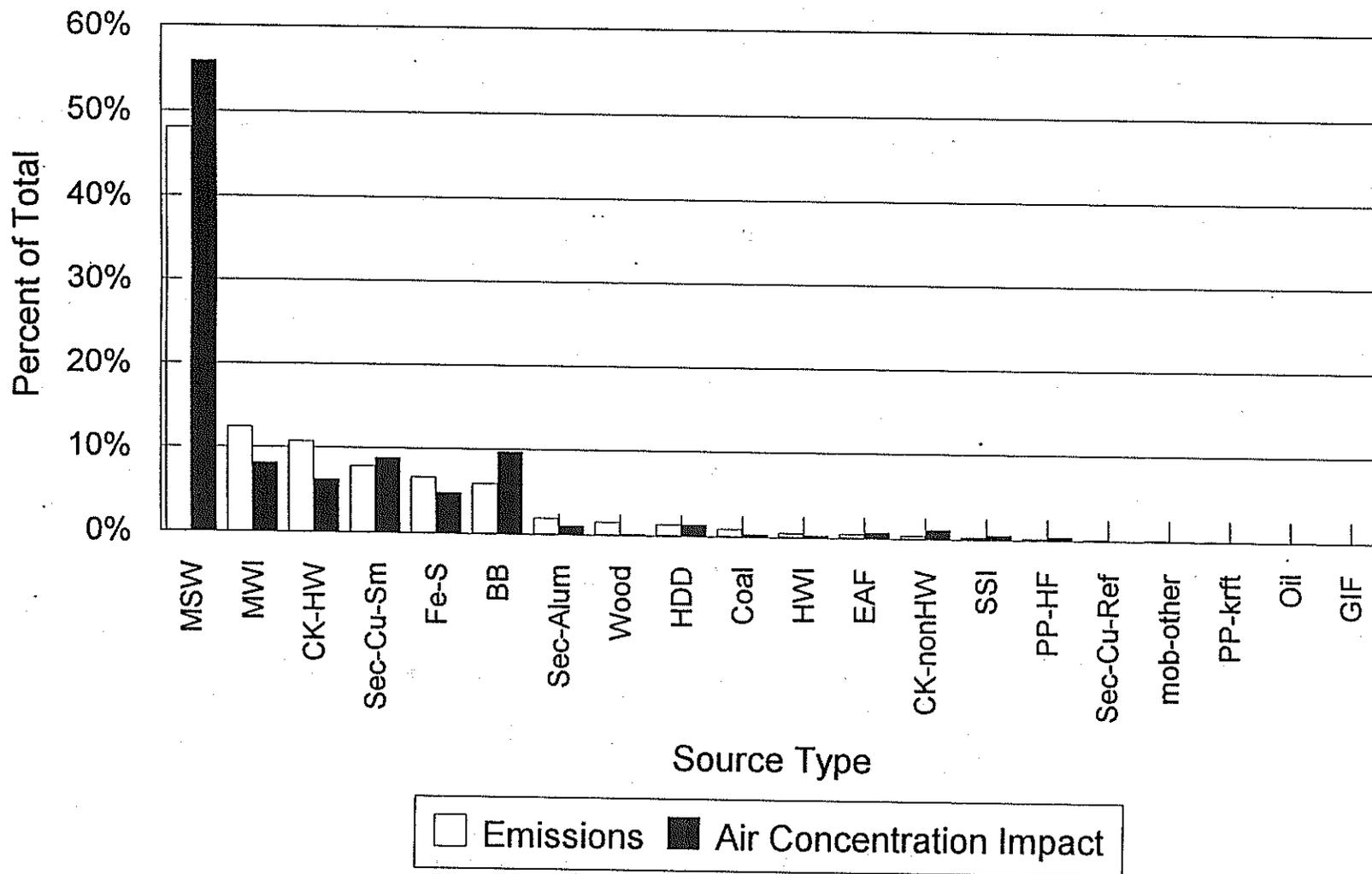
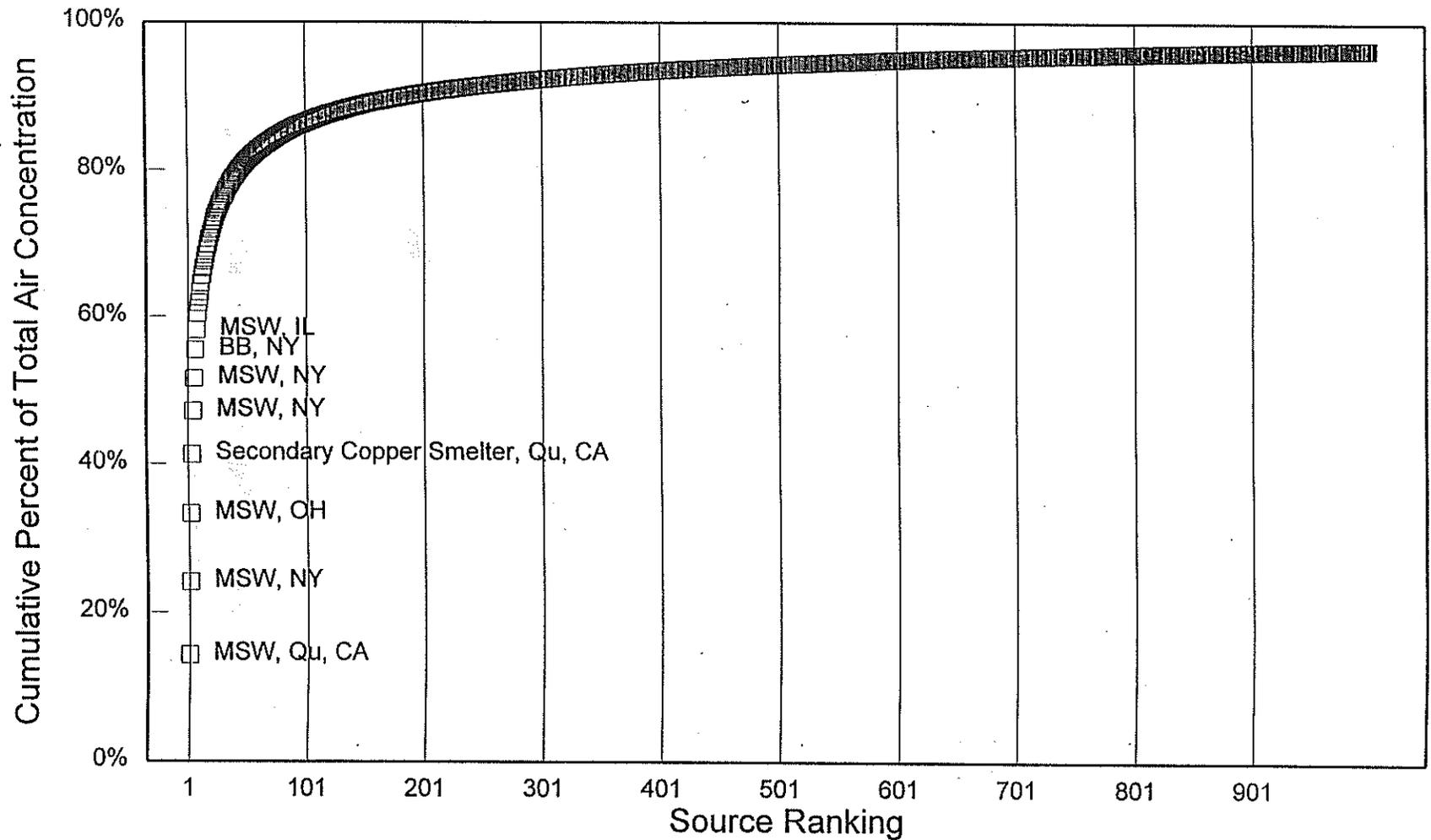


Figure 4-D Contribution of 1000 Highest-Ranked Sources to Air Concentration of Dioxin (TEQ) at Farm VT-C (Central Vermont)



(In descending order of percent contribution to total air concentration)

VT-A. Note that these sources are in New York rather than Vermont; Vermont bans backyard waste burning.

The location of municipal waste incinerators is shown in Figure 8. There are a number of such incinerators southwest of the test site, some of them as close as 150 km and others at distances ranging up to 600 km or more. A smaller group of these incinerators lies due west of the test site, bordering Lake Erie, and appears to account for the dioxin reaching the test site from the NW quadrant. These data also show that, remarkably, despite the heavy concentration of municipal waste incinerators west and south of VT-C in New Hampshire and southern Maine, none of these sources appear among the top-ranked sources shown in Figure 4.D. This is also true of the VT-A test site and appears to result from the strong effect of prevailing winds in the region, which are generally from the north in August.

#### Western Wisconsin

At receptor WI-D, emissions are chiefly (81%) from sources 400-2,000 km distant from the receptor, while the dioxin that reaches the receptor largely (79%) originates from sources 100-1,000 km distant (Figure 5.A). Figure 5.B shows that sources in the SE sector account for 74% of the emissions and 60% of the dioxin concentration at the receptor; air transport is most efficient from sources in the NW sector. Figure 5.C shows that, while — in common with the Vermont sites — municipal waste incinerators have a large effect, at WI-D medical waste incinerators also have an appreciable impact on airborne dioxin concentration (14%). In addition, the dioxin emitted from iron sintering plants and backyard waste burners is transported to this receptor rather efficiently, so that these sources make notable contributions to the airborne dioxin concentration (11% and 13%, respectively).

The cumulative contributions of the 1,000 highest-ranked sources to airborne dioxin concentration at WI-D are noticeably different from those of the Vermont sites. In contrast with the latter, the WI-D curve is less steep. As Figure 5.D shows, the eight highest-ranked sources account for only about 44% of the total concentration, while the comparable figures in Vermont are about 60%. At WI-D the cumulative curve reaches

Figure 5A. Dioxin (TEQ) Emissions and Air Concentration Impact as a Function of the Distance of Sources from the Western Wisconsin Sampling Site

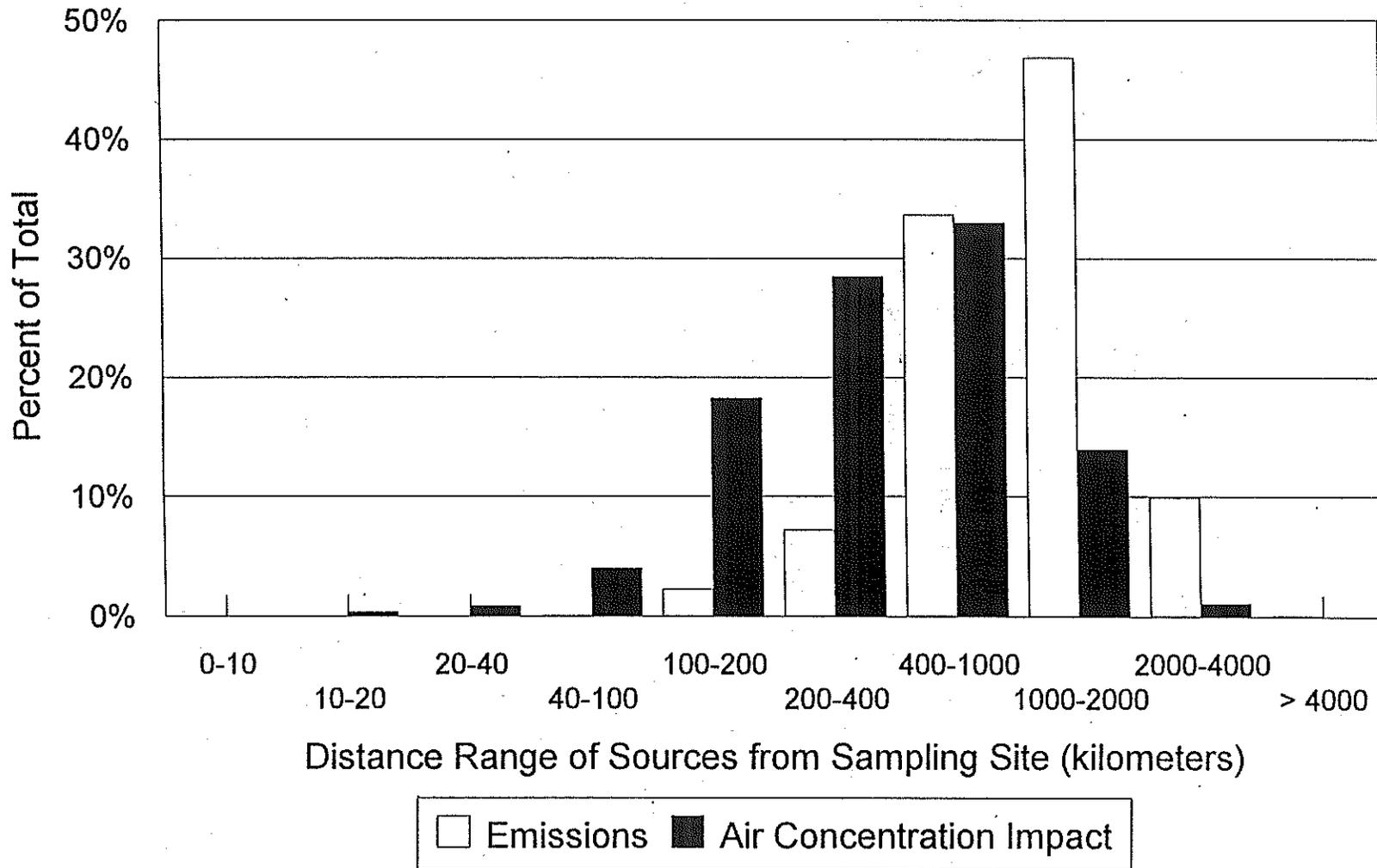


Figure 5B. Dioxin (TEQ) Emissions and Air Concentration Impact from Different Directional Orientations for the Western Wisconsin Sampling Site

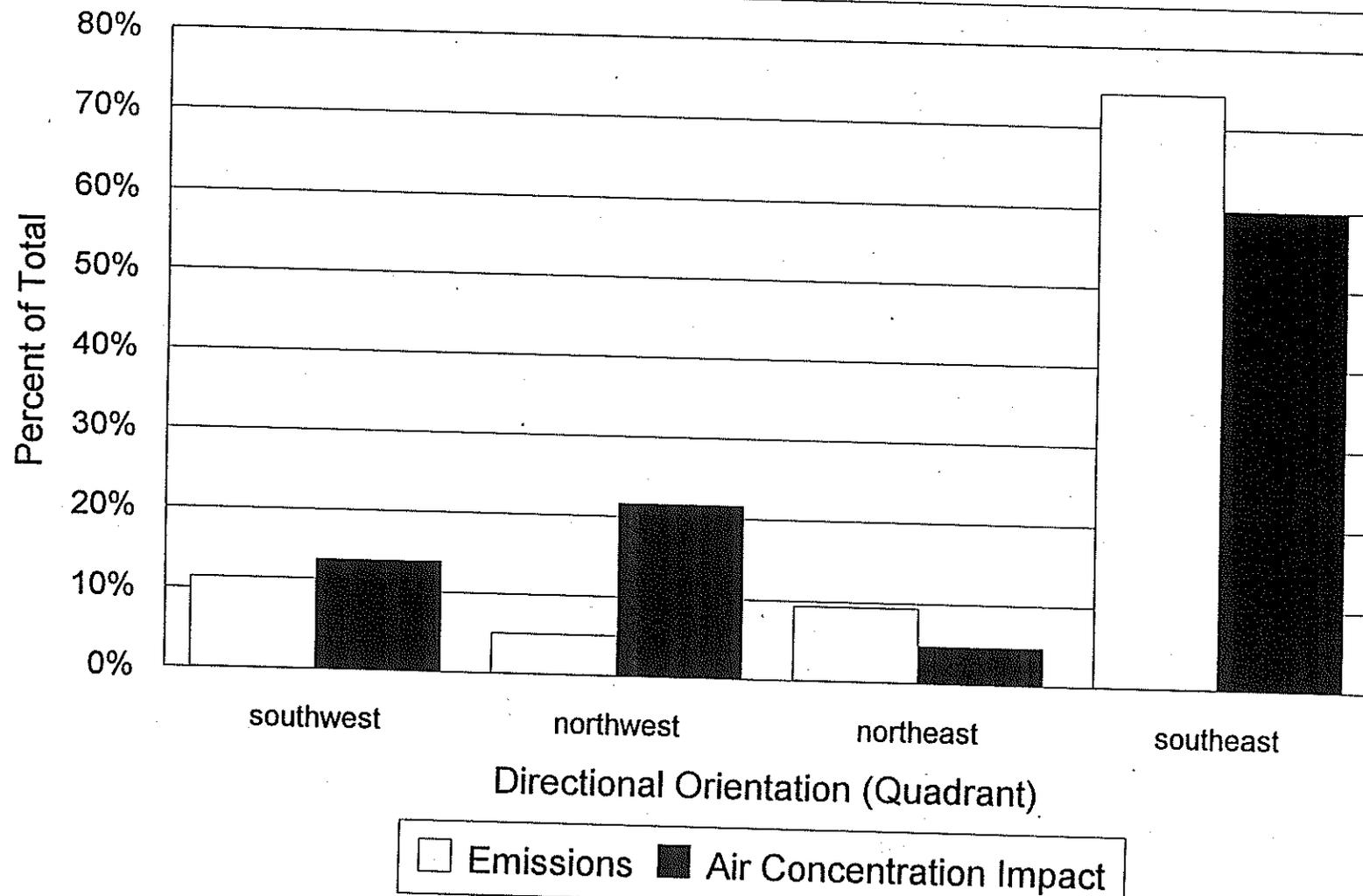


Figure 5C. Dioxin (TEQ) Emissions and Air Concentration Impact of Different Source Types for the Western Wisconsin Sampling Site

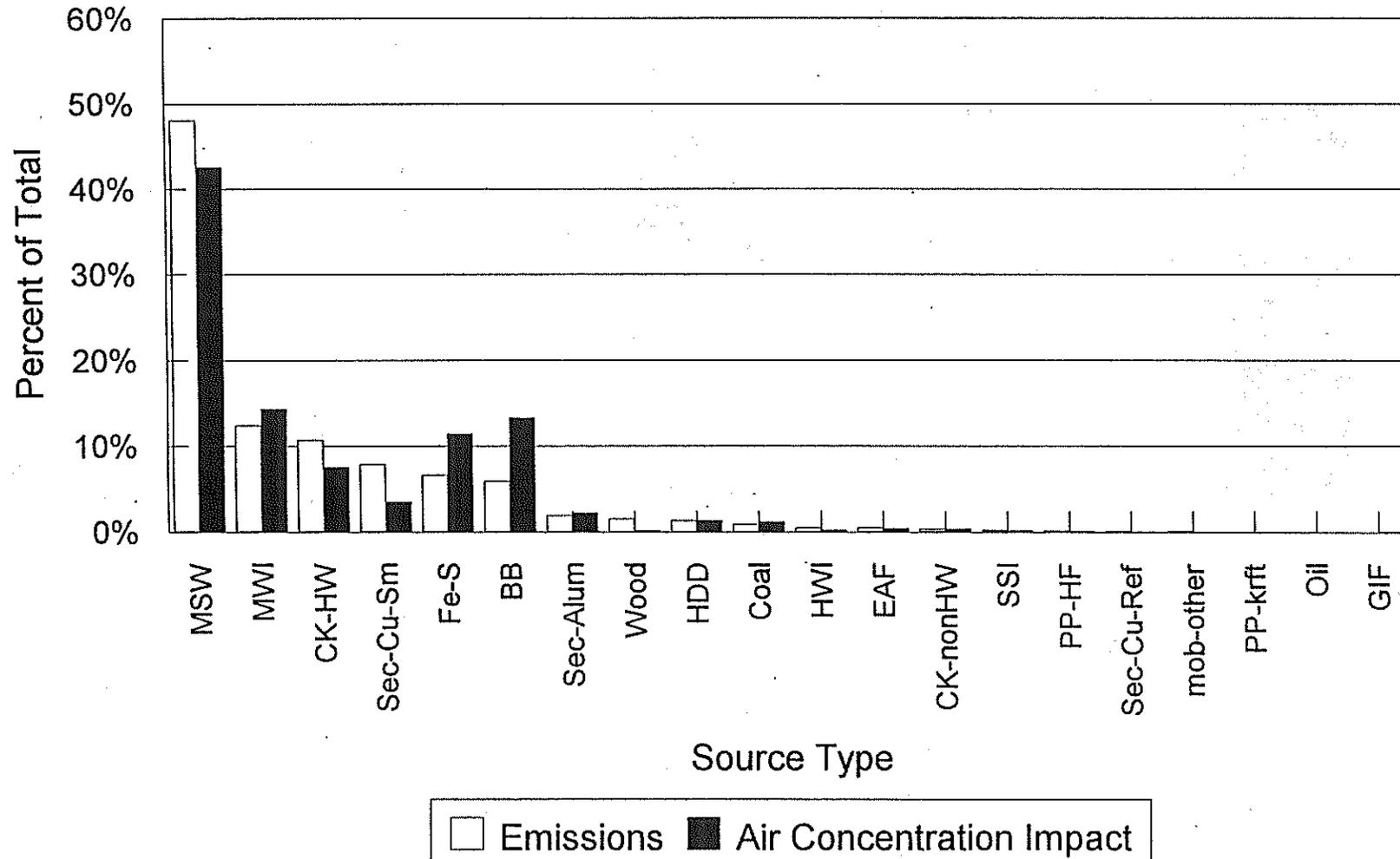
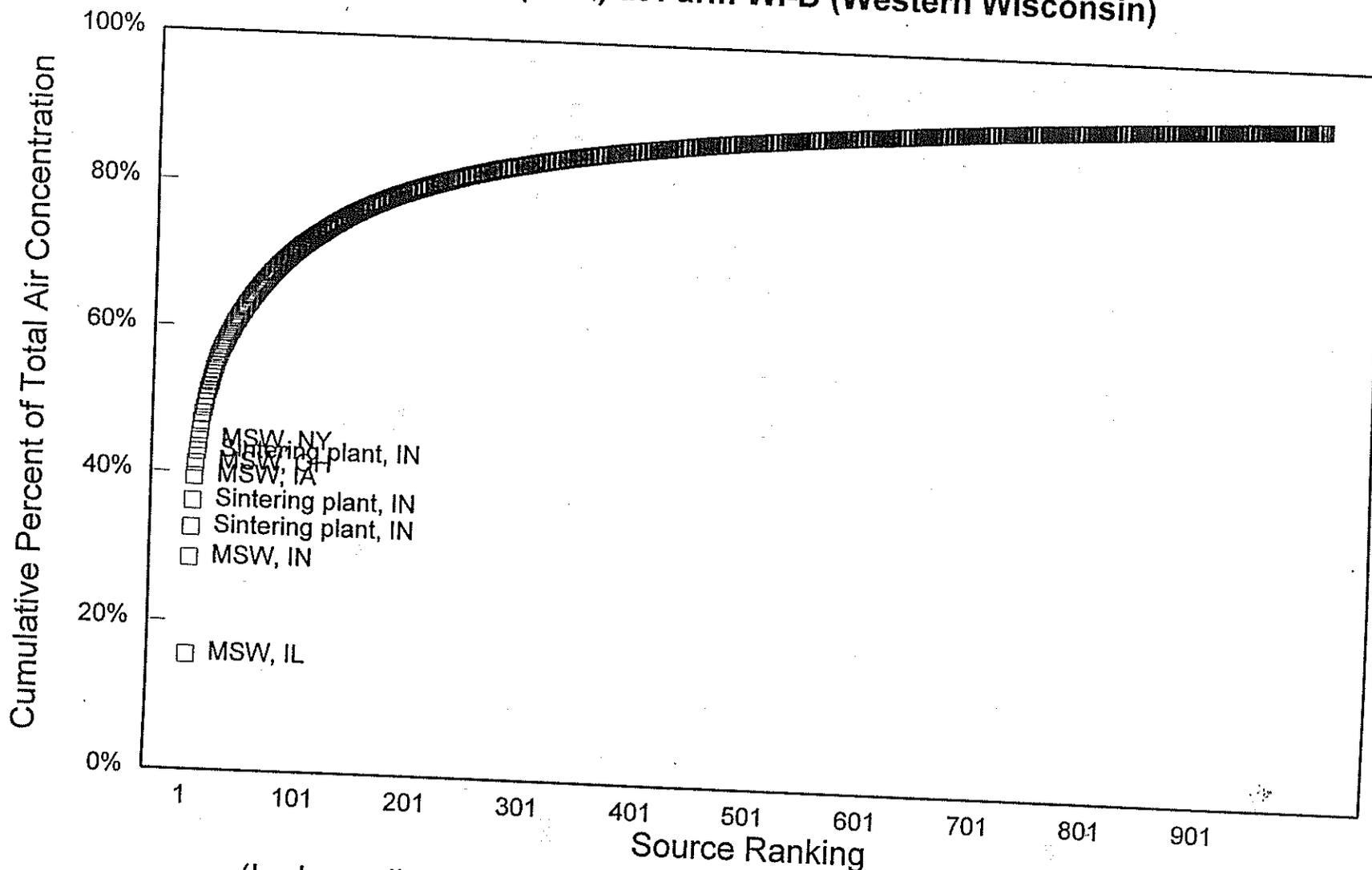


Figure 5-D Contribution of 1000 Highest-Ranking Sources to Air Concentration of Dioxin (TEQ) at Farm WI-D (Western Wisconsin)



(In descending order of percent contribution to total air concentration)

80% of the total concentration with about 200 sources, while the comparable figures at the two Vermont sites is less than 50 sources. These results indicate that, in comparison with the Vermont sites, the dioxin concentration at WI-D is considerably affected by relatively small sources — that is, those which are numerous but individually emit relatively little dioxin. These observations are supported by the map showing the locations of medical waste incinerators, which are numerous, but individually emit relatively low amounts of dioxin. Figure 9 shows that there is a heavy concentration of medical waste incinerators, upwind of Wisconsin in Illinois and Indiana. Figure 9 also helps to explain the minor impact of medical waste incinerators on the Vermont test sites: the eastern half of New York, which, based on proximity and weather patterns might be expected to influence airborne dioxin at the Vermont test sites, is notably free of medical waste incinerators.

#### Southeastern Wisconsin

The receptor is farm WI-A. The effect of source-to-receptor distance (Figure 6.A) shows, as expected, that the efficiency of air transport falls with distance. The emissions chiefly occur in sources at distances ranging from 400 to 2,000 km, while most of the airborne dioxin concentration at the receptor originates from sources at distances of 40-1,000 km. As shown by Figure 6.B, 61% of the emissions come from sources in the SE quadrant, and 59% of the total airborne dioxin concentration reaching the receptor originates from that quadrant. Although municipal waste incinerators are the largest contributors to airborne dioxin concentration at this receptor (39%), medical waste incinerators rank next, at 18% (see Figure 6.C). This unusually high contribution of medical waste incinerators to airborne dioxin at this receptor, and its efficient air transport, suggest that such incinerators are located relatively close to WI-A. Figure 6.D shows that the contributions to airborne dioxin at the receptor of the eight highest-ranked sources are due to seven municipal waste incinerators (in Illinois, Ohio, Minnesota, New York, Indiana, Iowa and Michigan), and two iron sintering plants (in Indiana). Like WI-D—and unlike the Vermont sites—the cumulative dioxin concentration curve is relatively flat, indicative of significant contributions, collectively,

Figure 6A. Dioxin (TEQ) Emissions and Air Concentration Impact as a Function of the Distance of Sources from the Southeastern Wisconsin Sampling Site

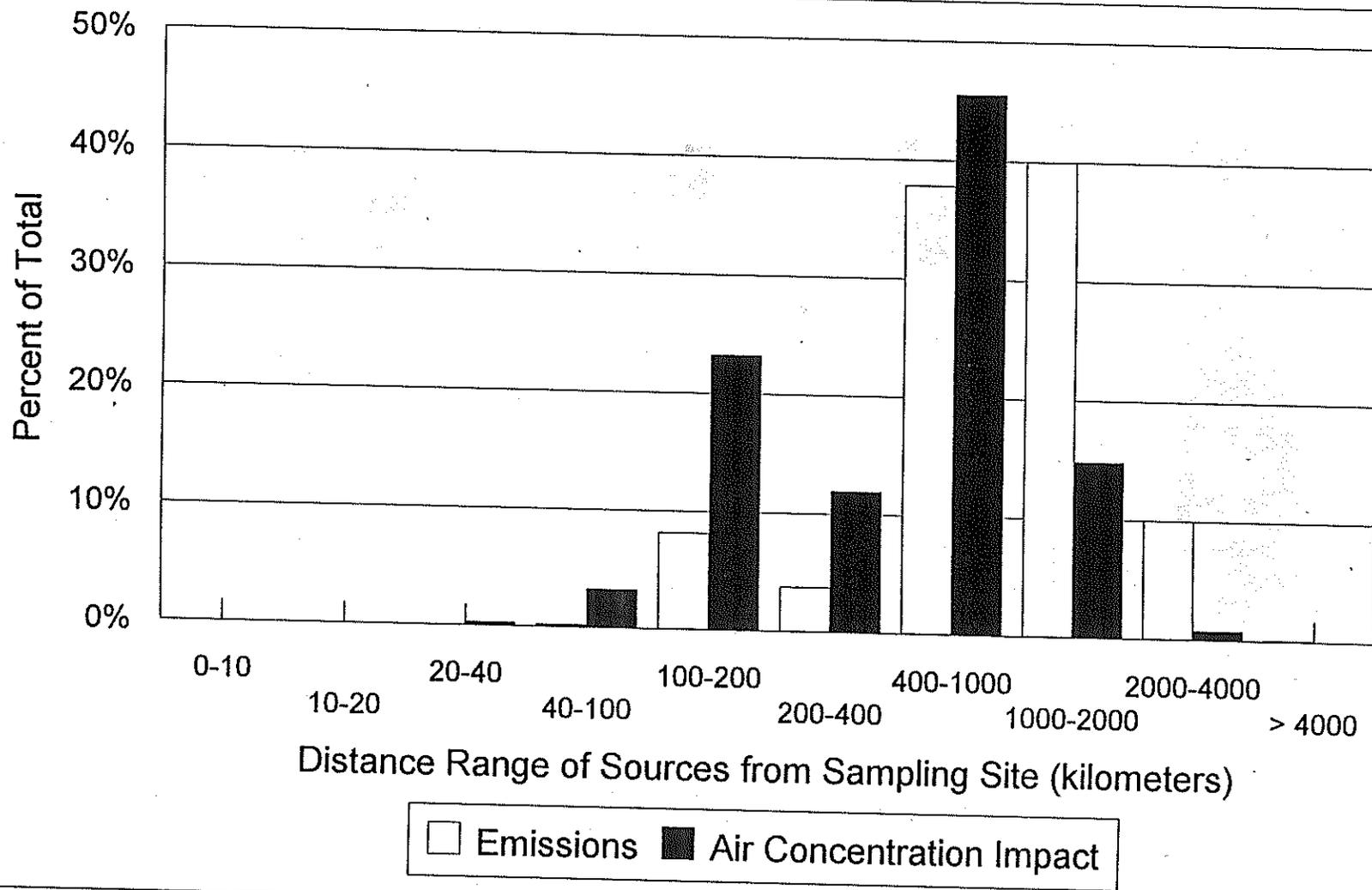


Figure 6B. Dioxin (TEQ) Emissions and Air Concentration Impact from Different Directional Orientations for the Southeastern Wisconsin Sampling Site

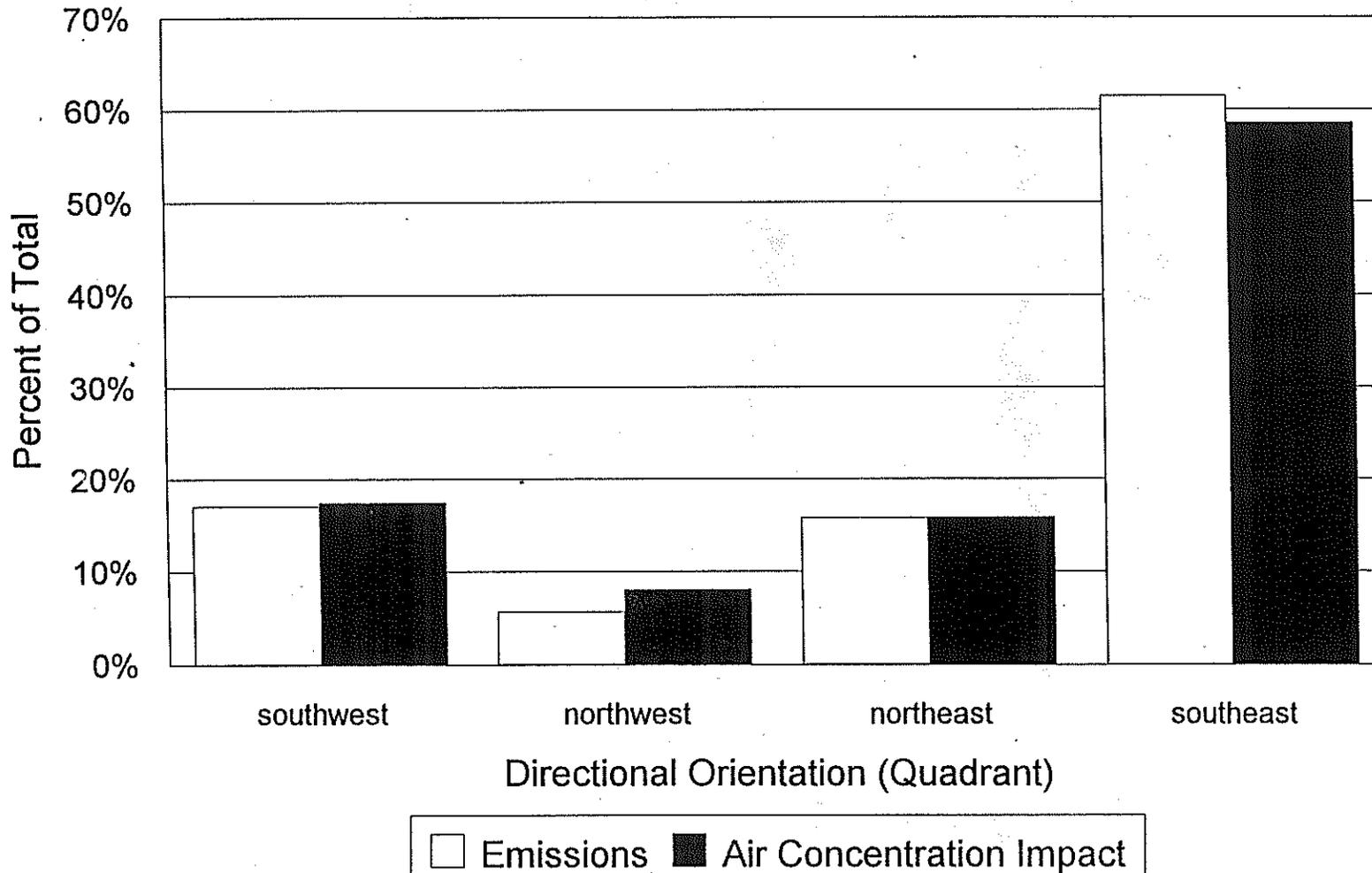
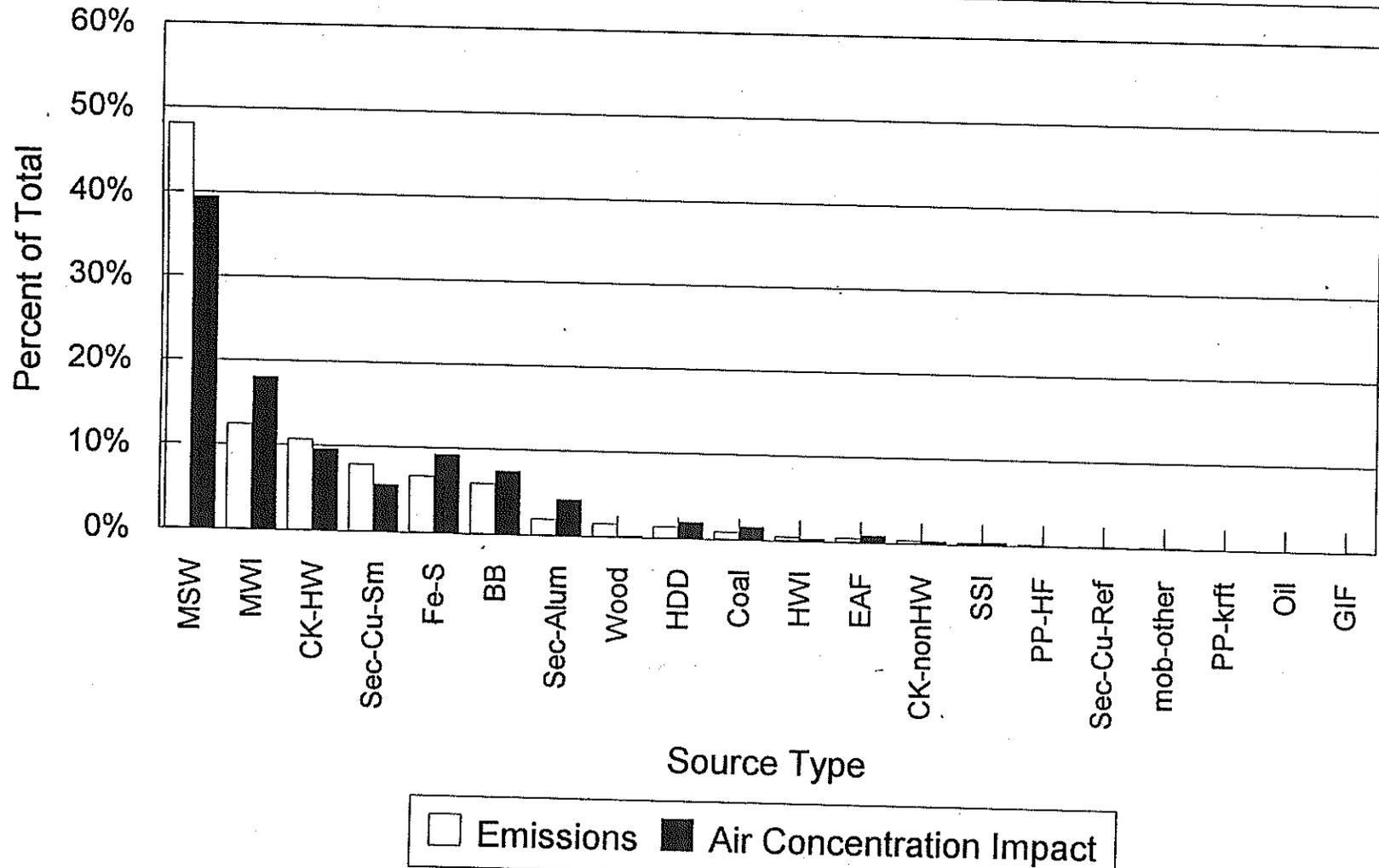
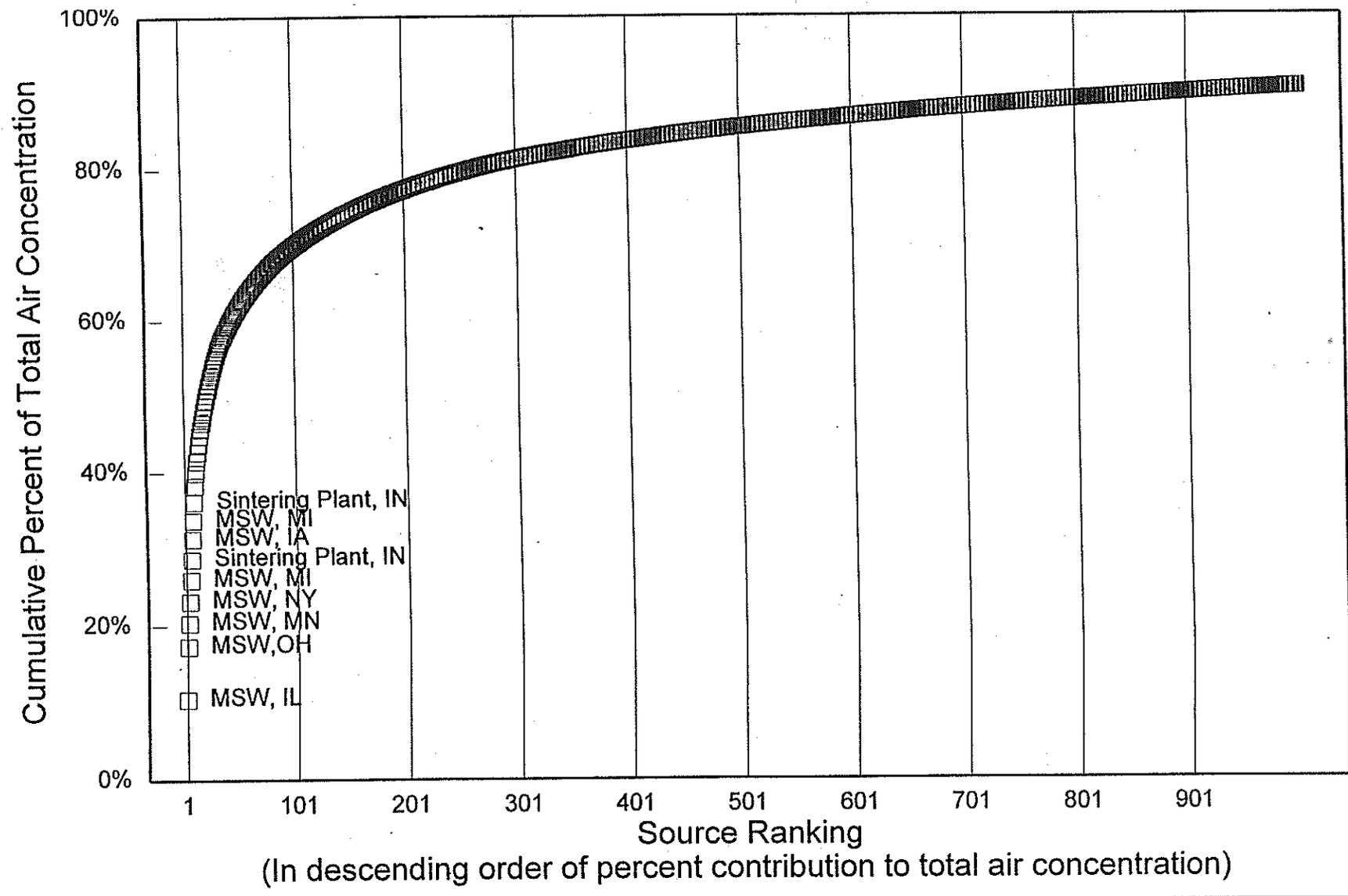


Figure 6C. Dioxin (TEQ) Emissions and Air Concentration Impact of Different Source Types for the Southeastern Wisconsin Sampling Site



**Figure 6-D Contribution of 1000 Highest-Ranking Sources to Air Concentration of Dioxin (TEQ) at Farm WI-A (Southeastern Wisconsin)**



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such as medical waste incinerators that are numerous but are individually low-emitters.

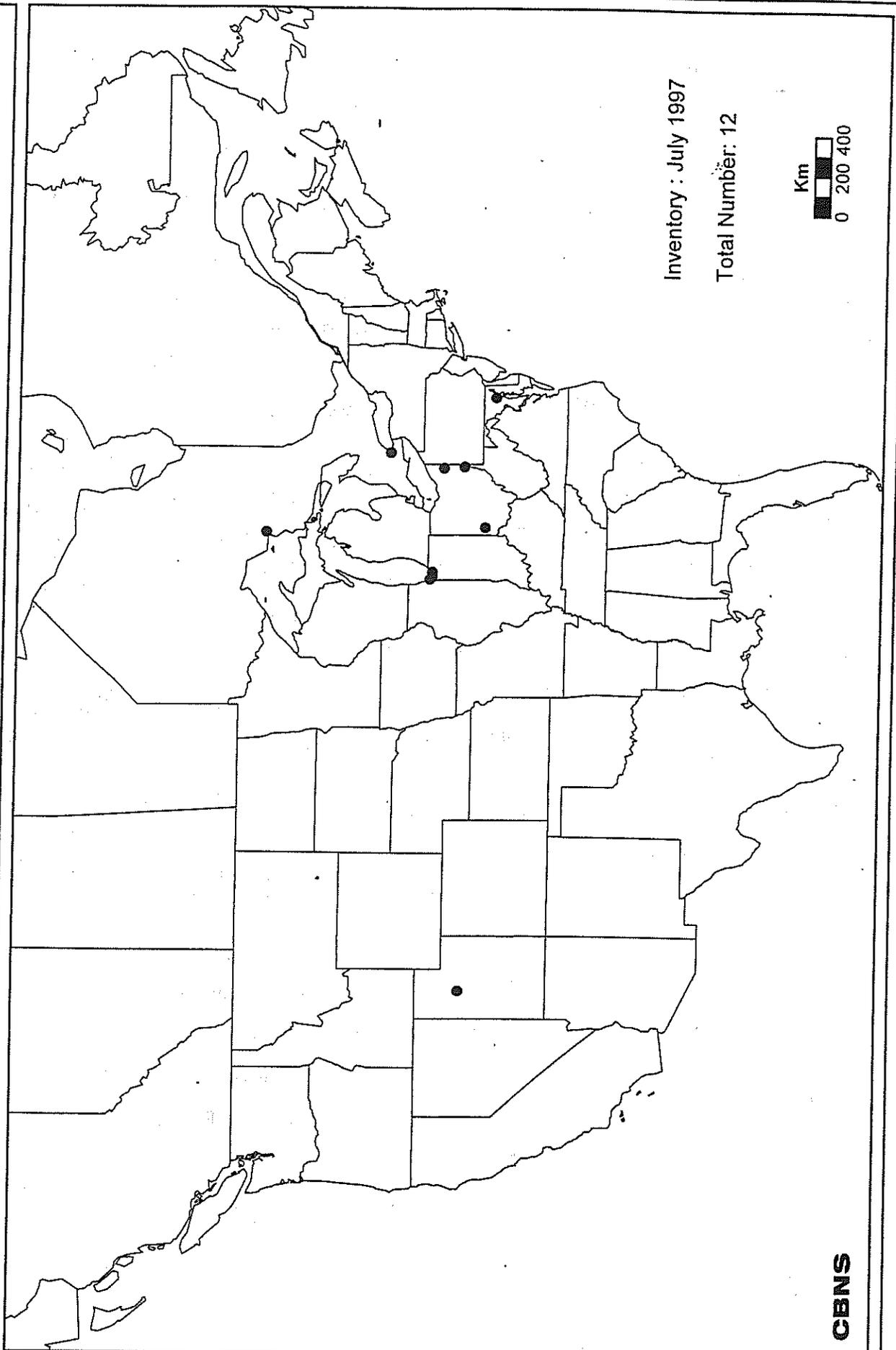
These indications are confirmed by the relevant source location maps. Figure 9 shows that WI-A is within 100 km of a very heavy concentration of medical waste incinerators in northeastern Illinois and consequently even more affected by these sources than WI-D in western Wisconsin. Finally, as noted in Fig. 10, there is a heavy concentration of iron sintering plants at the southern end of Lake Michigan, which accounts for their notable contribution to airborne dioxin at the Wisconsin receptors.

#### **D. Dioxin Transfer from Air to Crop**

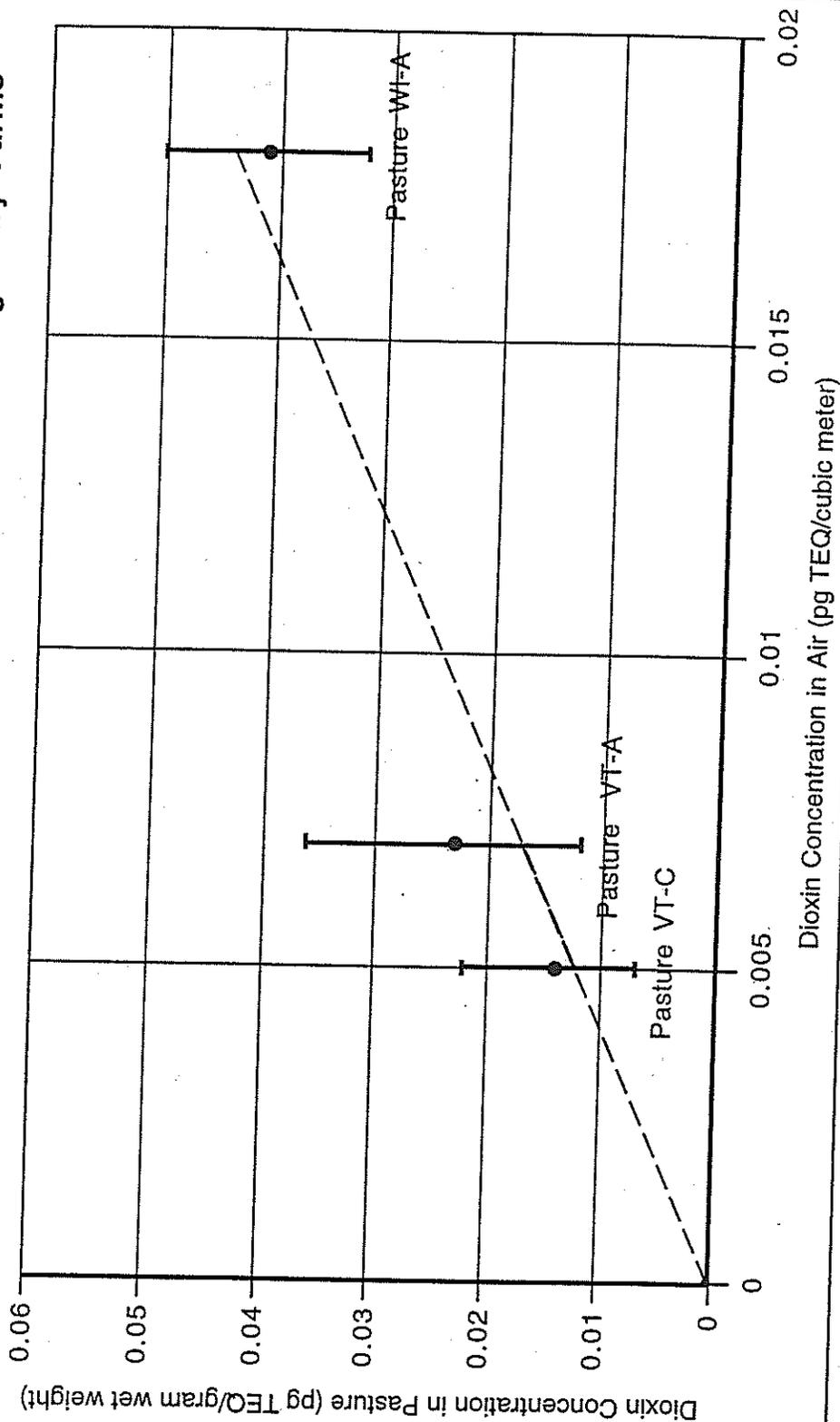
As noted earlier, samples of air were collected continuously at four test farms for dioxin analysis during the month-long test period. At VT-A VT-C and WI-A, which are managed grazing farms, the diet consisted chiefly of pasture (85-95%) and grain or other supplements. Only the pasture component of the diet represented vegetation that was exposed to the local, measured dioxin air concentration. Thus, samples taken from the pasture to represent growth that occurred during the test period were expected to contain the amount of dioxin that was taken up by the pasture vegetation from the air during the test period and would therefore bear some relation to the measured concentration of dioxin in the air during that period.

At VT-A and WI-A pasture samples were collected at the end of the test period from fenced, ungrazed sections of the pasture that had been closely clipped on the day the test began. At these farms samples were also collected weekly from the pasture areas that were actually being grazed by the cows during the test period. Since, at the start of the period, such samples contained vegetation grown in the preceding month, the final composite of the weekly samples represented a rolling average of the vegetation grown in July and August. The data from these samples were averaged with those from the ungrazed fenced areas; the single sample taken at farm VT-C was only from such grazed pastures. The resulting data are plotted in Figure 11. Although the data are necessarily limited to the three pasture farms, they indicate that the dioxin concentration in the pasture vegetation grown during the test period is generally

**Figure 10 Location of Iron Sintering Plants**



**Figure 11 Dioxin Concentrations in Air & Pasture at Grazing Dairy Farms**



Notes: Air and Pasture Samples were collected concurrently during the following periods: Aug 1-28, 1996 at VT-A, C; Aug 22-Sept 17, 1996 at WI-A.

: The Regression line was computed with intercept set at zero.

proportional to the concentration of airborne dioxin to which the pasture was exposed during that time.<sup>3</sup>

This result is in keeping with the evidence that dioxin enters pasture plants from the air, in the form of vapor, through stomata. Given the three-fold difference between the measured values of airborne dioxin at the Vermont farms and the farm in Southeastern Wisconsin, it is likely that dioxin levels in pasture and similar vegetation varies over a comparable range, regionally in the United States. This may contribute to regional differences in the dioxin concentration of milk as well.

#### **E. Dioxin Transfer from Diet to Milk**

##### Diet and Milk Samples

For this purpose, in keeping with the procedures described earlier, the total diets at the eight farms were sampled for dioxin analysis to represent the cows' dioxin intake during the month-long test period. At the five confinement farms, such samples were taken from the Total Mixed Ration. At the pasture farms, the dioxin concentration of the overall diets was computed from the weighted dioxin concentrations of samples of pasture and supplements. (The resulting values are therefore greater than they are in the pasture only samples referred to above.) In all cases, the original analytical data were in the form of dioxin concentration per gram wet weight of sample. However, what is of interest here is the dioxin flux — the amounts of dioxin ingested by the cows and excreted in the milk during the test period. The diet dioxin flux was computed by multiplying the concentration of dioxin in the diet by the herd average of the amount of the diet ingested, computed as pg TEQ of dioxin per cow per day. In the same way, the milk dioxin flux was computed, as pg TEQ per cow per day, from the measured concentrations of dioxin in the milk samples multiplied by the herd average milk production per cow per day. These data are shown in Table 4.

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<sup>3</sup>Alfalfa produced during the test period at WI-D, where the airborne dioxin concentration was 0.033 pg TEQ per cubic meter, contained a concentration of 0.174 pg TEQ per g (wet weight), a ratio about twice that of pasture vegetation.

Table 4: CARRYOVER OF DIOXIN FROM DIET TO MILK

FARM	TYPE (HERD SIZE)	DIET				MILK			DIOXIN FLUX RATIO: MILK/DIET (percent)
		COMPO- SITION	CONSUMPTION (lb/cow/day) <sup>5</sup>	DIOXIN CONCEN- TRATION (pg TEQ/g) <sup>5</sup>	DIOXIN FLUX (pg TEQ/ cow/day)	PRODUCTION (lb/cow/day)	DIOXIN CONCEN- TRATION (pg TEQ/kg)	DIOXIN FLUX (pg TEQ/ cow/day)	
VT-A	Grazing (140)	Pasture <sup>2</sup>	160	0.0270	1,960	46	9.450 <sup>1</sup>	197 <sup>1</sup>	10
VT-B	Confinement (275)	TMR	100	0.2082	9,444	60	9.333	254	3
VT-C	Grazing (95)	Pasture <sup>3</sup>	184	0.0354	2,955	84	21.96	837	28
VT-D	Confinement (28)	TMR	89	0.3462	13,976	68	26.64	822	6
WI-A	Grazing (86)	Pasture <sup>4</sup>	154	0.0406	2,836	49	4.815	107	4
WI-B	Confinement (53)	TMR	44	0.1019	2,034	53	43.48	1,045	51
WI-C	Confinement (175)	TMR	100	0.0920	4,173	89	11.68	472	11
WI-D	Confinement (240)	TMR	100*	0.0509	2,309	51	19.19 <sup>1</sup>	434 <sup>1</sup>	19
Average									17

Diet and milk sampled for dioxin content during Aug. 1996, at all farms.

<sup>1</sup> Average of two duplicate samples.

<sup>2</sup> Diet included 6.25% grain (0.01 pg TEQ/gram wet weight).

<sup>3</sup> Diet included 8.14% grain (0.17 pg TEQ/gram wet weight) and 7.02% haylage (0.08 pg TEQ/gram wet weight).

<sup>4</sup> Diet included 8.79% grain (0 pg TEQ/gram wet weight).

<sup>5</sup> Wet weight

\* Estimate

### Factors that Influence Diet to Milk Carryover

The results summarized in Table 4 show that the dioxin flux ratio, or carryover — i.e., the percentage of the dioxin ingested in the diet that is excreted in the milk — varies considerably among the different farms, from 3 to 51 pg TEQ/cow/day. Several factors are expected to influence the carryover value:

- The amount of dioxin ingested with the diet;
- The degree to which dioxin stored in the cow's body fat is mobilized during lactation and adds to the dietary dioxin that appears in the milk. Mobilization carries body fat, and with it the dioxin it contains, into the blood plasma; both fat and dioxin are then accessible to milk secretion.
- The "bioavailability" of the dioxin present in the diet — i.e., the degree to which dioxin present in the diet is absorbed by the cow and hence is capable of being transferred to milk. Bioavailability is particularly high — of the order of 80% — if dioxin is ingested in animal fat or vegetable oil. Little is known at present about differences in bioavailability that may be associated with various other diet components, such as pasture, corn or hay silage or supplements, except that the fat content of supplements such as cottonseed may tend to enhance bioavailability.

Each of these factors may be involved in the carryover rates exhibited at the different farms. There is more than a ten-fold variation in the dioxin concentrations in the diets of the eight test farms, although, as shown in Table 4, there is no apparent systematic relation between dioxin concentration in the diet and carryover rate. This suggests that variations in body fat mobilization and bioavailability, may largely influence the carryover rate. In particular, the extensive information about lactation in dairy cows indicates that the rate of fat mobilization probably varied considerably among the 1,092 cows, living under a diversity of management systems, that were involved in the study.

## Lactation

Cow lactation begins when the calf is born, a time called cow "freshening." Lactation will then typically last 305 days, during which time the cow is bred again and readied for the next year's lactation cycle. A typical confinement farmer is likely to aim for production levels of 100 pounds of milk per cow per day at the beginning of lactation, which drops by 10% per month over the lactation period to a low of 30-40 pounds at the end of the lactation cycle. A common average production is about 70 pounds of milk per cow per day.

At freshening cow body weight peaks, milk production is very high for several weeks, and the cow will mobilize body fat at a very high rate, adding significantly to the dioxin ingested with the diet. Appetite will initially be low and there will be some weight loss as lactation begins. As the appetite catches up with milk output, the cow then accrues body fat, and with it, stored dioxin. When the cow is bred (normally artificially inseminated), and the fetus develops, the cow will gain weight. This cycle is not uniform within a dairy herd, and not all cows will be successfully bred, so that their lactation cycles will not be the same as those that have been bred.

A number of interconnected factors influence body fat mobilization in the cow. The cow's genetic milk production potential sets the absolute limit of milk production. Nutrient intake and available body stores influence the *actual* milk production; the less a cow is fed, relative to her genetic potential, the greater will be the mobilization of body stores in the initial phase of lactation. Feeding high levels of high-energy grains and other supplements can increase milk output within genetic limits, and a high producer will mobilize fat if thereby "pushed" to produce more milk. Mastitis and metabolic stress related, for example, to calving or cow herd behavior patterns may increase fat mobilization. The management style on many low-input pasture farms may tend to create somewhat less "stress" on animals with respect to milk production, which is typically lower than on confinement farms, as seen at WI-A and VT-A. At VT-C, one of the pasture farms, the somewhat elevated levels of dioxin in milk may be due to several of the factors listed above. For example, most of the herd had recently freshened, milk

production per cow was very high, many of the cows had been bred and brought in from the Midwest, and farm management practices had been undergoing changes in recent weeks. The variable composition of the diet, especially among the five confinement farms, is also likely to influence the observed differences in dioxin carryover rate (see Table 5). For example, the dioxin levels in dietary fat and oil components are not only generally higher than they are in plant components, but these components also enhance dioxin bioavailability.

#### Analytical Uncertainties

In interpreting these results, it is also important to consider the effect of analytical uncertainties on the estimated carryover rates. This is especially true of the data on the dioxin concentrations in the diet samples, where the values are very low in comparison with the sample blanks. The error bars shown in Figure 12, which are almost entirely due to the diet data, represent the range of possible values of the diet-to-milk carryover rates. It is evident that except for the rates at VT-B and WI-A, all of the carryover rates may lie in the range of 20-25%, which is the value observed in several reported tests of this process on single cows. It is interesting, in this respect, that the average value of the individual carryover rates shown in Table 4 is 17% — close to the range of the values observed on single cows in relatively controlled (from a research perspective) circumstances.

#### Dioxin Concentration in Milk: Comparative Data

Finally, the overriding practical significance of these results is revealed by the dioxin content of the milk, which is usually expressed as pg TEQ per g lipid (milk fat). The data of Table 4, converted to this form, show that the values for the milk produced by the eight farms vary from a minimum of 0.12 pg TEQ per g lipid (WI-A) to a maximum of 1.10 pg TEQ per g lipid (WI-B); the overall average is 0.45 pg TEQ per g lipid.

It is useful to compare this result with the most comparable data, obtained in a survey of U.S. commercial milk supplies by U.S. EPA (Lorber *et al.*, paper presented at Dioxin '98, Stockholm, Sweden, August 1998). The survey was based on composite

Table 5: Analysis of the Components of the Total Mixed Ration Used at Farms VT-B, VT-D, WI-B, WI-C and WI-D

Component	VT-B		VT-D		WI-B		WI-C		WI-D	
	% of TMR	Dioxin Concentration pg TEQ/g	% of TMR	Dioxin Concentration pg TEQ/g	% of TMR	Dioxin Concentration pg TEQ/g	% of TMR	Dioxin Concentration pg TEQ/g	% of TMR	Dioxin Concentration pg TEQ/g
Corn silage	37	--	26	.17	--	--	--	--	20	0.14
Haylage	32	--	30	.08	--	--	43	--	58	0.10
High-moisture corn	15	--	--	--	11	0.02	25	--	--	--
Soybean meal	7	--	--	--	--	--	--	--	--	--
Corn meal	--	--	22	0.01	--	--	--	--	--	--
Custom grain mix	--	--	15	0.29	--	--	--	--	20	0.02
Cottonseed	--	--	7	0.28	--	--	3	--	--	--
Hay	2	--	--	--	61	0.17	5	--	--	--
Canola oil	6	--	--	--	--	--	--	--	--	--
Fat <sup>2</sup>	--	--	0.29	2.96	--	--	--	--	--	--
Protein mix <sup>2</sup>	--	--	--	--	27	0.07	6	--	--	--
Distillers corn	--	--	--	--	--	--	6	--	--	--
Wet brewers grain	--	--	--	--	--	--	13	--	--	--
Meat & bone meal <sup>2</sup>	--	--	--	--	--	--	--	--	2	0.26
Minerals, urea	1	--	--	--	--	--	--	--	--	--
Total TMR <sup>1</sup>	100	0.18	100	0.31	100	0.06	100	0.06	100	0.05

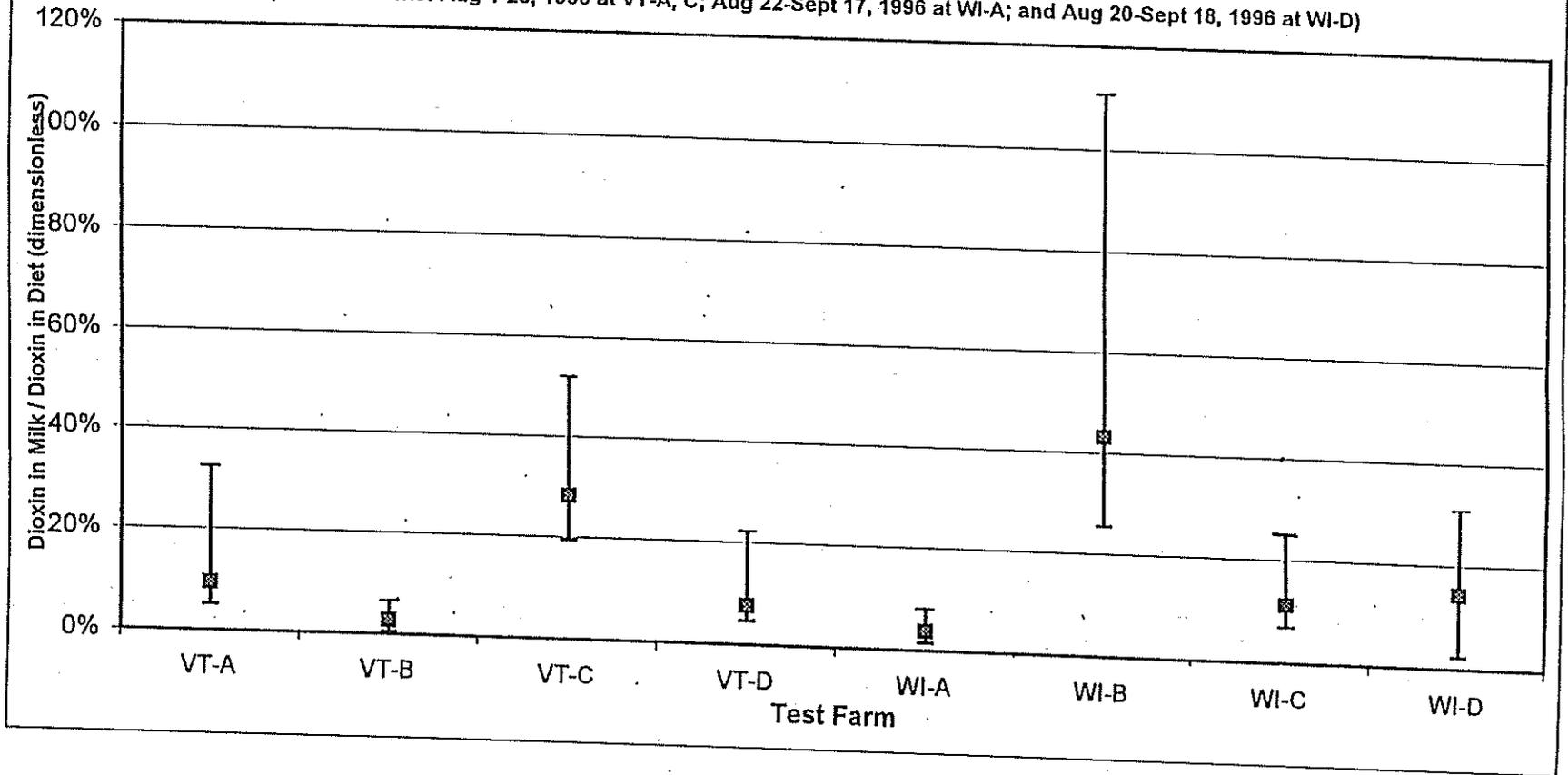
Diet sampled for dioxin content during Aug. 1-28, 1996, at VT farms, Aug. 22-Sept. 17, 1996, at WI-A, and Aug. 20-Sept. 18 at WI-D.

<sup>1</sup> Data from dioxin analysis of TMR fed during the test period.

<sup>2</sup> The current ban on ruminant-to-ruminant feeding had not yet come into effect at the time of this study.

**Figure 12 Ratio of PCDD/F in Milk to Diet for Study Farms**

(Measurements: Aug 1-28, 1996 at VT-A, C; Aug 22-Sept 17, 1996 at WI-A; and Aug 20-Sept 18, 1996 at WI-D)



samples of pasteurized milk collected from commercial dairies in U.S. metropolitan areas in April, July and October 1996 and January 1997. The national average dioxin concentration was 0.82 pg TEQ per g lipid. The lowest concentrations tended to occur in the July 1996 samples. For the purposes of comparison with our own results, the samples taken from a Boston, MA and St. Paul, MN dairy are most relevant. These values are 0.68 and 0.74 pg TEQ per g lipid respectively. In comparison, the average values for our Vermont and Wisconsin farms for August/September 1996 were 0.40 and 0.50 pg TEQ per g lipid respectively. It would appear that while our samples are somewhat lower in dioxin content than the regional composite samples reported by EPA, both sets of data suggest that the dioxin concentration of milk is somewhat higher in the Midwest than in the Northeast.

It is significant that the range of variation in the dioxin content of the composite regional samples tested by EPA (minimum 0.75 and maximum 0.94 pg TEQ per g lipid) is considerably smaller than the range of our eight samples from individual farms (minimum 0.12, and maximum 1.10 pg TEQ per g lipid). This suggests that farm-to-farm differences that result from differences in on-farm levels of airborne dioxin and/or differences in diet composition may be observed when milk from numerous farms are pooled for analysis.

#### **IV. CONCLUSIONS**

The results of this study make certain useful contributions to our understanding of the role played by dairy farms in carrying dioxin, emitted by thousands of often distant sources, from the air through feed crops into the milk distributed for human consumption. Most of the earlier studies have dealt with very limited numbers of cows over brief time periods, in a relatively limited range of circumstances. In contrast, we have studied eight farms in two widely separated regions, with herds comprising a total of 1,092 cows over a continuous, one-month test period. For this reason, it has been possible to compare, from farm to farm and region to region, the quantitative relation of the dioxin concentration in pasture to its concentration in the air to which the pastures

were exposed, and the relation of the dioxin in the diet to dioxin in the milk.

Recognizing that such estimates are subject to the significant uncertainties that are inherent in the considerable analytical constraints, certain useful generalizations can nevertheless be drawn from the results.

- The measured concentration of airborne dioxin in the Vermont and Wisconsin study regions varies over a three-fold range, indicative of the geographic variation in exposure to dioxin that is inherent in the non-uniform distribution of the most intense sources, such as municipal waste incinerators and in the regional, and even local, differences in weather patterns. **Thus, the level of overall exposure of dairy farms to environmental dioxin can be expected to vary from region to region in the nation as a whole.**
- The variation in the concentration of airborne dioxin among the test farms is reflected in a comparable range of dioxin concentrations in the pasture vegetation exposed to it. Moreover, as Fig. 11 shows, albeit based on only three observations, there is a reasonably linear proportionality between the concentrations of dioxin in the air and in the vegetation grown concurrently. This result probably reflects the similar ways in which the different plants that grow in pastures absorb dioxin from the air. A common dominant pathway seems to be involved: airborne dioxin enters the leaf via stomata, and tends to remain there. Moreover, this result confirms that the air is essentially the only environmental route of dioxin into the plant; no dioxin is absorbed from the soil, despite the amount that has accumulated in soil by deposition from the air over recent decades. This leads to an important practical outcome: **If the presence of airborne dioxin at a dairy farm is eliminated, within the next growing season the dioxin content of the feed crops grown on-farm will fall to zero. The cows bred at that farm and fed farm-grown crops would then produce dioxin-free milk, within the time required to replace**

**older milk cows by new ones — three to four years at most.**

- The dioxin air transport model is an effective means of characterizing the types and locations of the groups of sources that are chiefly responsible for the airborne dioxin that reaches the farms. The model is also effective in ranking the numerous sources with respect to their contributions to the concentration of airborne dioxin at the farms and hence to the resultant dioxin levels in the local dairy feed crops. **Despite the very large numbers of sources of various types in the total dioxin inventory, very few are responsible for the major part of the dioxin that reaches a given farm. This greatly facilitates designing preventive action.**
- The passage of dioxin from feed crops to milk is not well defined by this study, largely because of the inherent inaccuracies in the analytical results of dioxin measurements of crop samples. Overall, the results suggest that, in keeping with reports in the literature, about 20% of the dioxin ingested with the diet is excreted in the milk — as an average of widely varying individual analytical results at eight different farms. On theoretical grounds — for example, the influence of the cow's freshened state, or the varied bioavailability of dioxin occurring in different diet components — one might expect certain real differences in carryover rate among the eight test farms. However, any conclusion regarding the possible authenticity of such differences will require further study. **Such studies could identify farm practices that might minimize the degree to which dietary and body fat dioxin appear in the milk.**
- Our results show that the initial event, at a dairy farm, that results in dioxin contamination of the milk is the presence of dioxin in the air to which the local feed crops are exposed. Although the absence of airborne dioxin would solve this problem, this cannot, of course, be accomplished at the farm. Rather, preventive action can only be taken at the source. For that purpose, the sources that emit the dioxin that is transported through the

air to the farm must be identified and ranked. Our results show that the air transport model that we have developed to accomplish this task does so effectively. **Thus, the air transport model, together with the necessary dioxin inventory, is an essential tool for developing environmental policies that can successfully eliminate the threat of dioxin to dairy farms and to the people who depend on them for their livelihood and, more generally, for food.**

The indicated policy considerations are discussed below.

## V. POLICY CONSIDERATIONS

### A. Remedial Strategy: Control v. Prevention

Given that the present levels of dioxin in the general U.S. population are unacceptable, and that the ingestion of milk and dairy products accounts for a significant part of this exposure, remedial action is strongly indicated. There are two generic approaches to the remediation of environmental exposure to such toxic substances: The installation of pollution control systems to reduce the source emissions (although controls are never perfect and become exponentially more costly as they are improved), and, second, preventing the generation of the pollutant at the source, for example, by substituting a suitable dioxin-free process and thereby reducing emissions to zero. It is now widely understood that the prevention strategy is by far the most effective method of remediation. This is the position of major government agencies, such as the U.S.-Canada International Joint Commission and the U.S. Environmental Protection Agency. However, although EPA's "Pollution Prevention Policy Statement" was formally adopted in January 1989, the Agency still relies on control systems for most regulatory purposes. Nevertheless, prevention is to be strongly preferred as the basis of remedial policy.

Fundamental to this strategy is the recognition that the sources that emit pollutants into the environment are themselves production processes. They are governed by a fundamental operational condition: that the same process that produces

the source's goods also produces the environmental "bads," such as dioxin. Thus, incineration of municipal waste, which produces the desired good of trash disposal, is the same combustion process that produces dioxin as well. (Chlorine in certain trash components combines with unburned organic substances to form dioxin in the cooler parts of the incinerator system.) The basic preventive measure is to break the link between the production of goods and pollutants — transforming the production process so that it achieves the goods without at the same time generating dioxin. Thus, recycling municipal waste instead of incinerating it produces the sought-for good — trash disposal — without producing dioxin.

The choice between control or prevention as the strategy of dioxin remediation has important practical consequences. As a regulatory measure, the control strategy has the advantage of uniformity; once an emission limit that the control system can achieve has been established, it can be applied universally to all sources of a given type, such as medical waste incinerators or hazardous waste incinerators. However, if their efficiency is improved in order to reduce the present, unacceptable level of exposure to dioxin, control systems become progressively more costly. Thus, if environmental levels are improved, further progress becomes exponentially more difficult to achieve and, in practice, zero emissions are unattainable.

#### **B. Implementing the Prevention Strategy**

In the prevention strategy, dioxin exposure is reduced by successively replacing each source by a dioxin-free alternative — a process that makes the next step easier, rather than more difficult. Moreover, since the dioxin sources are very numerous and vary greatly in their individual contribution to overall emissions, a great deal can be gained in environmental quality by concentrating the remedial effort on the worst offenders and, one by one, reducing their dioxin emissions to zero.

The results of this study provide a basis for applying this approach to the practical task of sharply reducing dioxin exposure at receptors such as dairy farms. As noted earlier, it is evident from our analysis of the source/receptor relationships that most of the exposure is due to an extremely small fraction — of the order of one percent

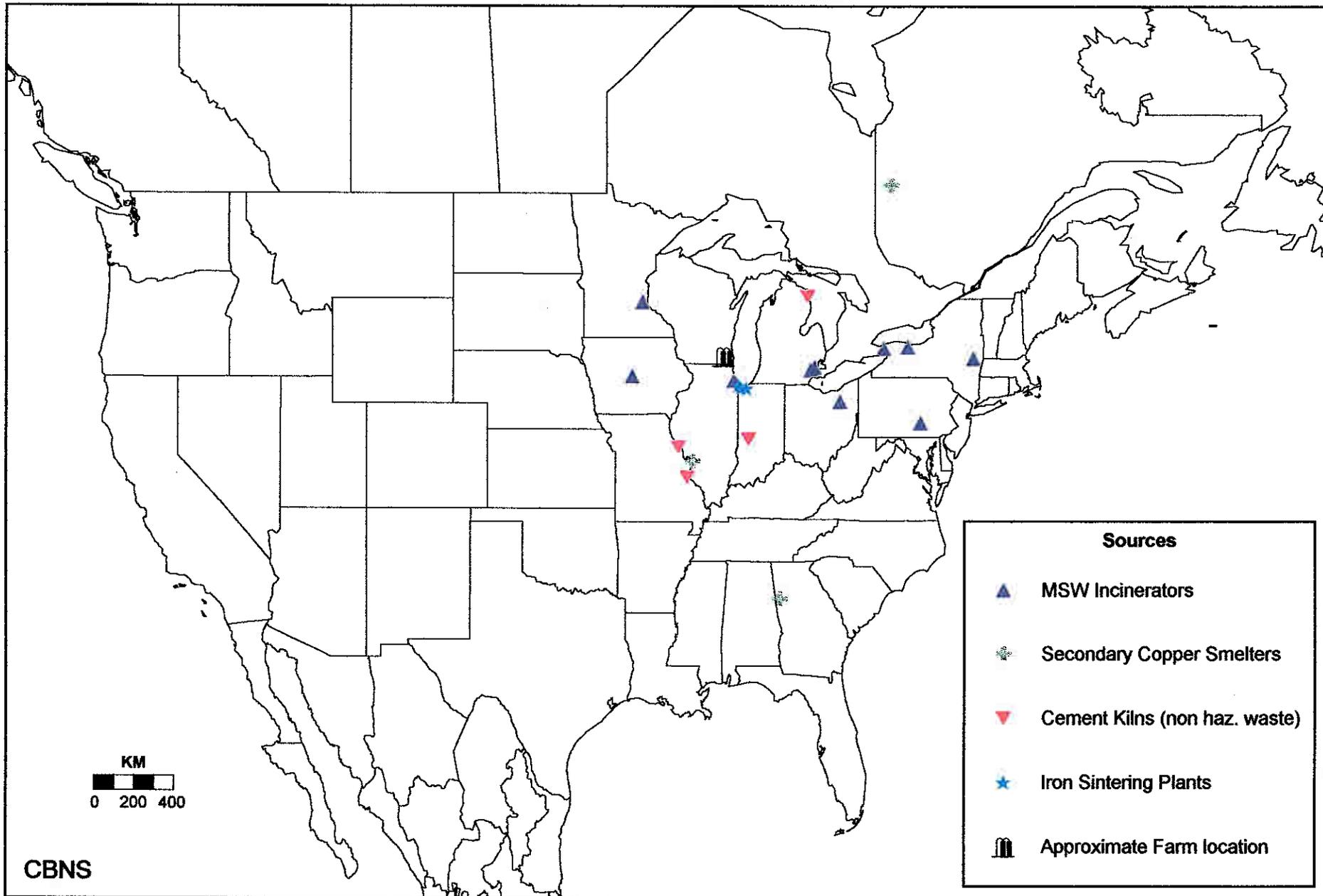
or less — of the 24,644 sources in the dioxin inventory and is limited to relatively few of the 20 source types. In order to arrive at a practical remedial policy, it is useful to identify, for each of the receptors, those individual sources which, if replaced by equivalent dioxin-free processes, would have a major effect on the farms' dioxin exposure. Initially, it is also instructive to evaluate the relationship between the size of the sought-for remedial effect and the number of sources that need to be replaced to achieve it.

Relation Between Remedial Effect and the Number of Sources  
Replaced by Dioxin-Free Alternatives

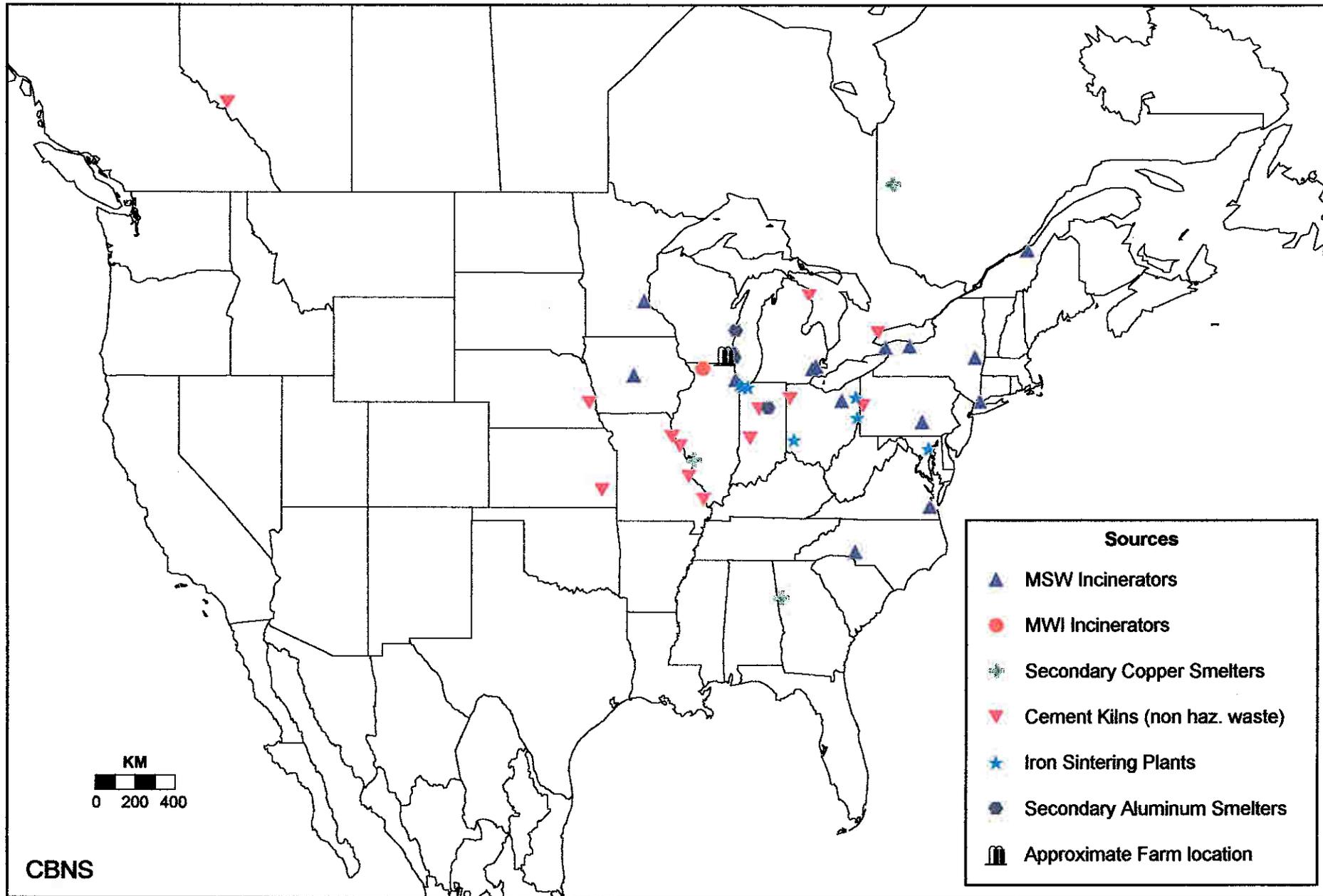
In Figures 13.A, B and C we map the locations of those highest-ranked sources, which — if replaced by dioxin-free alternatives — would eliminate, respectively, 50%, 60% and 75% of the total airborne dioxin concentrations at farm WI-A. Several important conclusions emerge:

- *50% reduction:* This can be accomplished by taking preventive measures at only 21 sources. Only four source types are involved — 10 municipal waste incinerators, three secondary copper smelters, four cement kilns, and four iron sintering plants. None of these sources are in Wisconsin; about half are in states south-southeast of Wisconsin: Illinois, Indiana and Ohio; one secondary copper smelter is in Georgia, and another in Quebec.
- *60% reduction:* This goal requires preventive transformation of 43 sources of six different types, dominated by 14 municipal waste incinerators. Now two Wisconsin sources (secondary aluminum smelters) are involved, and the new sources (i.e., those absent in the 50% case) tend to be clustered in the Illinois, Indiana and Ohio.
- *75% reduction:* At this level, 161 separate sources contribute to dioxin at the receptor; they are included in 11 different source types. Most of the newly added sources are clustered in Illinois, Indiana, Michigan, Ohio, and Wisconsin itself. Among them are several that are characterized by

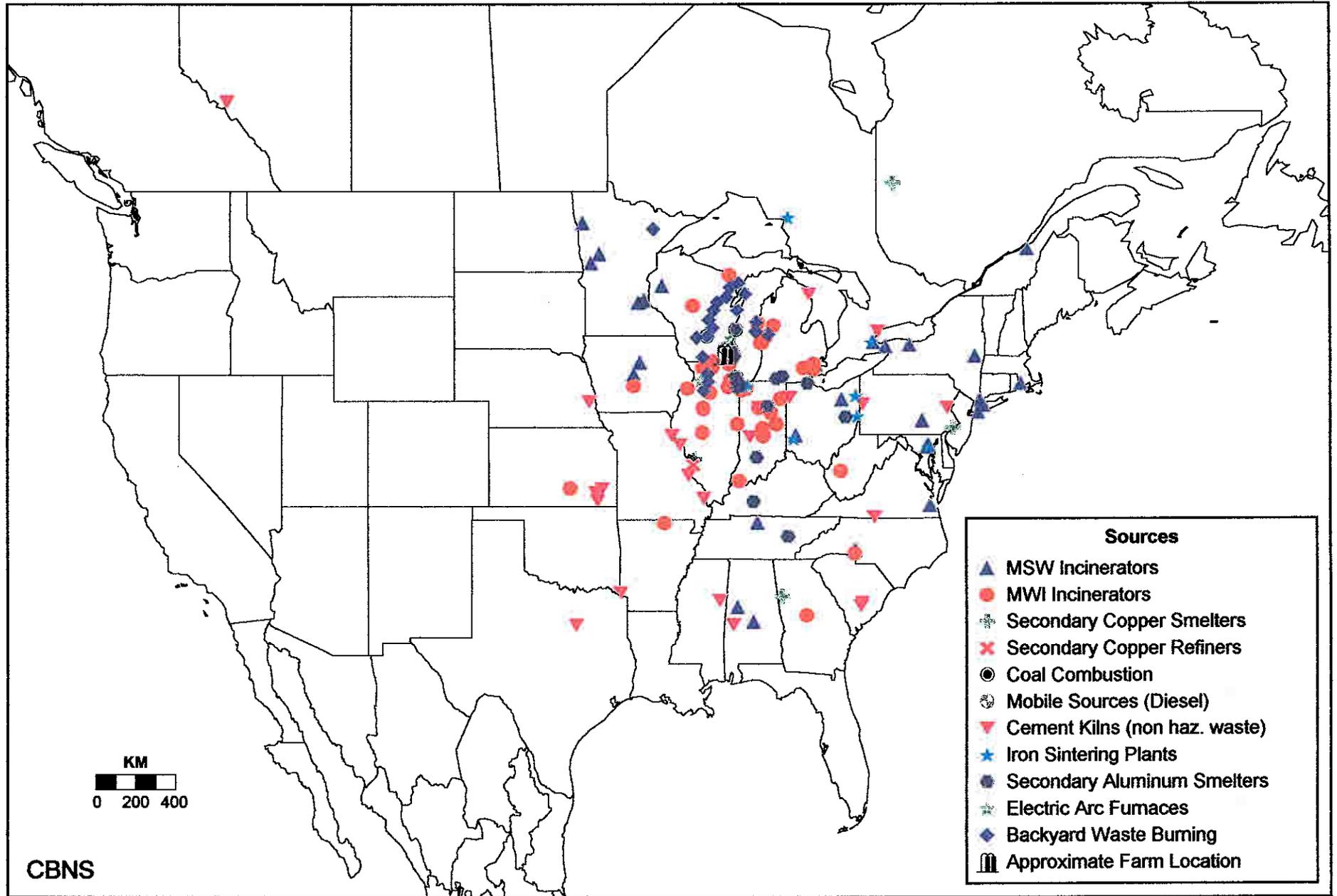
**Figure 13-A**  
**Highest-Ranked Sources (21) Contributing to 50% of Total Concentration**  
**of Airborne Dioxin at Farm WI-A (Southeast Wisconsin)**



**Figure 13-B**  
**Highest-Ranked Sources (43) Contributing to 60% of Total Concentration**  
**of Airborne Dioxin at Farm WI-A (Southeast Wisconsin)**



**Figure 13-C**  
**Highest-Ranked Sources (161) Contributing to 75% of Total Concentration**  
**of Airborne Dioxin at Farm WI-A (Southeast Wisconsin)**



relatively small emissions per source: medical waste incinerators, backyard waste burners, coal combustion, and mobile sources (diesel). Because these sources are low-emitters of dioxin, they have a significant impact on airborne concentration at WI-A only if they are relatively close to it. Thus, most of the medical waste incinerators are in states adjacent to Wisconsin, or within it, and most of the contributing backyard burners are in counties within Wisconsin or just south of it in Illinois.

This exercise suggests several useful guides to the design of practical remedial policies applicable to farm WI-A.

- Preventive measures capable of eliminating more than half of the airborne dioxin at the receptor farm — a good start toward elimination — are most readily accomplished if directed toward the six most heavily-emitting source types.
- Nearly all of these major sources are outside Wisconsin, chiefly in nearby Midwestern states, within about 750 km of the receptor farm. Policies must therefore be regional in scope, encompassing the main stakeholders: in Wisconsin, the farmers (and the local dairy industry); and in the nearby Midwestern states both the source operators and the consumers of Wisconsin milk. Fewer, large sources, located outside the region at distances up to 1200 km, are also part of these major contributors to the problem.
- In order to progress toward the elimination of three-fourths of the airborne dioxin at the receptor, remedial policies must address low-emitting sources relatively close to farm WI-A, many of them within Wisconsin itself. These sources — for example, medical waste incinerators and backyard waste burners — are relatively numerous and, in practice, must be dealt with as a group, for example by regulatory legislation.
- As noted earlier (see Figures 3-6.D) to address preventive policy toward the elimination of 90% or more of the airborne dioxin at the receptor farms

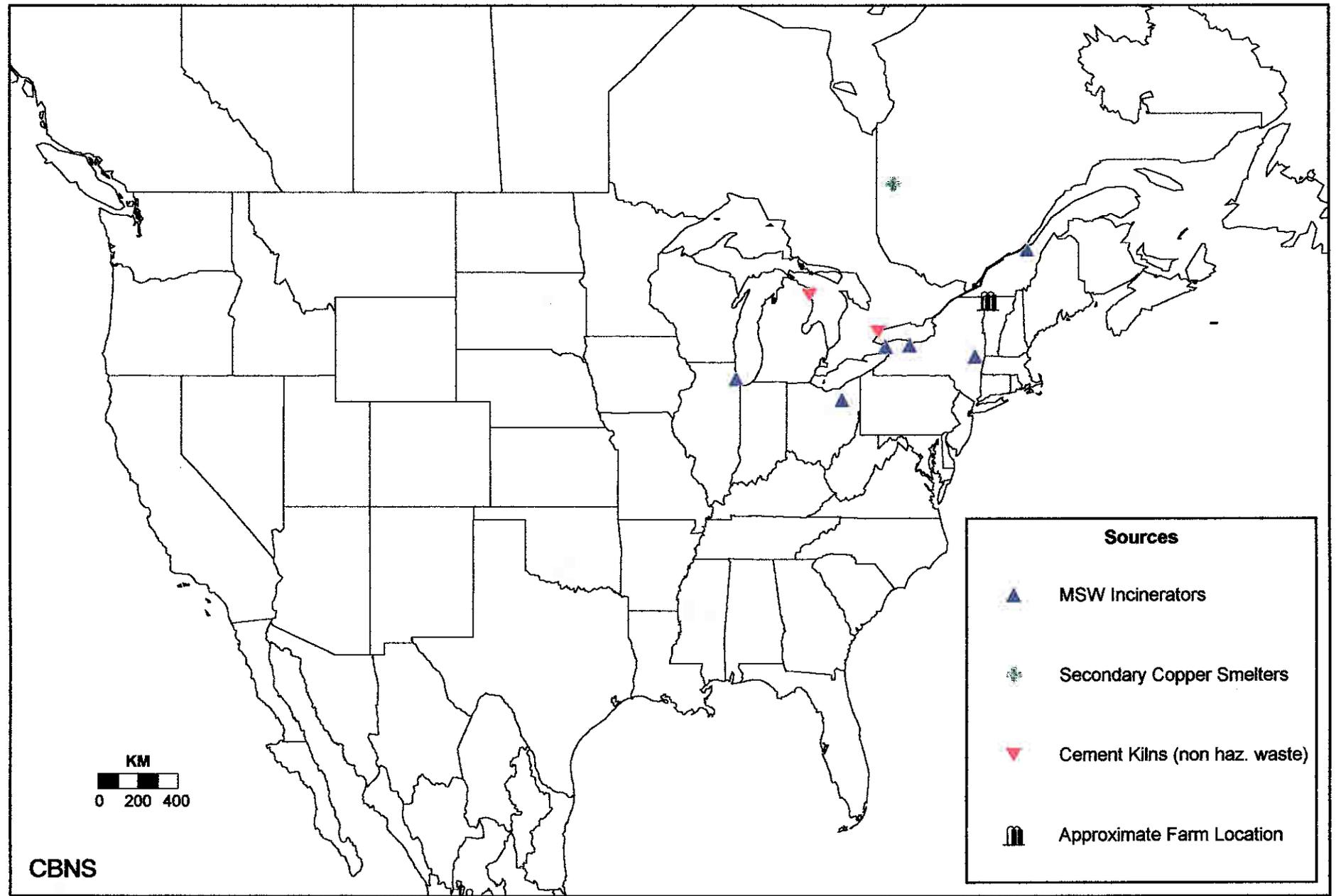
requires action at of the order of 1000 sources. However, most of these will be rather low-emitters and relatively close to the receptor.

### Regional Differences

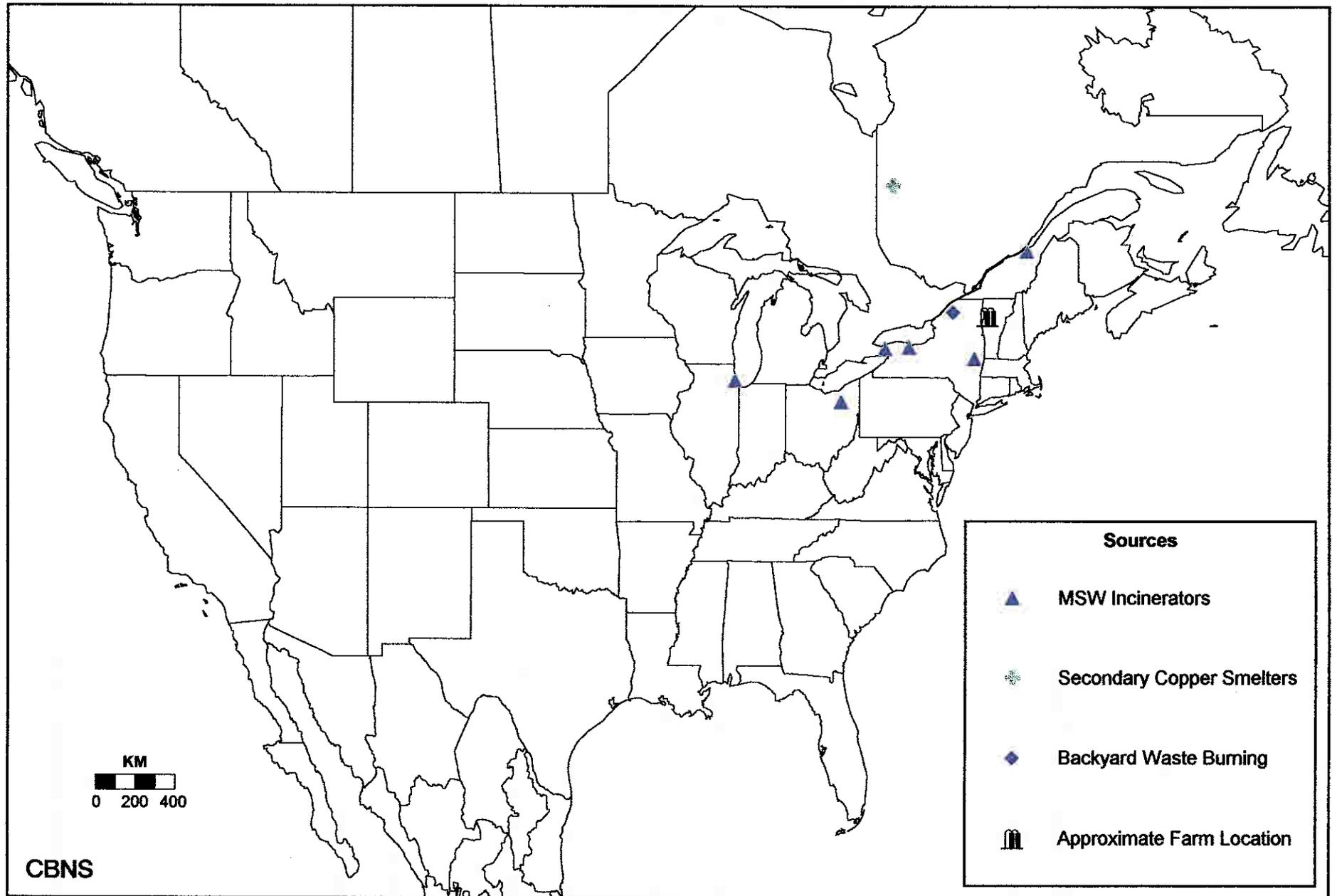
A major aspect of remedial policy relates to the degree of uniformity among different regions of the country with respect to the source/receptor relationships that govern their level of exposure to dioxin. For this purpose, in Figures 14.A, B, C and D we examine the geographic patterns of highest-ranked sources that — if replaced with dioxin-free alternatives — would eliminate 60% of the airborne dioxin at each of four farms, two in Vermont and two in Wisconsin. The results are informative.

- *Vermont farms:* Figure 14.A maps the relevant sources that contribute 60% of the airborne dioxin concentration at farm VT-A. Only nine sources are involved: six municipal waste incinerators, half of them in New York; two cement kilns (not burning hazardous waste), and the secondary copper smelter in Quebec, Canada. At farm VT-C, the contributing sources are the same as those at farm VT-A except that the cement kilns are excluded and backyard trash burners in a single New York county are included. (See Figure 14.B) This degree of uniformity between the two sites is to be expected, for they are only 56 km apart.
- *Wisconsin farms:* These data are shown in Figures 14.C and 14.D. They reflect source/receptor relations that are distinctly different from those that characterize the Vermont farms. Whereas only three source types are involved in Vermont, six are involved in Wisconsin. In Wisconsin the total number of sources that are responsible for 60% of the airborne dioxin concentration at the receptors is much higher than in Vermont: 43 and 35, respectively, at WI-A and WI-D, as compared to eight and nine at VT-A and VT-C. The additional sources that appear in the Wisconsin maps are largely due to industrial facilities (cement kilns, iron sintering plants, secondary aluminum smelters, and secondary copper smelters), reflecting the concentration of such types of sources in the Midwest. In contrast

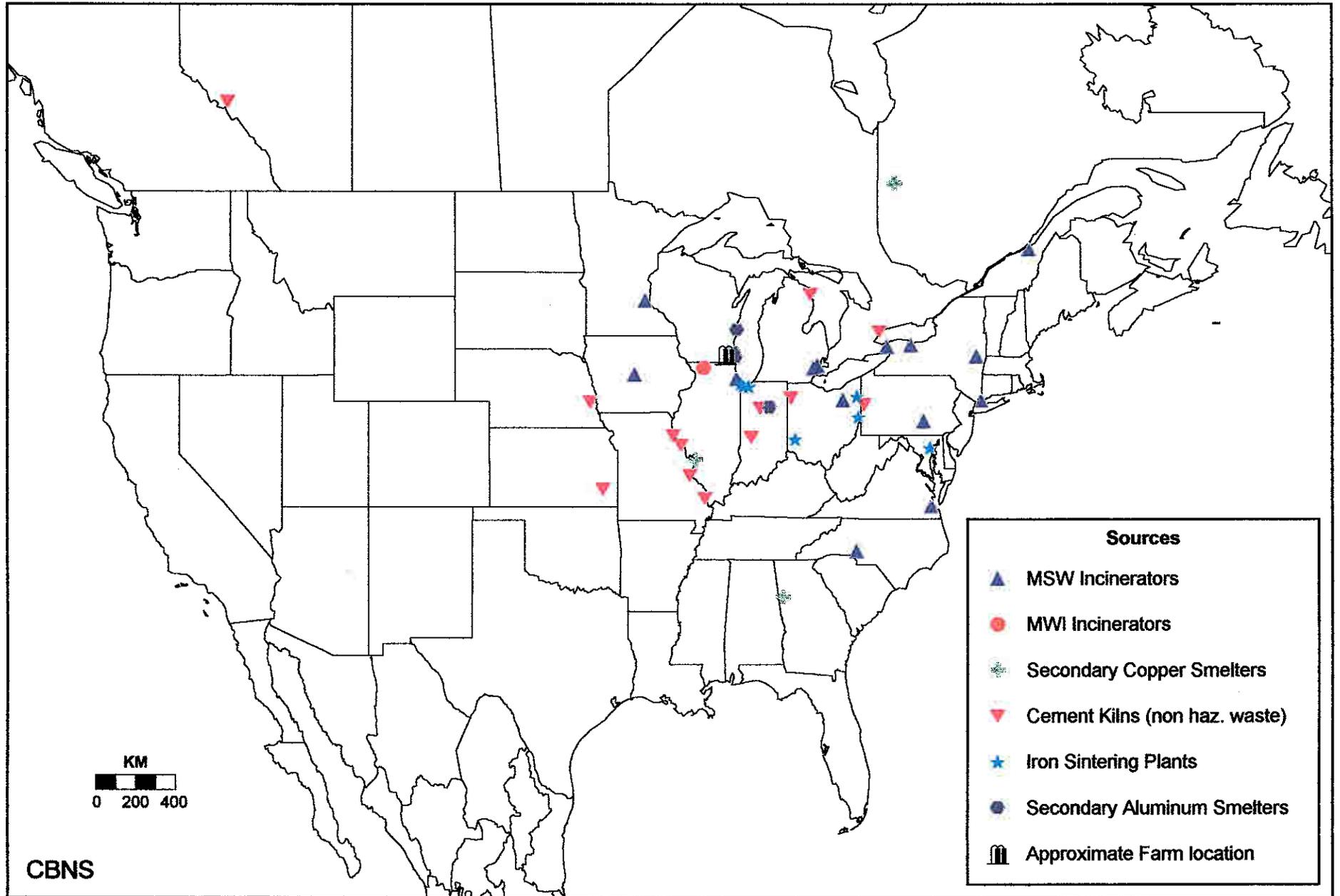
**Figure 14-A**  
**Highest-Ranked Sources (9) Contributing to 60% of Total Concentration**  
**of Airborne Dioxin at Farm VT-A (Northern Vermont)**



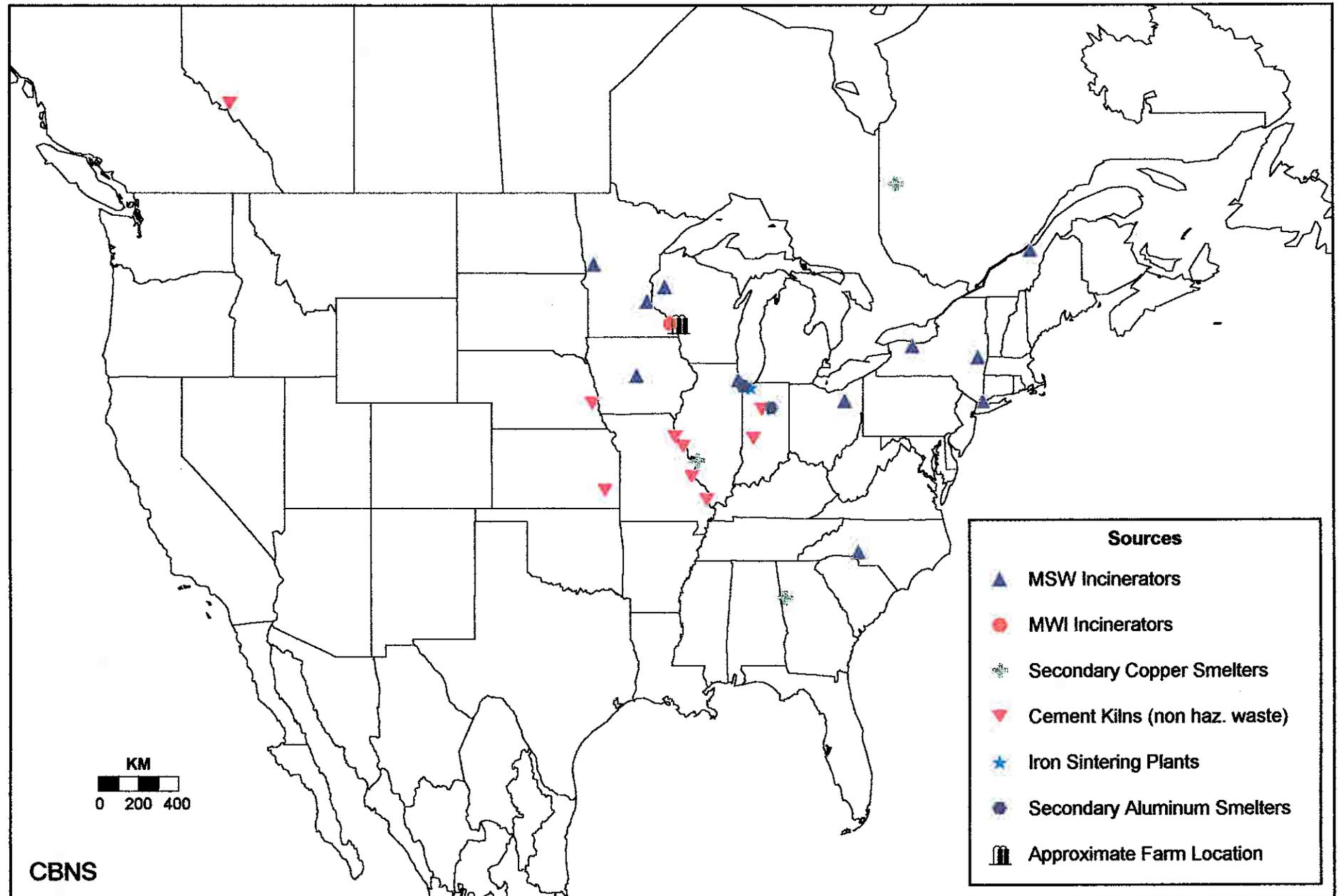
**Figure 14-B**  
**Highest-Ranked Sources (8) Contributing to 60% of Total Concentration**  
**of Airborne Dioxin at Farm VT-C (Central Vermont)**



**Figure 14-C**  
**Highest-Ranked Sources (43) Contributing to 60% of Total Concentration**  
**of Airborne Dioxin at Farm WI-A (Southeast Wisconsin)**



**Figure 14-D**  
**Highest-Ranked Sources (35) Contributing to 60% of Total Concentration**  
**of Airborne Dioxin at Farm WI-D (Western Wisconsin)**



with the Vermont farms, the two Wisconsin receptors differ significantly in their source-receptor patterns, with farm WI-A (southeastern Wisconsin) more affected by sources in Michigan, Ohio, and Pennsylvania than farm WI-D (western Wisconsin). Significantly, while the Vermont farms are only 56 km apart, the Wisconsin receptors are 272 km apart.

In sum, the foregoing data serve to identify, for each receptor, the distinctive array of sources which — if replaced by dioxin-free alternatives — would have a major remedial effect on dioxin exposure. By narrowing down the major sources to an exceedingly small fraction of the total number of them, these data provide a practical physical basis for preventive remedial policy. By specifying the geographic locations of the major sources, the data define the regulatory and political features of such policy.

The data indicate that preventive remedial policy should be regional in scope. At least with respect to receptors in Vermont and Wisconsin, areas that are some 1500 km apart, the source/receptor patterns are qualitatively different. Differences in source/receptor patterns between the two farms within each of these regions tend to be quantitative rather than qualitative; in Vermont, where the farms are considerably closer to each other than in Wisconsin, such differences are very small.

#### What Can Dairy Farmers Do?

It is the purpose of environmental policy to facilitate action, in this case to reduce or preferably eliminate the dioxin content of milk. Given the variety and geographic distribution of the sources that emit airborne dioxin, preventive action will take time. Is there any action that dairy farmers can take in the meantime that might reduce the level of dioxin in their milk? As indicated earlier, certain farm practices may affect the dioxin content of milk, in particular the dioxin content and "bioavailability" of diet components and lactation practices. There is some evidence in our results to suggest that such differences, which are likely to be largest between intensive grazing and confinement farms, do occur. As Table 4 indicates, the intensive grazing farms tend to have lower dioxin concentrations in their milk: a three-farm average of 0.034 pg TEQ per g of milk as compared with a five-farm average of 0.160 pg TEQ per g milk in confinement farms.

Another indication is given by the results of the EPA survey of regional composite milk samples. These were taken in five regions of the country at four different consecutive seasons in 1996 and 1997. The average seasonal values for the five regions were: April '96, 0.74 pg TEQ per g lipid; July '96, 0.64; October '96, 1.00; and January '97, 0.78. Feeding practices in July, when the dioxin concentration of milk is lowest, are likely to favor freshly grown vegetation — with reduced fat content and hence lower in dioxin content and bioavailability. It is possible, depending on further analyses, that farm practices that emphasize increased reliance on grazing and low fat diets (especially animal fat) may tend to reduce dioxin levels in milk, as a partial remedy, apart from action taken at the sources.

#### The Economic Feasibility of Dioxin-Free Alternatives to Major Sources

As already noted, there are technical changes that can be taken at the source to reduce its dioxin emissions to zero. However, since such preventive action requires that the production process, which is the generator of economic value, be fundamentally transformed, the economic feasibility of doing so must be considered as well. Such data are available for some of the dioxin sources that are major contributors to the airborne dioxin that reaches the dairy farms that we have studied: municipal waste incinerators, medical waste incinerators, and iron sintering plants. An earlier CBNS study of the sources that contribute to the deposition of airborne dioxin in the Great Lakes evaluated the economic feasibility of converting these activities to dioxin-free processes in the Great Lakes region (the eight U.S. states and the province of Ontario that are adjacent to the Lakes).<sup>4</sup> These regional results are applicable nationally as well. The relevant results are summarized below.

- *Municipal Waste Incinerators:* Municipal waste can be disposed of without at the same time producing dioxin by means of an intensive form of recycling. Intensive recycling collects all forms of residential waste that

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<sup>4</sup>Commoner, B., Cohen, M., *et al.*, 1996: Zeroing Out Dioxin in the Great Lakes: Within Our Reach. CBNS report to The Joyce Foundation, June.

are recyclable or compostable, generally of the order of 90% of the total waste stream. The net economic effect of substituting intensive recycling for the residential waste incinerated in the Great Lakes region in 1995 (11.7 million tons) would increase disposal costs by \$601 million per year (for additional collection and public education and retiring the outstanding debt on the incinerators); however, this increase would be outweighed by the revenue of \$1,137 million annually from marketing the recycled materials and avoiding incinerator tipping fees. The net result would reduce municipal waste disposal costs by \$536 million annually.

- *Medical Waste Incinerators:* Autoclaving and subsequent landfill disposal is a dioxin-free alternative to incinerating medical waste. For a typical hospital incinerator, this would add about \$0.60 per patient per day to the average cost of hospital operations, or about one-tenth of one percent of the hospitals' total operating costs (about \$800 per patient per day). Another alternative is to eliminate the extensive use of disposable chlorinated plastics in hospitals, a change that is being carried out in some hospitals with minimal economic effects.
- *Iron Sintering Plants:* The steel industry uses such plants in order to agglomerate finely powdered iron ore so that it is suitable for use in a blast furnace. This is done by mixing the ore with powdered coal and coke and heating the mixture in a specially designed furnace. However, the process is also used to recover iron from steel plant residues, such as dust and scale. Since these residues are usually contaminated by chlorine-bearing materials such as cutting oils or hydraulic fluids, sintering furnaces are known to emit significant levels of dioxin. Landfilling the chlorine-contaminated residues is a dioxin-free alternative. This would result in an economic loss from the failure to recover iron from the plant residues that would amount to an additional 1% in the price of steel. Another alternative is to eliminate the use of chlorinated materials in steel

operations. Certain plants have claimed to achieve this change without any ensuing economic difficulties.

Such information about the economic feasibility of dioxin-free alternatives is not available for the other sources that may play a major role in contributing to the airborne dioxin at Vermont and Wisconsin dairy farms: backyard burning and secondary copper smelters. In general, it would appear to be economically feasible to replace backyard burning with collection systems and intensive recycling of the collected material. As noted earlier, backyard burning is a significant contributor to airborne dioxin at the Wisconsin farms, but not the Vermont farms. It is significant that Vermont state regulations ban backyard burning.

To our knowledge, there has been no systematic analysis of the origin of chlorinated substances in the wide variety of waste materials used by secondary copper smelters, such as insulated copper wires, electronic circuit boards, telephone equipment and automotive parts. If one accepts the economic necessity of recovering scrap copper, then the process could be made dioxin-free only by substituting non-chlorinated materials in the manufacturing of wire insulation and electronic and similar equipment. Such substitutions have been made in other products, for example disposable medical equipment, and should be feasible in this case as well. However, unlike hospitals, the secondary copper industry has not as yet addressed this problem, and we lack the information needed to specify how it could feasibly be done.

In sum, apart from secondary copper smelters, it appears that economically feasible, dioxin-free alternatives exist for all of the other major contributors to the airborne dioxin impacting the test farms in Vermont and Wisconsin.

#### Putting Pollution Prevention Policy Into Practice

The remaining policy question is how to effectuate such pollution prevention measures. They necessarily involve the interests of dairy farmers, the dairy product industry, the operators of the dioxin sources, regulatory agencies, and the general public who consume milk and dairy products. Since these separate interests diverge widely and conflicts are inevitable, there is reason to be skeptical about the possibility

of taking action to replace the major sources with dioxin-free alternatives.

On the other hand, in two of these dominant source types — municipal waste incinerators and medical waste incinerators — significant progress has already been made. Inventory surveys, including our own, show that in the last few years a significant number of hospital medical waste incinerators have been replaced by autoclaves or by shipping the waste to regional facilities, many of which now use autoclaves as well. In addition, there have been innovations in waste reduction, recycling and disposal at major hospitals. In Vermont, for example, the work of the Fletcher Allen Hospital, stimulated in part by this research, has already put into place new policies regards dioxin and mercury waste that are a marked improvement over earlier practices. They have also led the way in providing educational materials to medical practitioners and others in both the United States and Canada. Such policies adopted by the operators of individual medical waste incinerators have contributed significantly to the marked reduction in emissions from this group of dioxin sources in recent years. In the same period, there has been a notable decline in new proposals for municipal waste incinerators, and a number of proposed facilities have been abandoned, often in favor of expanded recycling. In New York City, where official municipal waste management plans once called for the construction of eight massive incinerators, today all such plans have been abandoned. Where New York once had dozens of medical waste incinerators, none now exist.

It is informative to examine the factors that have influenced such positive changes. One reason is that municipal and medical waste incinerators are usually under public governance, either operated directly by the municipality, or if privately operated, funded in part and often wholly by public funds. Hence, these facilities are most directly accessible to public opinion through elected officials. Another reason is economic: it is now generally agreed that both of these types of incinerators are more costly than the dioxin-free alternatives. A third factor has been the efforts of community and environmental organizations, who are not only concerned about the environmental hazards, but informed about how they arise and can be remedied. These organizations

are able to meet the incinerator professionals on their own ground and win the day with scientifically sound policy alternatives.

These factors are applicable to the dairy problem as well, albeit with certain important modifications. First, the issue here is not the fate of a particular dioxin-generating facility — a trash-burning incinerator, for example — proposed by the same community that would likely suffer from its environmental hazards. Rather, we are dealing with a number of sources of different types, far removed from the farming communities they affect. The air transport data tell us that an incinerator in Albany, New York, will surely expose the people of that city to unwanted dioxin, but indirectly; dioxin will return from its airborne voyage to the farms of Vermont in the form of the milk sold in Albany food stores. Clearly, both the people in Albany and the farmers in Vermont have a common interest in calling for action toward the goal of dioxin-free milk. The same is true of the industry that collects, bottles and distributes the milk or converts it into cheese and ice cream. These relations create a community of interest in preventive action that has diverse capabilities for accomplishing it. In states like Vermont and Wisconsin, farmers' organizations have considerable influence with public officials and the media; the dairy industry has similar influence through the business community; and consumers' intense interest in healthy food is also a persuasive force — witness the surprising rapid development of the organic food market.

We have met with representatives of this diverse group of stakeholders, especially in Vermont, to discuss the purpose of this study and its policy implications (see Appendix C). It is apparent from these discussions that there is a wide interest in learning more about the problem and in devising ways to reduce and eventually eliminate dioxin from milk and dairy products. There is an opportunity for a coalition that brings all these forces to bear on this common goal.

Such a regional alliance for dioxin-free milk would respond to the nature of the source/receptor relationship revealed by our study. The relationship is inherently regional, not only because of the diverse geographic distribution of sources and weather patterns, but also because of the regional nature of the dairy industry,

especially with respect to fresh milk. This suggests that remedial efforts can most readily be organized on a regional basis.

Yet it is equally true that dioxin contamination, not only of milk, but of more widely distributed products like cheese and beef, is also a national problem. Indeed, it is an international problem as well. This can be seen from the impact of Canadian dioxin sources on the Vermont dairy farms, and, conversely, from the likely impact of U.S. dioxin sources on dairy farms in southern Canada. Nevertheless, in practical terms, just as the public outcry about localized dioxin contamination at Love Canal, or the Columbus, Ohio, municipal waste incinerator has generated a much wider range of action, regional efforts can be expected to do the same, nationally and internationally as well.

To this end, it is evident that studies such as those we have carried out on selected farms in Vermont and Wisconsin need to be extended more broadly in each of these regions, to other dairy regions, and to the "balance of trade" in airborne dioxin between the United States and its North American neighbors. It is our hope that such studies will further these wider efforts toward dioxin-free milk and, in time, secure for us all a dioxin-free future.

**APPENDIX A**

**ON-FARM OBSERVATIONS  
AND DATA COLLECTED**

Farms represent dynamic, complex agricultural ecosystems which respond to seasonal and annual changes. Farmers come from a range of cultural and educational backgrounds. Thus a farm is not the simplified laboratory where research controls are in place, measurements standardized, and confounding variable reduced or eliminated. Field research, in contrast, requires that a large number of confounding variables be identified before, during, and after the research has been completed, and assessed as to their possible role in the final data analysis. Thus every effort was made to collect detailed, site-specific data and related information that could be of some use in explaining some of the measurements either in this or subsequent studies.

The field research methods were developed based on the relevant literature, but also in close conjunction with the farmers themselves on the test farms. Farmers do not wish to have their milk contaminated with anything potentially unhealthy. At the test farm, they wanted to know if we could identify anything that they were doing in their farm practices that might increase the likelihood of dioxin reaching the milk they produced. Thus, working closely with the test farmers in each state, the field researchers collected the following types of contextual information:

**General farm data:**

*Milking Barn Type and Material*

*Bedding Material Used*

*Sanitizing Procedures & Chemicals Used (Sanitizers, Soaps, Acids, etc.)*

*Milking procedures*

*Total Number of Cows in the Herd*

*Sketch maps of farms, field and barn layout, fields, water sources etc.*

**Data regarding the dairy herd:**

*Number of Milking Cows*

*Number of Different Milking Groups*

*Types of Milking Cows*

*Cow Age Distribution*

*Average Stage of Lactation, and/or period since partial seasonal milking resumed*

*Cow Weight (average)*

*Whether the milk cows on the study farms were bred and raised on the farm or imported from somewhere else in North America.*

**Milk production data**

*Amount of Milk at Each Pickup During Sampling Period*

*Monthly Milk Production Totals*

**Cow Diet Data**

*Estimated Average Amounts of Each Diet Component Consumed by Milking Cows at the Farm.*

*Estimates of dry matter content and/or moisture content of the material.*

*Source of Each Diet Component (i.e., Where, When and How Grown).*

*Ensiling Practices at the Farm*

*Source of Water for the Cows*

*Approximately How Much Water Consumed by the Cows*

**Crop Data**

*Dairy Feed Crops Grown on the Farm*

*Acreage Planted for Each Crop*

*Detailed Species Information for Crops, If Known*

*How Each Crop Grown (e.g., planting, tilling, cutting practices and schedules)*

*Fertilizers Used on Each Crop (e.g., rates and schedule of application of each type of*

*fertilizer used, including manure)*

*Pesticides, Herbicides, Fungicides, or any other Biocides Used on Each Crop*

*Current Usage on crops*

*What has been used on crops in the past*

*How much is/was used in each application*

*When it is/was applied*

*Yield of Crops (e.g., bushels/tons per acre)*

**Other Pesticides, Herbicides, Fungicides, or Biocides Used on the Farm**

*Anything used for fly control*

*Anything else, such as any treated wood*

**Data Regarding Diesel-Fueled Vehicle Use**

*Equipment on the farm operated by diesel fuel*

*How the equipment is operated, and how often.*

*Approximate fuel usage for each piece of equipment (e.g., how often do they fill it up, and how much do they put in each time).*

*Where do they purchase the diesel fuel used.*

*Fuel storage on farm, size of tank, location etc.*

**Data Regarding Heating Fuels Used at the Farm**

*Type of fuel or fuels for heating at the farm (e.g., natural gas, fuel oil, wood)*

*Different boilers or furnaces, and where located at the farm*

*Approximate fuel usage for each boiler or furnace*

*Where they get the fuels used, and where stored, location of storage tanks*

### **Local Knowledge About Potential Air Pollution Sources**

*Each farmer provided information about various activities in the area surrounding their farms... such as the type of heating fuel that surrounding farmers use, the presence of nearby industrial facilities, etc.*

### **Other Characteristics of the Farm That Might be Relevant**

*Anecdotal and other information was noted. For example, at the pasture farm near East Troy, WI, we learned that sewage sludge from Milwaukee was used as fertilizer for many years.*

**APPENDIX B**

**UNCERTAINTIES IN SOURCE EMISSIONS  
AND DIOXIN SAMPLING**

## Uncertainties in Emission Estimates

The basic approach used in estimating emissions in this study was the following. For a given source category, a list of sources were generated based on industry, federal, state, and/or local government information sources. For each source, the throughput was estimated — e.g., the tons per year being burned in an incinerator — based on such data sources, or when these were lacking, on scientific judgement. In some cases, attempts were made to obtain throughput information directly from representatives of individual facilities, a procedure that occasionally yielded useful data. Area-based sources were handled in a directly analogous manner, with the throughputs expressed on a county basis, e.g., amount (tons/year) of backyard burning per county. Emissions from each source were then estimated from an emissions factor. The emissions factors were based on existing stack test data on similar facilities or processes. These emissions factors were then multiplied by the throughputs for a given facility to estimate the emissions for that facility. By agreement with staff of the U.S. EPA National Center for Environmental Assessment, certain inventory data were shared.

The emissions inventory was used as a direct input to the modeling analysis, and any errors in the inventory will be propagated directly through to the results. There are several areas of uncertainty in the emissions inventory. The most important of these will be briefly discussed here.

### **1. Source Existence**

Some potential sources of PCDD/F were not included in our inventory due to lack of data or resources. We attempted to include the most significant sources, of course, but some potentially significant sources were omitted. These included: accidental structure fires, e.g., consuming PVC; forest fires; magnesium manufacturing; and small commercial and industrial waste incinerators. In addition, some U.S. sources that were included were not available for Canada. These included mobile sources, backyard burning, and the various types of residential, commercial, industrial and utility-

related fuel combustion (e.g., wood and coal combustion).

Finally, for the source categories that were included, it is obviously possible that particular facilities were missed. It is difficult to assess the quality of inventory information obtained from industry groups or government sources, as the quality assurance/quality control procedures utilized are rarely specified.

## **2. Source Throughputs**

As mentioned above, estimated source throughputs were obtained when possible from industry or government data sources. It is impossible to judge the accuracy of the throughputs in such databases, as the uncertainties are not generally discussed or even acknowledged. Lacking other estimated throughput data, scientific judgement was used. Generally, there is a substantial amount of uncertainty in the throughputs used in this analysis.

In addition, unknown temporal variations in throughputs introduced additional uncertainties. Except for residential fuel consumption, all sources were assumed to have a constant throughput. The effect of time-varying throughputs was ignored, because as a practical matter, it would have been essentially impossible to obtain such data.

## **3. Emissions Factors**

By far the largest contributor to the uncertainty in the emissions estimates used in this report are the emissions factors. For some of the MSW incinerators, the emissions factors were based on facility-specific stack tests. For essentially all other source classes, emissions factors were based on stack tests of representative facilities. The emissions of PCDD/F from a particular facility depends in a complex and poorly understood way on a number of process variables, including the chemistry of the inputs, the detailed nature of the process environment (e.g., the combustion environment), and the detailed characteristics of the pollution control equipment, if present. The details of each of these factors changes continuously. Thus, it is not surprising that repeated tests on a given facility can yield dramatically different results. Even the use of stack

test data from a given facility collected on a particular day is not likely to be representative of the emissions of this facility at all times. Moreover, the application of data from stack tests on one facility to another facility — even a very closely related one — is acknowledged to be a very approximate procedure.

Emissions factors are used because, short of ongoing tests at all sources, there is no practical alternative to estimate emissions. The uncertainty in emissions factors is likely to be on the order of a factor of ten.

An illustrative example is secondary copper smelters. The emissions of dioxin from these smelters likely arises as a result of residual PVC insulation from wires being recycled or from potential inputs of other chlorinated substances to the process (e.g., chlorinated solvents or lubricating oil additives). These inputs are likely to vary substantially from facility to facility, and at a given facility, may vary substantially over time. The stack tests upon which the emissions factors for this source category are based were apparently performed some time ago (about 10 years ago). The relation of the chlorinated compound input during these earlier stack tests to that currently occurring at each facility is unknown.

#### **4. Overall Assessment**

Given the methods involved in assembling the dioxin source inventory, the greatest level of uncertainty is associated with the emission value of any *individual* source. When emission values are estimated for a number of sources of the same type, over-estimates and under-estimates may tend to cancel each other to some degree, so that the overall emission value may be more accurate than any individual one. The same consideration applies to large groups of sources of different types, for example when source estimates are classified by distance from the receptor, or by geographic orientation to the receptor. Finally, at least for the modeled estimates of the dioxin air concentration at the receptors in Vermont and Connecticut, the substantial agreement with actual measured values indicates that the total amount of emissions from all inventory sources is a reasonably good approximation of the actual figure.

The inaccuracies that we have noted here are common to all the dioxin

inventories known to us, nearly all of which have been produced by government agencies and/or commercial consultants under contract to them. Some of these problems result from the simple fact that dioxin analyses are very expensive and that both governments and the private firms that operate the sources are reluctant to fund more than a very small fraction of the necessary tests. In some cases the necessary data are simply unavailable to researchers because of confidentiality restrictions imposed by government practice, both in the United States and Canada. Even simple information, such as source locations, throughput and control systems, which are generally required by government agencies for regulatory purposes, are often unavailable from the agencies. We have endeavored to overcome some of these difficulties by undertaking, whenever possible, intensive inquiries through telephone interviews and literature searches to find missing data and check doubtful inventory entries — procedures that, in our experience, are not widely practiced.

#### Sampling Uncertainties

In this study, samples of air, milk, diet components, and crops were analyzed for PCDD/F. It is important to consider uncertainties in interpreting the results of this sampling.

#### **5. Quality Assurance / Quality Control Procedures**

First, several field-initiated quality assurance procedures were performed. These included collection and analysis of duplicate samples of air, milk, and vegetation. Except for the special case discussed below, there was satisfactory consistency between the results for the duplicate samples in all cases. In addition, field blanks for milk and vegetation samples were collected. Unfortunately, the field blanks collected in Wisconsin were irretrievably lost at the laboratory. Field blanks collected in Vermont showed very low levels of PCDD/F. A number of laboratory-initiated procedures were carried out as well, including the preparation and analysis of method blanks and method spikes. These procedures yielded satisfactory results.

Despite the quality assurance/quality control approaches discussed above, there

were several significant sources of uncertainty in the sampling results that emerged in this analysis. The most important of these will be discussed briefly below.

## **6. Very Low PCDD/F Levels in Vegetation and Cow Diet Samples**

The largest element of uncertainty in the sampling results concerns the samples of diet components and vegetation. The levels of PCDD/F in most of these samples were very low, relative to the overall analytical sensitivity of the sampling and analysis procedures. In many cases, concentrations of individual congeners were below the detection limit. Most importantly, even when PCDD/F was detected, it was, for most of the vegetation/diet samples, only at levels comparable to — or slightly higher than — the laboratory method blanks. In reducing the data, the values of the laboratory blanks for a particular set of analyses were subtracted from the actual sample results. This procedure yielded a blank-corrected sample value for each PCDD/F congener and homologue group. For example, if the concentration of 2,3,7,8-TCDD in a sample was reported to be 1.1 pg/g, and the laboratory method blank was reported to have a 2,3,7,8-TCDD concentration of 1.0 pg/g, the blank value was subtracted from the sample value to yield a blank-corrected sample value of 0.1 pg/g. Because the sample values were generally comparable to or only slightly higher than the blank values, the uncertainty in these blank-corrected values is very significant. To estimate the magnitude of this uncertainty, the following data analysis procedure was carried out.

The starting point for the analysis were the data reported on each sample analyzed by Midwest Research Institute (MRI) for each of the seventeen 2,3,7,8-substituted PCDD/F congeners and eight homologue groups (e.g., total TCDD, total HxCDF). Thus, a total of twenty-five analytes were available for each sample. If a particular congener or homologue group was found at concentrations above the detection limit, the result was reported by MRI as a quantified concentration value (e.g., pg/g of dried sample). However, in many cases, one or more of the analytes were not found in the analytical procedure for a given sample. In these cases, the analyte was reported as "non-detected" in the sample. MRI used a criterion of 10:1 sample to noise ratio as the threshold for detection of a given analyte. That is, if the analyte's signal

was greater than 10 times that of the background electronic noise signal in the instruments, then it was reported as being detected. The samples were analyzed by MRI in batches, generally with on the order of 5-10 samples analyzed in each batch. In each batch, a method blank was analyzed. For vegetation and cow diet samples, this method blank was cleaned quartz sand. For milk samples, this method blank was lab-purified water. In some cases, analytes were found at concentrations above detection limits in these method blanks.

The analysis of MRI-reported data was conducted in the following way. For each sample, three initial sets of results were prepared. In the first set, it was assumed that the concentration of all non-detected analytes was zero. In the second set, it was assumed that the concentration of all non-detected analytes was one-half the detection limit. In the third set, it was assumed that the concentration of all non-detected analytes was equal to the detection limit. For detected compounds, the same concentration (i.e., the reported concentration) was used in each of the three initial data sets. These three sets comprise the first data *suite* for each sample.

Then, a second suite of results was prepared by *subtracting* the relevant method blank from each sample. In this second suite of results, "parallel" treatments of non-detects were used, i.e., the same assumption was made regarding non-detects in the sample *and* the method blank. Thus, in the first set, it was assumed that the concentration of all non-detected analytes was zero in *both* the sample and the method blank. In the second set, it was assumed that the concentration of all non-detected analytes was equal to one-half the detection limit in *both* the sample and the method blank. Similarly, in the third set, it was assumed that the concentration of all non-detected analytes was equal to the detection limit in *both* the sample and the method blank. In each of the three data sets in this suite, if the concentration of a given analyte was greater in the method blank than in the sample, then a value of "0" was assigned to the analyte for that set for that sample. This second suite of results for each sample comprise one portion of the basic "blank corrected" results from this analysis, i.e., the parallel blank correction method results.

A third suite of results was prepared using an alternate method. In this

treatment, an attempt was made to estimate the maximum and minimum possible analyte concentrations in each sample. A minimum value for each analyte was estimated by assuming that the non-detected analytes in the sample were present at zero concentration, and non-detected analytes in the method blank were present at the detection limit. In either case, if the analyte was detected, it was simply recorded at its reported value. Then, the maximized method blank values were subtracted from the minimized sample values to create a set of minimum possible sample concentrations. A maximum possible sample value was estimated in an analogous way, i.e., by subtracting a minimized blank value (non-detects = zero) from a maximized sample value (non-detects = detection limit). In either of these treatments, if the concentration of a given analyte was greater in the method blank than in the sample, then a value of "0" was assigned to the analyte for that set for that sample. An arithmetic average of these minimum and maximum possible sample values was also estimated. This third suite of results for each sample comprises another portion of the basic "blank corrected" results from this analysis, i.e., the non-parallel blank correction method results.

The overall range of results encompassed by the above methods was used to estimate the uncertainty in the sampling results.

For the milk and air samples, the sample concentrations were generally relatively higher than the method blanks and there were far fewer non-detected congeners or homologue groups. Thus, the level of uncertainty in the milk and air sample data would appear to be much less than that of the vegetation and cow diet samples.

## **7. Representativeness of Sampling**

At three of the farms, weekly total diet composite samples were collected and analyzed, as were weekly samples of the individual diet components. When properly weighted by the proportions in the diet, the sum of the PCDD/F contents of the individual components should equal the PCDD/F level in the total diet composite sample -- *if* each sample is perfectly representative and there are no sampling or analysis errors.

It was found that the mean PCDD/F levels — estimated in the two different ways described above — varied by approximately a factor of two. However, the uncertainty ranges in the two results generally overlapped, indicating that the results *could* actually be the same. Nevertheless, the differing midrange estimates for total diet PCDD/F perhaps gives a sense for the overall uncertainty in the diet sampling results, i.e., the overall results may be accurate within approximately a factor of two.

**APPENDIX C**

**MEETINGS WITH STAKEHOLDERS**

Typically, scientific research and policy research are undertaken as two distinctly separate aspects of scholarly endeavor. The scientific research is completed and handed to the policy maker to determine what the science means in terms of possible policy implications. Citizens may not be involved in the process until policies are proposed. In contrast, this study aimed to provide both scientific findings and policy recommendations. For this purpose, we endeavored to work with stakeholders throughout the process.

*Farmers:* The farmers at the test farms in both states were actively involved in developing many aspects of the field research. They received briefings by the field researchers who were available to answer questions as the work progressed. In Vermont, each farmer and sometimes their staff people met with Jean Richardson and the other field researchers, who shared with them the preliminary results and sought their perspectives on possible sources of dioxin contamination. Meetings also took place one-on-one with farmers when the data were developed, and follow-up will continue with the completion of this report.

*Dairy Cooperatives:* In Wisconsin one of the Dairy Cooperatives provided names of farmers from whom the test farms were selected, working through the University of Wisconsin Extension System personnel. In Vermont, a detailed briefing of the proposed research was given by the Vermont research team to the regular monthly meeting of the St. Albans Dairy Cooperative, the largest such cooperative in the State. The members had been informed that we would be making a presentation and thus there was a large turnout and a useful discussion ensued. Vermont is a small state with a small number of dairy farms. They expressed considerable concern over the possible economic impact of our findings, and a desire to understand which instate or out of state emission sources were causing the air pollution reaching their farms.

*Agency Personnel:* In Vermont, personnel at the state Agency of Natural Resources, Air Pollution Control Division were actively involved in several aspects of the research, from sharing inventories, to suggested analysis of emissions modeling data, and review of manuscript. They also have provided, through these regular

conversations, guidance on the more useful forms of policy recommendations as seen from their practical aspect in State government.

*Group Stakeholder meetings* were also scheduled at the beginning of the research with the State Department of Agriculture, meeting together with representatives from each of the offices of the Vermont Congressional Delegation, one Democrat, one Republican and one Independent. This was a well-attended meeting with a detailed discussion. A similar meeting will be held following public release of this report in early Fall 1998.

*Congressional meetings:* In addition to the above group meetings, the principal investigator in Vermont has met individually the Vermont Congressional delegation senior staff. All three Vermont Congressmen have continued to follow this research and provided useful policy comments.

*Stakeholder meeting with Vermont Governor, senior staff, Commissioner of Health and Commissioner of Agriculture.* Jean Richardson provided a briefing for the Governor and his senior staff early in the research. The discussion that followed focused on the possible economic implications of the results of the research.

*University Colleges of Agriculture.* Staff at both the university of Vermont and the University of Wisconsin-Madison provided assistance with aspects of the research.

*Health Professionals:* In Vermont, as the preliminary dioxin research results began to come in, Jean Richardson, was invited to work with several personnel at the university regional hospital (Fletcher Allen), and throughout New England. This resulted in an educational teleconference in Spring 1998, funded by the John Merck Fund, and led Ellen Cepetelli, a nurse. The Conference focused on causes of dioxin and mercury contamination in hospitals, and provided concrete examples of how the health care industry can reduce, and in some areas eliminate dioxin and mercury pollution.

*Advocacy groups:* although no formal meetings have taken place, informational conversations between individuals have taken place regularly, and following release of the report citizen activists will be invited to stakeholder meetings.

In sum, the research undertaken is interdisciplinary in nature; no one person can understand all aspects in detail. Similarly, there are several diverse and possibly

conflicting policy solutions that must be evaluated by the stakeholders, and the foregoing discussions were devoted to that end.