A_o = relative cross-sectional area acted upon by single drop
A_s = relative projected surface per particle, square feet
d_a = air density, pounds per cubic foot
d_w = particle density, pounds per cubic foot
g = gravity constant, feet per minute per minute
K = constant applying to all test data
L = length of pattern, feet
m = particle mass corrected for buoyancy effect, $m\left(\frac{d_w - d_a}{d_w}\right)$
n = number of holes in pan
N = relative number of drops per hole
P = pressure
Q_A = net air flow, cubic feet per minute
Q_w = water flow, cubic feet per minute
r = relative particle radius = hole radius, feet
R = air resistance to falling particle, pound feet per minute per minute
s = distance of fall, feet
V = velocity of any falling body, feet per minute
V_1 = velocity intercept at $Q_A = 0$, feet per minute
V_A = air velocity, feet per minute
V_T = terminal settling velocity of particle, feet per minute
V_w = maximum velocity of water droplet, feet per minute
W = width of pattern, feet

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Sulfur Dioxide in the Atmosphere and Its Relation to Plant Life

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Sulfur dioxide from industrial gases in low concentrations is widely distributed in the atmosphere. In exposures of sufficient duration to concentrations higher than about 0.40 p.p.m. it may be toxic to sensitive plants at periods during the growing season when physiological activity is high and the conditions for rapid absorption of this gas by the leaves are at a maximum. However, low concentrations, in the range up to 0.10 to 0.20 p.p.m., have been demonstrated to be without influence on plant life, in the absence of visible markings. There is no effect, in this case, after long-continued exposure on rate of growth, yield of crop, photosynthesis, respiration, or on the daily march of the stomata. The effects may be beneficial if the plants are growing in a sulfur-deficient soil or nutrient. No basis has been found for the theory of invisible injury. The literature on the subject has been reviewed and the results of investigations carried out by the writer and his collaborators on various aspects of the sulfur dioxide pollution problem have been presented: occurrence of sulfur dioxide in the atmosphere of industrial areas; sulfur content of vegetation; effect on soils; symptoms and diagnosis of injury from sulfur dioxide and other factors; retardation of diameter increment in conifers; experimental studies on the influence of environmental factors on susceptibility, the effects on conifers in natural habitat and transplanted stock, yield of crop plants, stomatal behavior, and photosynthesis and respiration. It is hoped that the methods and results described will serve as a guide in investigations of effects of other industrial waste gases on plant life.

CONTAMINATION of the atmosphere by sulfur dioxide from combustion products of industrial and smelting operations and the resultant effects on vegetation have been the object of intensive investigation for nearly 100 years. Early work on this subject was hampered by the lack of accurate methods for determining concentrations in the air in the vicinity of affected vegetation. Consequently, the observations were more or less qualitative in character and generally confined to a study of the symptoms on the leaves. This was later supplemented by more or less crude experiments in which incredibly high concentrations were applied to experimental plants under cabinets to reproduce the gross symptoms of damage. It is probable that experimentation with reliably determined concentrations of sulfur dioxide, under known conditions, was first introduced into this subject by the Selby Smelter Commission (*33*) in 1915. However, more accurate and systematic observations were rendered possible after the development of a continuous, automatic method of analysis by Thomas in 1928. Subsequent modifications and adaptation of this apparatus to carbon dioxide and other gases have introduced a high degree of precision in atmospheric pollution investigations (*92–95*, *103*, *104*).

One of the most comprehensive investigations ever undertaken on this subject was organized in 1929 when the National Research Council of Canada was requested to study the problem of alleged damage in Stevens County, Wash., by smelter fumes emitted from the stacks of the Consolidated Mining and Smelting Company of Trail, B. C. The investigation was initiated with measurements of the sulfur dioxide concentrations in the atmosphere at various points in the Columbia River valley by means of Thomas automatic recorders, surveys of the condition of field and forest crops, and the sulfur content of vegetation. But, as time went on, the scope of the work was enlarged to include a great amount of experimental work, as well, on the effect of sulfur dioxide on conifers and crop plants under controlled conditions. The work covered a period of about 8 years and the results were published in book form in 1939 (*35*).

Previous investigations were concerned mainly with the influence of high concentrations of sulfur dioxide on vegetation—that is, concentrations in excess of 1 p.p.m. It soon became evident, however, that the majority of the fumigations in the Columbia valley were of comparatively low intensity but long duration, and concentrations above 1 p.p.m. were encountered only infrequently.

The problem presented by the effects of low concentrations of sulfur dioxide on plant life is of outstanding importance to industry, agriculture, and forestry because such conditions are encountered wherever industrial operations and centers of population are located in areas adjacent to rural and forest regions. The damaging character of high concentrations on vegetation can be readily ascertained by the trained observer because of the visible and specific symptoms produced on the foliage of sensitive plants. Until recently, however, the influence of sulfur dioxide in long exposures below the toxic level, where visible symptoms were not produced on the leaves, was not properly understood, and, in fact, was the subject of a long-standing controversy.

The experimental difficulties involved in a proper solution of this problem were probably responsible for the theory of "invisible injury" advanced by earlier investigators such as Wieler (*110, 112, 113, 115*), Stoklasa (*86*), and others (*23, 34, 85*), who maintained that sulfur dioxide in any concentration in the atmosphere, no matter how small, was harmful to plants and reduced the carbon dioxide assimilation and rate of growth. The opposite view was held by some who claimed that growth could be stimulated by such low concentrations (*29, 69*).

A direct attack on this question was made by the author and his collaborators (*45–48*) in field plot experiments, under controlled or measured conditions, dealing with the influence of low, nonmarking concentrations on the rate of growth, carbon dioxide assimilation, stomatal behavior, and chemical composition of sensitive plants. In many of these experiments the plants were grown to maturity in a continuous atmosphere of sulfur dioxide. Independent investigations on various aspects of the same problem were conducted by Thomas and Hill (*96–98, 102*) in both field plot and greenhouse experiments involving careful studies of the photosynthesis and respiration, yield, and sulfur nutrition. Swain and Johnson (*90*) have described the effects of exposures for long periods to low concentrations of sulfur dioxide on wheat plants grown in nutrient solutions under almost ideal and constant conditions of light intensity, humidity, and temperature. Finally, Setterstrom, Zimmerman, and Crocker (*80, 84*) attempted to determine the "invisible" effects of sulfur dioxide under conditions unfavorable as well as favorable to plant growth in greenhouse experiments in which water supply, nutrient supply, and age of plants were varied to simulate natural field conditions. The bearing of these investigations on the "invisible injury" theory will be discussed in subsequent sections of this paper.

Other aspects of atmospheric pollution by sulfur dioxide, such as the concentrations usually found in the atmosphere of industrial areas, the sulfur content of vegetation and its usefulness in defining the limits of gas penetration in a given area, the effect on soils and water supplies, the influence of injurious fumigations on diameter increment of conifers, the experimentally determined effects of environmental factors on susceptibility, and finally, the influence of low, medium, and high concentrations on stomatal behavior, carbon dioxide assimilation, yield, and chemical composition of plants are also presented here in the hope that the facts will serve to broaden our understanding of the general problem of atmospheric pollution.

SULFUR DIOXIDE CONTENT OF THE ATMOSPHERE

The Trail Area. The smelter at Trail, B. C., is situated in a relatively narrow valley on the west bank of the Columbia River at a point about 11 miles by river from the international boundary. The valley here is flanked on both sides by mountain ranges which rise to a height of several thousand feet. This constitutes a natural channel for the smelter gases which are confined to the valley and drift either northward or southward according to the prevailing winds. On occasion the smelter fumes also penetrate up the various tributary valleys of the Columbia River. It is evident, therefore, that the topography of the Trail area has an important bearing on the problem of dilution of the sulfur dioxide emitted and on the smoke control measures subsequently employed to reduce the hazard to vegetation.

The first complaints of damage in northern Stevens County, Wash., coincided with an increase in sulfur emission and the use of two high stacks to serve the enlarged plant in 1925. During the period 1926 to 1930, the rate of sulfur emission rose to about 9000 to 10,000 tons a month from a previous high of 5000 tons, owing to the increased production of metals. During these years the greatest damage to vegetation was produced in northern Stevens County, Wash.

Measures to reduce the emission of sulfur have been a feature of the smelting operations at Trail throughout the history of the Consolidated Mining and Smelting Company. These consisted of improvements in flotation, smelting practice, dilution by the use of 400-foot stacks and, since 1930, recovery of sulfur dioxide as sulfuric acid and elemental sulfur. The sulfur recovery plants now form the basis of a huge heavy chemicals industry and include the following steps:

1. Concentration of sulfur dioxide
2. Sulfuric acid production
3. Nitrogen fixation and production of ammonia
4. Phosphoric acid production
5. Reduction of sulfur dioxide to elemental sulfur with coke
6. Production of phosphate, ammonium sulfate, and ammonium phosphate–sulfate fertilizers (*10, 12*).

By 1939 the total expenditures on these recovery plants was in excess of $15,000,000 and during that year about 70% of the sulfur charged to the furnaces was recovered. The total recovery capacity had been increased to about 600 tons of sulfuric acid and 140 tons of elemental sulfur per day.

In addition to the above measures, a weather plan of smoke control was instituted in 1934 by the company under the direction of F. Matthews and has been in successful operation ever since. This consisted in reducing the sulfur emission from the lead and zinc roasters or shutting them down entirely during periods of the growing season when the meteorological conditions were favorable for injury to vegetation. This program was at first carried out by visual observations of the extent of diffusion of smoke in the valley, correlated with barometric pressure readings and the indicated concentrations at two sulfur dioxide recorder stations.

However, a more specific régime was applied to prevent the occurrence of injurious fumigations of sulfur dioxide in the State of Washington by regulation of the maximum emission from the smelter stacks according to wind turbulence, other meteorological data, and recorded sulfur dioxide concentrations. Some of the general restrictions were that during the growing season, when turbulence was bad and the wind direction unfavorable, the emission should be reduced by 4 tons of sulfur per hour or stopped completely if an automatic recorder at Columbia Gardens (about 5.5 miles southeast of Trail) indicated 0.30 p.p.m. or more of sulfur dioxide for 40 minutes, until the recorded concentration was reduced to 0.20 p.p.m. or less for 1 hour. In the nongrowing season a similar restriction was enforced, if the Columbia Gardens recorder indicated 0.50 p.p.m. or more of sulfur dioxide for 1 hour (*10*).

The International Tribunal appointed by the United States and Canada to adjudicate the Trail smelter question was assisted by two technical consultants, Dean and Swain, who supervised a thorough study of the influence of meteorological factors on the diffusion and dispersion of the fumes emitted from the Trail smelter stacks during 1938 to 1940 (*10*). The results indicated the relation between meteorological conditions in this valley and the mode of occurrence of fumigations which were classified into diurnal and nondiurnal types. The former occur most frequently during the growing season as a result of the disturbance of the thermal balance of the atmosphere by the action of the sun on the earth's surface causing a differential heating (or cooling) of the sides of the Columbia valley. The gas contained in the upper

Table I. Average Atmospheric Sulfur Dioxide during Growing Season

Year	Monthly Emission of SO_2 to Atmosphere, Tons	Average Concentration, P.P.M.					
		April	May	June	July	August	September
	15 Miles, by River, South of Trail, B. C.						
1930	20,000	...	...	0.030	0.050	0.040	0.080
1931	14,200	0.034	0.029	0.062	0.023	0.018	0.026
1936	12,200	0.023	0.033	0.028	0.011	0.014	0.023
1937	9,600	0.020	0.013	0.010	0.007	0.009	0.020
	20 Miles, by River, South of Trail, B. C.						
1936	12,200	0.028	0.031	0.020	0.013	0.013	0.021
1937	9,600	0.016	0.011	0.008	0.006	0.007	0.019
1940	7,600	0.011	0.016	0.007	0.008	0.008	0.018
	30 Miles, by River, South of Trail, B. C.						
1930	...	0.020	0.022	0.018	0.028	0.032	0.046
1931	...	0.004	0.009	0.019	0.008	0.006	0.006

Table II. Maximum Atmospheric Sulfur Dioxide during Growing Season

Year	Maximum Concentration, P.P.M.					
	April	May	June	July	August	September
	15 Miles, by River, South of Trail, B. C.					
1930	...	...	0.92	0.90	1.05	2.11
1931	0.77	1.25	0.87	0.94	0.55	0.74
1936	0.68	0.76	0.60	0.38	0.42	0.42
1937	0.32	0.30	0.34	0.16	0.25	0.48
	20 Miles, by River, South of Trail, B. C.					
1936	0.58	0.75	0.31	0.46	0.56	0.80
1937	0.33	0.28	0.28	0.16	0.26	0.40
1940	0.37	0.23	0.22	0.19	0.17	0.23
	30 Miles, by River, South of Trail, B. C.					
1930	0.56	0.48	0.45	0.36	0.50	0.93
1931	0.35	0.25	0.41	0.45	0.17	0.20

atmospheric levels is thus brought to the surface almost simultaneously at recorder stations many miles apart. The nondiurnal type predominates during the nongrowing season and is brought about usually by surface winds, the sulfur dioxide being recorded first at the station nearest the smelter and at the others at later intervals (*10, 30*).

Some indication of the concentrations found in the atmosphere south of the smelter at Trail, B. C., in the Columbia River valley on the United States side of the international boundary may be found in Tables I to IV. Determinations were made continuously by means of Thomas automatic sulfur dioxide recorders at stations about 15, 20, and 30 miles distant by river (*36*). In 1937 the recorders were set up closer to the smelter, one being installed at a point about 5.5 miles south and another at the international boundary, the third station was left at Northport, 20 miles distant. The maximum concentrations recorded at points 15 and 30 miles south of the smelter during the entire period of investigation were 2.11 and 1.11 p.p.m., respectively, and the highest monthly averages of all readings were 0.115 p.p.m. and 0.106 p.p.m., respectively.

As a rule the average and maximum concentrations were considerably higher during the winter months than in the crop growing season, and in this respect the situation was no different from that observed in many other industrial areas. With the reduction in emission of sulfur at Trail and the operation of the control régime, the duration of the higher concentrations, especially during the growing seasons, fell off quite sharply until by 1937 very few of the recorded concentrations were higher than 0.25 p.p.m. (Figures 1 to 4). In fact, by 1937 the gas conditions in this area were improved to such an extent that the recorded concentrations were considerably lower than those found in many of the industrial areas of the United States, Great Britain, and elsewhere.

Sulfur Dioxide in Other Areas. The Boyce Thompson Institute has reported for a 2-year period at Yonkers, N. Y. (*79, 82*), an average concentration, including zero readings, of 0.034 p.p.m. and a maximum of 0.75 p.p.m. Studies by the Mellon Institute (*59, 60*) of the atmosphere within a radius of 25 to 30 miles of some of the larger cities in the United States indicated, during the heating season from October 1936 to April 1937, a maximum of 2.266 p.p.m. and an average of 0.261 p.p.m. in the 0- to 5-mile zone of East St. Louis, a maximum of 1.07 p.p.m. in the Pittsburgh district, and lower average and maximum concentrations in the Philadelphia, Camden, and Detroit areas, but nevertheless comparable in order of magnitude to the concentrations found 15 miles south of Trail in 1937. With reference to Detroit, an investigation carried out by the author in 1934, during March and April, in the Windsor-Detroit area, indicated a maximum of 1.93 p.p.m. and averages in individual fumigations of 0.039 to 0.396 p.p.m. in portable tests at such times when the wind was carrying smoke from the Detroit area (*36*). During this period, measurements made by means of an automatic recorder showed the presence of sulfur dioxide in concentrations of 0.10 p.p.m. or over on about 60% of the days.

Reports of the Department of Scientific and Industrial Research of Great Britain (*11*) indicate comparatively high maximum and average sulfur dioxide concentrations in the industrial areas of large towns and cities; the air of Glasgow, Manchester, and Leeds showed a similar degree of pollution to that of the London area (*72*). However, sulfur dioxide occurrence is less and the fumigations are milder during the growing season than in the winter months. This is due not only to the increased volume of smoke emitted during the winter months but to the less favorable meteorological factors governing dilution, such as poor turbulence, low temperatures, low wind velocity, and a considerably higher incidence of fog. Under such unfavorable weather conditions, concentrations up to about 10 p.p.m. have been reported in many industrial areas, (*4, 33, 36, 60, 77, 88*).

The monthly fluctuations in the occurrence of sulfur dioxide, as influenced by the seasons and corresponding meteorological factors, have been shown to give rise to an annual cycle in the form of a sine-shaped curve, with a maximum in midwinter and a minimum in midsummer. This cycle may be typical of condi-

Table III. Average Atmospheric Sulfur Dioxide during Nongrowing Season

Year	Average Concentration, P.P.M.					
	January	February	March	October	November	December
	15 Miles, by River, South of Trail, B. C.					
1930	...	...	...	0.115	0.110	0.060
1931	0.109	0.105	0.049	0.016	0.025	0.048
1936	0.071	0.092	0.025	0.032	0.066	0.048
1937	0.069	0.094	0.096	0.049	0.085	0.039
	20 Miles, by River, South of Trail, B. C.					
1936	0.061	0.086	0.024	0.026	0.049	0.065
1937	0.062	0.080	0.073	0.042	0.040	0.030
1940	0.056	0.075	0.030	...	...	...
	30 Miles, by River, South of Trail, B. C.					
1930	0.036	0.069	0.059	0.054	0.080	0.043
1931	0.106	0.052	0.022	0.005	0.010	0.037

Table IV. Maximum Atmospheric Sulfur Dioxide during Nongrowing Season

Year	Maximum Concentration, P.P.M.					
	January	February	March	October	November	December
	15 Miles, by River, South of Trail, B. C.					
1930	...	...	...	1.31	1.36	1.08
1931	1.30	1.47	1.31	0.48	1.00	0.78
1936	0.85	0.75	1.47	0.60	1.25	0.72
1937	0.84	0.64	0.80	0.55	0.45	0.45
	20 Miles, by River, South of Trail, B. C.					
1936	0.56	0.82	1.04	0.60	1.24	0.51
1937	0.92	0.66	1.15	0.76	0.30	0.32
1940	0.71	0.97	0.88	...	...	...
	30 Miles, by River, South of Trail, B. C.					
1930	0.59	0.80	0.59	0.61	0.85	0.48
1931	1.00	0.51	0.54	0.11	0.44	1.11

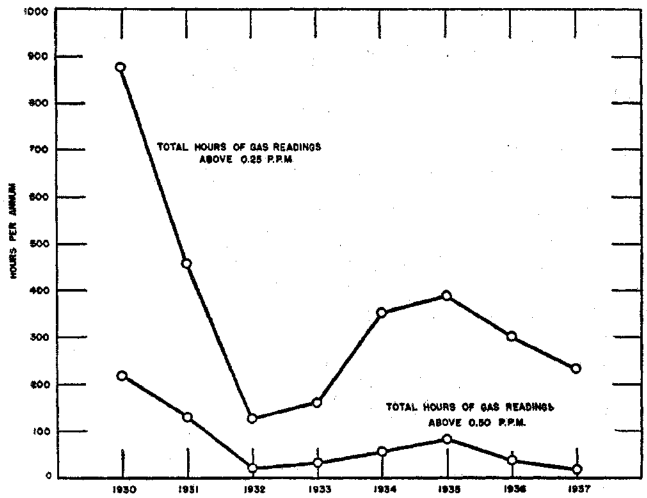

Figure 1. Sulfur Dioxide Readings at Stroh Recorder

Total hours above 0.25 and above 0.50 p.p.m. per year, approximately 15 miles south of Trail, B. C.; 1930 values estimated from the period June to December

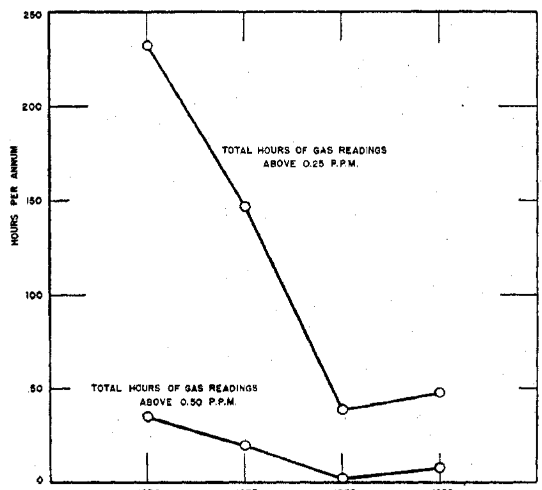

Figure 2. Sulfur Dioxide Readings at Northport Recorder

Total hours above 0.25 p.p.m. per year, approximately 20 miles south of Trail, B. C.

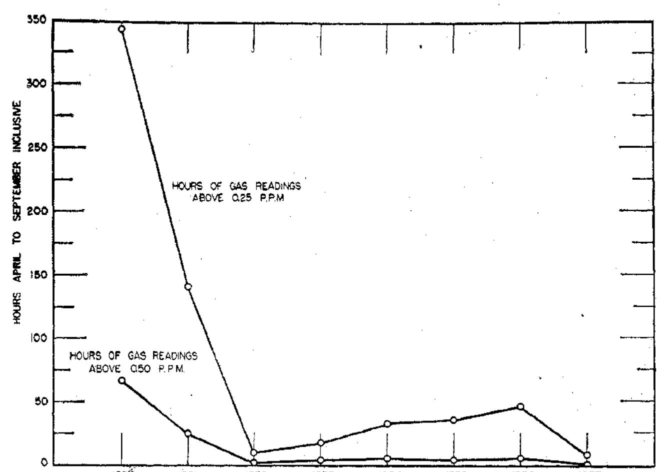

Figure 3. Sulfur Dioxide Readings at Stroh Recorder during Growing Seasons

Total hours above 0.25 and above 0.50 p.p.m., April–September, inclusive; 1930 values estimated from the period June to September

tions in most industrial areas as shown by data obtained in surveys at such widely separated locations as at the Boyce Thompson Institute (*120*), at Leicester, England (*13*), and in unpublished results obtained by the author at St. Catharines, Ontario. However, the reverse is true at Selby, Calif. (*32*).

Oxidation Products of Sulfur Dioxide. Sulfur dioxide, after release to the atmosphere, undergoes a slow oxidation to sulfur trioxide and sulfuric acid; the reaction is catalyzed by sunlight and fine dust particles, especially by minute metallic oxide particles present in smelter smoke. Observations in the Sudbury, Ontario, smelter region by means of portable equipment in the path of the smoke stream during field fumigations indicated the presence of a measurable amount of sulfuric acid or sulfate with considerably higher amounts of sulfur dioxide. The apparatus consisted of a pair of carefully matched aspirating units containing absorbers equipped with fritted glass disks of medium porosity and balanced flowmeters. Measured volumes of air were passed simultaneously, in finely dispersed bubbles, through 25 ml. of a solution containing 0.006% hydrogen peroxide in $5 \times 10^{-5} N$ sulfuric acid in one absorber and a similar volume of stabilized iodine-starch solution, $6 \times 10^{-5} N$, in the other absorber. The aspirated solutions were washed out of the absorbers into Pyrex bottles and the change in conductivity measured by a Wheatstone bridge for the hydrogen peroxide solutions, while the iodine solutions were analyzed by a calibrated photoelectric method.

The results of over 550 sets of simultaneous observations were grouped according to the character of the fumigations into heavy, medium, and light smoke visitations. In heavy fumigations, with an average of 1.16 p.p.m. (calculated as sulfur dioxide), the conductivity values were 10.4% higher than the specific sulfur dioxide concentrations as indicated by the iodine method. In medium fumigations, of an average value of 0.48 p.p.m., the conductivity measurements were 17.1% higher and in light fumigations, averaging 0.20 p.p.m., the conductivity values were 19.8% higher. The differences between the means were subjected to the t test and were found to be significant to better than the 0.01% level of probability. As it is known that high concentrations persist for a much shorter time in a given area than low concentrations, the data indicate in an indirect manner the extent to which sulfur dioxide is oxidized in the atmosphere before its complete dispersal or removal by natural processes.

Removal of Atmospheric Sulfur Dioxide by Natural Processes. Although the total amount of sulfur dioxide released to the atmosphere may be enormous in large industrial or smelter areas, its dilution and subsequent removal by natural air-cleaning phenomena, when the meteorological conditions are not unfavorable, are comparatively rapid processes. Dilution and removal may be brought about by eddy-diffusion and thermal tur-

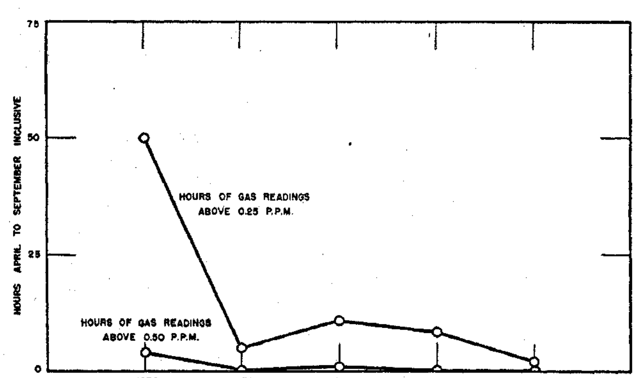

Figure 4. Sulfur Dioxide Readings at Northport Recorder during Growing Seasons

Total hours above 0.25 and above 0.50 p.p.m., April–September inclusive

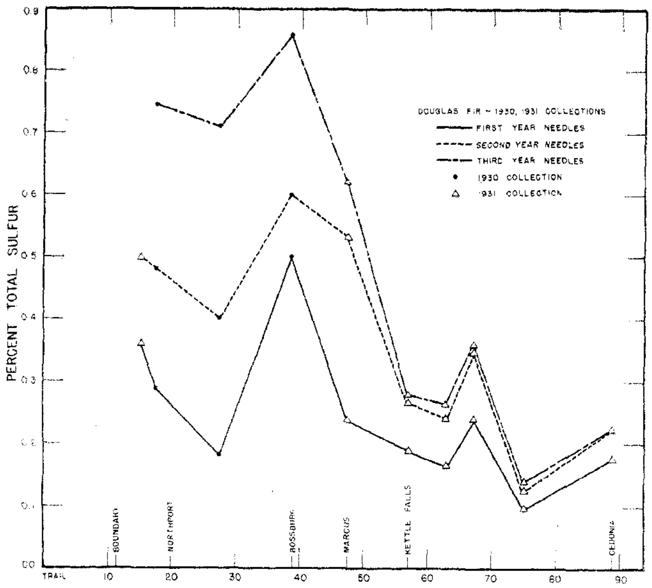

Figure 5. Effect of Smelter Smoke on Absorption of Sulfur by Leaves of Douglas Fir

Columbia River Valley, Stevens County, Wash.

bulence, absorption by land and water surfaces and by vegetation, rain clouds, rain, dew, and snow.

The rate of dilution with distance on principles of eddy-diffusivity has been derived theoretically by Bosanquet and Pearson (*5*) as an inverse square law of distance where the mass rate of emission from a stack, without regard to the extent of dilution of the stack gas, is the controlling factor (*32*). This law has been applied with some success in evaluating ground concentrations of sulfur dioxide in level terrain where the mass rate of emission from the stack and wind conditions are known. More accurate expressions have been deduced by Sutton (*87*) by the introduction of a parameter for the diffusing power of the turbulence and by use of virtual diffusion coefficients which depend on the value of the above parameter and the gustiness of the wind. Sutton's analysis is more applicable to the diffusion of sulfur dioxide in the mountainous Trail area.

On many occasions in the Trail and Sudbury regions the fumigations have been observed to decay exponentially with time if the main smoke stream has been diverted by a change in wind direction to another path. The concentrations decrease according to a relation, $C = C_0e^{-bt}$, where C_0 and C are the initial and final concentrations, t is the time and b is a coefficient denoting the rate of mixing with clean air or the number of air changes in unit time. It has been estimated that the average life of a suspended smoke particle in the air is about 6 days and of a sulfur dioxide molecule, about 1.5 days (*13*).

SULFUR CONTENT OF VEGETATION

The sulfur content of leaves of plants exposed to atmospheric sulfur dioxide increases substantially over a period of time, provided the leaf tissue is functioning in a normal manner (*38*). This increased sulfur may reach a value several hundred per cent greater than the average value normally found in plants of the same species. However, unless the concentration and exposure to gas and other factors are known accurately, there is no quantitative relation between this increase and the degree of injury because the sulfur content is subject to great variation in normal plants (*24, 63, 91*). Nevertheless such data, from comprehensive collections of certain sensitive plants, may be used to define the area within which this gas occurs. Thus, in the Trail area, both conifers and broad-leaved trees and shrubs had a substantially higher sulfur content within the smoke zone, and the values decreased with increasing distance from the smelter until at a sufficient distance the sulfur content became similar to that of control collections from areas on similar soil but situated beyond the influence of smelter fumes (*38*). If proper care is exercised in the collection of the samples this method may be extremely useful in defining the farthest limits of penetration of gas in a smelter area. In general, an abnormal sulfur content in vegetation may be traced far beyond the limits of any recognizable injury to forest or crop plants. Conifer needles remain on the trees for 3 years or longer, and throughout most of this time there is a slow accumulation of sulfur unless the plant tissue has been injured by the presence of gas. Conifers accumulate sulfur by absorption of sulfur dioxide mainly during the growing season; there is little, if any, accumulation in the winter period as shown in Table V.

The curves in Figures 5, 6, and 7 indicate the trend in sulfur content with distance for conifers and broad-leaved species within the Columbia River valley. In the case of yellow pine and Douglas fir there is a considerable spread in sulfur content between the needles of different years within the area influenced by smelter fumes. In 1930 to 1931 the extreme limits of penetration of sulfur dioxide extended to about 58 to 62 miles south of the smelter, but in 1934 and later years the distance was considerably less. Similar results for this area were reported by Fisher, Goldsworthy, and Griffin (*17*) and more recently by Griffin *et al.* (*19*).

Among other investigations which have shown a significant trend in sulfur content of vegetation with distance from the source of pollution may be mentioned those of Rusnov (*75*), Haywood (*27*), Lorenz (*56*), Cohen and Ruston (*8*), and McCool and Johnson (*57*).

INFLUENCE OF SULFUR DIOXIDE ON SOILS AND WATER SUPPLIES

The early literature on the effect of sulfur dioxide on soils has been reviewed critically by Haselhoff, Bredemann, and Haselhoff (*23*). This gas may be absorbed directly by the soil, may reach the soil by deposition with the atmospheric precipitation, or more indirectly through absorption by vegetation. Because sulfur is an element which is essential to plant life (*61, 62, 70*) and is being constantly removed from the soil in agricultural areas by the harvesting of crops (*22*) or by the drainage of soluble sulfates during periods of precipitation, some method of renewal is necessary if serious depletion is to be avoided. However, there is no doubt that this depletion has been averted in those regions where sufficient sulfur for plant requirements has been supplied in the form of atmospheric sulfur dioxide carried by the prevailing winds from industrial areas. Alway (*1, 2*) has suggested that

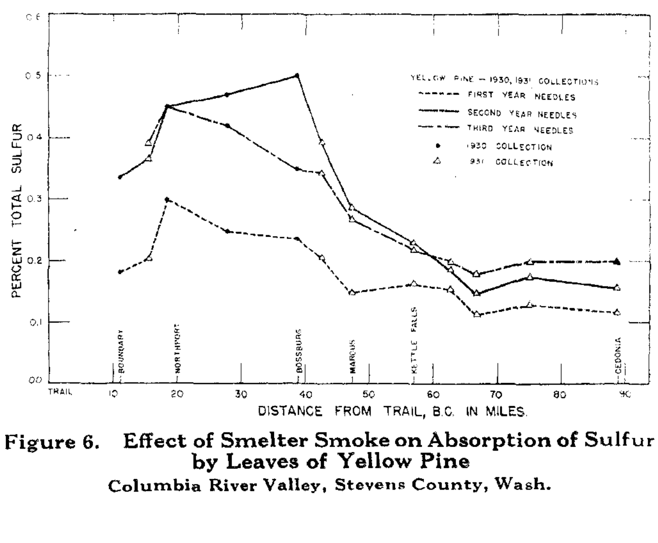

Figure 6. Effect of Smelter Smoke on Absorption of Sulfur by Leaves of Yellow Pine

Columbia River Valley, Stevens County, Wash.

Table V. Absorption of Sulfur Dioxide by Conifers

(Smelter area 15 miles south of Trail, B. C.)

Date of Foliage Collection	Total Sulfur (Dry Basis), %, Age of leaves, years: 1	2	3	4
		Douglas Fir		
June 29, 1933	0.10	0.23	0.39	0.45
Nov. 9, 1933	0.23	0.32	0.48	0.50
Jan. 9, 1934	0.22	0.33	0.45	0.52
March 7, 1934	0.24	0.30	0.42	0.52
		Yellow Pine		
June 29, 1933	0.13	0.25	0.27	0.34
Nov. 9, 1933	0.24	0.31	0.33	..
Jan. 9, 1934	0.26	0.26	0.27	..

plants may derive part of their sulfur requirements by direct absorption of this gas by the leaves as well as by the soil, and recent investigations by Setterstrom, Zimmerman, and Crocker (*84*) and Thomas, Hill *et al.* (*96, 97, 98*) indicate that the yield of sulfur-deficient plants is increased by low concentrations of sulfur dioxide in air.

However, long continued exposure to high concentrations may prove damaging to poorly buffered, virgin soils because of the increase in acidity and gradual loss of calcium and other bases. In rare instances where the accumulation of sulfuric acid may become abnormally high, the normal activity of soil microorganisms may be affected adversely and the decomposition of the humus may be interrupted. The adverse effects of sulfur dioxide on soils, particularly the removal of lime in forest areas in Europe, have been described by Wieler (*111, 117*). Kelley (*49*), however, could find no relation between smoke and soil acidity in the vicinity of Philadelphia where it was claimed that damage to crops was due to factory smoke. Also Rusnov (*76*) found that there was no correlation between the lime content of the soils he investigated and injury to the vegetation growing on them.

Recently, McCool and Mehlich (*58*) examined some soil characteristics in relation to distance from the industrial centers of Philadelphia, Pa., East St. Louis, Ill., and St. Louis, Mo. They concluded that soils near centers of considerable sulfur dioxide production have not been altered, notably, in spite of many years of exposure.

This question was investigated in the Trail region by the analysis of about 600 samples of virgin soils consisting mainly of sands, sandy loams, and silts. The hydrogen-ion concentration, exchangeable hydrogen, base-exchange capacity, and sulfate content of the surface inch and succeeding depths to 36 inches were determined at each location (*39*). Within an area extending 6 to 8 miles north and south of the smelter the soil was markedly low in pH and in the percentage of base saturation, and this was correlated with an abnormally high sulfate content, both in the soil and in the water supplies from the creeks and springs in this region. This smoke effect diminished rapidly with increasing distance and was confined mainly to the surface inch of soil. At points 15 to 20 miles away the soil was substantially normal, and even in the region of 8 to 15 miles distant there was not much effect noted in the top 5-inch profile of soil.

An investigation of the water supplies of the area undertaken to determine whether an appreciable amount of absorption of atmospheric sulfur dioxide was taking place in the water of creeks, springs, and wells of this region was largely negative, with the exception of a restricted area north and south of Trail, B. C. (*40*).

INFLUENCE OF SULFUR DIOXIDE ON GROWTH OF TREES

The classical investigations of Douglass of the Carnegie Institution of Washington (*15*) have portrayed the climatic cycles extending over hundreds of years by means of the annual growth rings of trees. These constitute a permanent record of the influence of environmental conditions such as water supply, soil quality, and the incidence of fire, disease, insect damage, and many other local factors. However, in semiarid regions it is probable that the dominant factor is the amount of available water, including both precipitation and subirrigation.

If, in addition to other factors, both beneficial and detrimental, there is sometimes imposed a polluted atmosphere, the resultant influence on tree growth may be traced provided the toxic agents exert a measurable retardation on the diameter increment. The effect of sulfur dioxide may not be harmful if the concentrations are below the toxic level and the soil is deficient in soluble sulfates.

The most successful studies on the retardation of tree growth by industrial gases have been those which dealt with conifers. Such trees are more likely to show a reduction in diameter increment because the needles remain on the branches for 3 years or longer in contrast to deciduous species. Reuss in 1892 (*73*) investigated the growth of spruce in a forest reserve in Upper Silesia, Germany, where 54 industrial plants, with a total yearly emission of about 35,000 metric tons of sulfur dioxide, were located within a radius of 6 kilometers. It was found that there was no constant relation between the sulfur dioxide content of the atmosphere and the observed retardation of growth; the latter, in large measure, was dependent on the age of the tree and its individual resistance to gas. Gerlach (*18*) also found a considerable degree of retardation in spruce from smoke injury, while Eicke (*16*), in growth studies of pine trees, noted that trees about 25 years of age were retarded to a greater extent than older trees. He described the almost complete recovery of a tree 52 years old which had shown an adverse smoke effect when young.

The damage to conifers from smoke and ash, chiefly sulfur dioxide and zinc sulfate, in the Naugatuck valley of Connecticut, has been estimated by Toumey (*105*) to have amounted to a reduction in growth of 25 to 50%, from measurements of increment borings in 1921. The conifers in this valley were gradually being displaced by the more resistant deciduous species. The early literature on the effect of smoke damage on the growth of conifers, with special reference to sulfur dioxide and smelter fumes, has been reviewed by Haselhoff and Lindau (*24*).

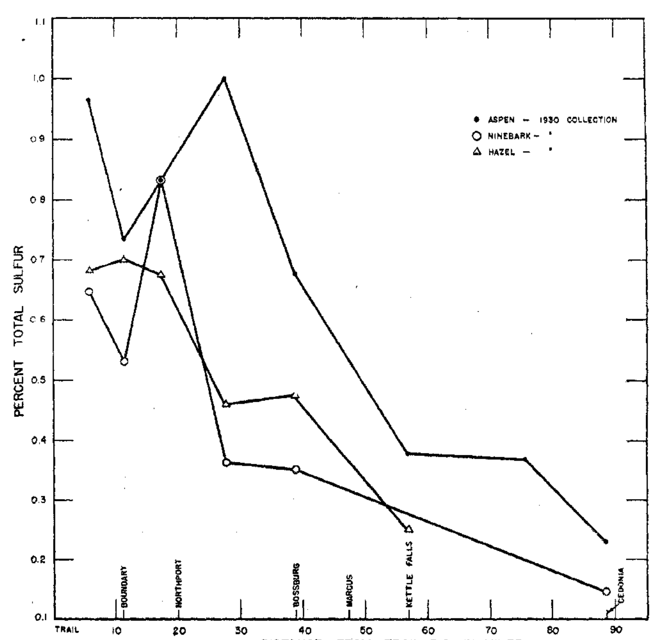

Figure 7. Effect of Smelter Smoke on Absorption of Sulfur by Leaves of Deciduous Species

Columbia River Valley, Stevens County, Wash.

A most comprehensive investigation on the influence of smelter fumes on tree growth was conducted by Lathe and McCallum for the National Research Council of Canada (*53*) in Stevens County, Wash., in order to define, accurately, the areas affected by operations of the smelter at Trail, B. C. Studies were made of the diameter increment of Douglas fir, yellow pine, and lodgepole pine from borings taken from over 10,000 trees in 1929, 1930, 1934, and 1936. The annual rings were measured from about 1915 to the date of sampling, and more than 175,000 measurements were made. There was good correlation between the precipitation from January to August of each year and the diameter increment during the period 1915 to 1936, except in the zone where sulfur dioxide was the factor limiting tree growth.

In the comparison of the growth curves for particular groups of trees, the trend of the curves was more significant than the actual magnitude of the growth year by year. The data for the control groups of trees showed a remarkable correlation with the precipitation for this region. The sulfur dioxide effect was clearly indicated by the failure of the affected trees in the smoke zone to respond to favorable periods of precipitation; the growth rate still showed a declining curve when the rate for the controls was ascending. Another feature of this study was that it disclosed the effects due to the operation of a small smelter at Northport during the period 1916 to 1921. The effect could be traced by the growth of trees both north and south of this town.

The affected trees showed a large deviation in growth trend as compared with the controls during the period of maximum sulfur dioxide emission, within a zone 12 to 15 miles from the Trail smelter, and smaller deviations coincident with the earlier operation of this smelter and also of the one which was located at Northport, Wash., about 20 miles south of Trail. This effect reached a vanishing point at approximately 40 miles from the source of emission. However, following the installation of units for the recovery of sulfur dioxide in 1931 and subsequent years, the yellow pine showed a growth trend rather more favorable than the controls, but the more severely injured Douglas fir still showed incomplete recovery even by 1936.

SYMPTOMS AND DIAGNOSIS OF INJURY

Sulfur dioxide produces characteristic symptoms on the leaves of plants which are generally classified under the broad groupings of acute and chronic injury. Within a given smoke zone, however, there is usually found a wide variety of markings which may be caused by other factors, such as drought, frost, winter injury, insects, disease, and various physiological disorders. Accurate diagnosis of the distribution and degree of injury is, therefore, a difficult matter unless some knowledge is available of the character and intensity of the gas visitations, the meteorological conditions, the sulfur content of the affected plants, and the reaction of susceptible indicator plants. The careful investigator must utilize, as aids in the diagnosis and estimation of the extent of injury, such knowledge as may be made available by a study of the factors enumerated above.

One of the earliest detailed studies of sulfur dioxide symptoms on vegetation was that conducted by Schroeder and Reuss in 1883 (*78*). They illustrated the types of acute markings which were found on conifers, broad-leaved trees, cereals, and grasses. Even in 1883 the question of "invisible injury" was under discussion and these authors were of the opinion that claims based on this theory were inadmissible and contrary to the basic principles of exact science. Chemical, botanical, and microscopic studies on spruce with special reference to invisible injury were described by Sorauer and Ramann (*85*) in 1899. Shortly afterwards, Haselhoff and Lindau (*24*) defined acute symptoms as those causing injury or death to vegetation in one or more fumigations in the same growing season. Chronic damage was considered to be manifested in a continued decrease in annual growth of trees.

The distinction between winter killing and the effects of smelter smoke injury was pointed out by Hedgcock (*28*) in 1912 in the case of conifers in the forests of Montana. He found that not only the leaves were killed in winter injury but often the terminal buds and twigs also, followed by the death of the whole tree in the growing season. Death of trees by sulfur dioxide, however, was found to take place slowly by a gradual defoliation process over a period of several years. The two forms of injury have different orders of susceptibility.

The report of the Selby Smelter Commission (*83*) described in detail the symptoms of acute injury to barley and other plants. It was pointed out that the symptoms of leaf maturity or the usual yellowing and death of the lower leaves on grain and grasses and of the older leaves of trees have often been confused with smoke or sulfur dioxide damage. In 1923 Stoklasa (*86*) classified symptoms into acute, chronic, and invisible. Acute injuries were brought about by high concentrations of sulfur dioxide and involved the rapid destruction of the chlorophyll. Chronic injury involved the more gradual breakdown of the chlorophyll, abnormal photosynthesis, and retardation in growth. Invisible injury was reflected in a lowering of the photosynthetic process and, according to Stoklasa, was always present wherever sulfur dioxide occurred in the atmosphere!

Neger (*64*) was of the opinion that there was no sharp line of demarcation between the forms of acute and chronic injury in trees. The chronic form was considered to predominate in winter fumigations, whereas the acute form was generally produced by the presence of gas during periods of high plant activity in the growing season. Wieler (*110*) considered the dark brown and red color tones of injured leaves to be due to the oxidation of tannins in sunlight. Where no tannin is present in the leaf, the tissue turns white—for example, cereals and alfalfa. Other investigators (*6*, *23*) have stressed the importance of not relying solely on botanical investigations; other factors, including analyses of soil, air, and leaf tissue, must be studied for an accurate diagnosis of injury.

Dorries (*14*) has proposed a spectroscopic method for detecting leaf injuries caused by various acid gases, based on the presence of phaeophytin in injured leaves. It is suggested that the acid gas acts directly on chlorophyll which is transformed into phaeophytin through the splitting off of magnesium. The transformation product is not present in leaves showing autumnal discoloration, drought, frost, or parasitic injuries, or in leaves showing symptoms of chronic sulfur dioxide injury.

Hill and Thomas (*31*) have described the initial stages and subsequent changes in the appearance of alfalfa leaf tissue injured by sulfur dioxide in experimental fumigations. The first symptoms were marked by an increased turgidity of the plant cells followed by the appearance of flaccid areas which bleached out to an ivory color after a short time in sunlight. Zimmerman and Crocker have discussed the differences in symptoms between mono- and dicotyledonous plants (*122*). The leaves of the former could be treated to produce markings varying from slightly mottled tips to completely bleached leaves, whereas in the latter type the intercostal type of marking predominated, the tissue about the veins being comparatively resistant.

A detailed study has been made by the author, Ledingham, and McCallum (*37*) of the various symptoms of injury on forest and crop plants from observations in the Trail smelter area and in experimental fumigations at Summerland, B. C. This has been supplemented by numerous plates of authentic specimens and of many types of discolorations not caused by sulfur dioxide. The various sulfur dioxide symptoms on leaves have been divided into broad classes—acute and chlorotic. The acute type has been subdivided into a number of well-defined categories. The essential difference is that in the acute type the leaf cells are killed, whereas in the chlorotic type only the chlorophyll is affected.

Changes in the internal leaf structure by treatment with high concentrations include a marked shrinkage, especially in the palisade cells, even prior to the appearance of the gross external symptoms. In cases of prolonged treatment with low, nontoxic, concentrations of gas, there were no clear-cut differences in the cell structure between treated and untreated leaves nor was there any evidence of the presence of dead cells.

EXPERIMENTAL STUDIES ON TREES AND CROP PLANTS

Extensive experiments were carried out by the author and his associates (*42–48*) on conifers and sensitive mono- and dicotyledonous plants under measured or controlled conditions to determine the influence of various factors on susceptibility, and the effect of low, medium, and high concentrations on stomatal behavior, rate of growth, and yield, chemical composition, photosynthesis, and respiration. The work was carried out at the Dominion Experimental Station, Summerland, B. C., in a semiarid region devoted to farming and fruit growing.

The experimental plots were 6 feet square and consisted of yellow pine and Douglas fir located in natural stands of reproduction, established plots of eleven species of conifers, and irrigated plots of barley, wheat, and alfalfa. A limited number of 12 × 12 foot plots were also used. The seedlings and young trees were obtained from the forest service nurseries at New Westminister, B. C., and Missoula, Mont., and from natural forest areas in the Summerland region. The crop plants were grown from certified seed.

The plots, plant chambers, and fumigation equipment have been described elsewhere (*41*). The plant chambers were of gastight construction and covered with a transparent celluloid material which allowed free access to light. Measurements were made of the volume of air supplied continuously to the plant chambers, the light intensity, temperature and humidity, soil moisture, and other factors. Precise control of the gas concentrations was maintained by careful metering and mixing of the sulfur dioxide–air mixtures and by continuous measurement of the concentrations at the entrance and exit ends of the chambers by three Thomas automatic recorders.

The rates of carbon dioxide assimilation and respiration of the plants were determined by two additional autometers of the Thomas type adapted to the continuous measurement of carbon dioxide (*93, 101*).

The design of fumigation equipment and technique required for accurate investigations of this type in field plot and greenhouse experiments have been described by Thomas, Hill, and their associates (*31, 99, 101*), who have made the most important contributions, within recent years, to the problem of continuous, automatic analysis of sulfur dioxide, carbon dioxide, and other gases in low concentrations in air. Improvements in design of fumigation equipment in greenhouse investigations have been described by Swain and Johnson (*90*), Setterstrom and Zimmerman (*81*), and more recently by Setterstrom (*80*). The latter has stressed the precautions necessary and the equipment required for precise control in experiments with low concentrations of sulfur dioxide for long periods of time.

ENVIRONMENTAL FACTORS AND SUSCEPTIBILITY

Conifers in Natural Habitat. About 450 trees in 43 groups in a mixed stand of Douglas fir and yellow pine were subjected to treatments designed to throw light on such factors as the effect of specific sulfur dioxide concentrations, relation between extent of absorption and injury, the seasonal variation in susceptibility, and residual effects. The age of the trees ranged from seedlings several years old to trees 25 to 30 years old. The concentrations were varied in the different experiments from 0.25 to 5.00 p.p.m., but one plot was fumigated with a concentration as high as 20 p.p.m. (*42*).

The conifers were found to be much more susceptible in the spring and early summer than in the autumn and winter months (*42, 116, 119*). The susceptibility in this respect parallels the seasonal variation in physiological activity of the leaves. Thus, it is known that in winter the transpiration losses are only 1/55 to 1/250 as great as those in the autumn (*106*). In fact, winter transpiration losses are scarcely greater than those from the defoliated twigs of broad-leaved trees. Similarly, the gaseous exchange of conifers is extremely low in winter; the lowest values for respiration or assimilation in pine and fir are obtained from the end of December to the beginning of February. At certain periods the respiration exceeds the assimilation in winter weather (*121*).

Owing to the comparatively mild winter at Summerland, B. C., there was a marked increase in susceptibility to sulfur dioxide early in March, corresponding to the end of the dormant winter period. Thus, Douglas fir and yellow pine, when fumigated at the end of March with 0.75 p.p.m. for 147 hours, continuously, showed discolorations on about 55% of the foliage but in the early autumn the same concentration could be applied for 334 hours without injury. In March 0.50 p.p.m. for 336 hours produced a trace of injury, but during the latter half of August to the end of September a similar concentration for 1008 hours had no injurious effect. During April and May, the most susceptible period, no adverse effect resulted from 450 hours of continuous treatment with 0.25 p.p.m. The trees were more susceptible in daylight than in darkness during the period of active assimilation (*42, 119*). The Douglas fir was in general more susceptible than the yellow pine.

Transplanted Conifers. Experiments were carried out on eleven species of transplanted conifers. These varied in age from 3 to 10 years. In addition, seedlings of yellow pine and Douglas fir from 1 to 3 years old were set out in flats and also subjected to experimental treatments (*43*).

The transplanted trees were more susceptible to sulfur dioxide than those growing in their natural habitat. Nevertheless, a long treatment with an average concentration of 0.22 p.p.m. for 1656 hours during the period October to December had no effect whatever on Douglas fir, yellow pine, and cedar. During October, 0.46 p.p.m. produced the first symptoms of injury on Engelmann spruce after 530 hours' treatment and on yellow pine, lodgepole pine, and white fir after about 700 hours. The transplanted conifers, like those in natural stands, were found to be extremely resistant to sulfur dioxide during the period of the year when there was a virtual cessation of growth.

The deciduous conifer, larch, was the most susceptible species to sulfur dioxide (*43, 116*), a fact which is in harmony with experience in the Columbia River valley. At the most susceptible time of the year, toward the end of May, slight symptoms were produced on larch foliage by a treatment with 0.30 p.p.m. for 8 hours during daylight at an average relative humidity of 67%. On the other hand, at an average concentration of 0.15 p.p.m., no symptoms were apparent after 70.5 hours. With higher concentrations of gas the first signs of injury appeared after 4.5 hours' treatment with 0.72 p.p.m. at an average relative humidity of 68%. The appearance of the characteristic symptoms on the leaves coincided with wilting of some of the terminal buds. The color of the needles varied in the initial stages from a very pale green to a straw yellow color, but after a few days the familiar reddish discolorations appeared.

Seedlings of Douglas fir and yellow pine were found to be much more susceptible than older trees. Thus slight markings were produced on 1-year-old seedlings by 0.29 p.p.m. for 44.5 hours and 0.78 p.p.m. for 8 hours. An 8-hour treatment with 1.08 p.p.m. resulted in slightly more injury to 1-year-old yellow pine and none on 2-year-old. However, 15 hours at 1.11 p.p.m. killed about 50% of the 1-year-old pine seedlings and about 21% of the 2-year-old seedlings.

Most of the experimental work on conifers, described in the literature, has been performed on transplanted stock grown in greenhouses, garden plots, or under other unnatural conditions. The results of such experiments cannot be applied directly to

Table VI. Absorption of Sulfur Dioxide by Yellow Pine in Fumigation Experiments

	Sulfur Dioxide Treatment		Increase in Total Sulfur over Period of Treatments, %			
			Age of leaves, years			
Plot	Average concn., %	Duration, days	1	2	3	Injury
44	0.22	69.0	0.280	0.260	0.120	None
11	0.50	14.0	0.084	0.074	0.055	None
39	0.50	42.0	0.134	0.047	0.037	None
19	0.75	6.12	0.046	0.047	0.039	Severe
30	5.00	1.67	0.021	0.083	0.107	Severe
12	5.00	5.0	0.069	0.029	0.055	Severe

trees in their native habitat. The latter are much more resistant to sulfur dioxide and this disparity in susceptibility is even greater between native and transplanted seedlings. Earlier investigations also suffer from the limitations of poor technique and apparatus, lack of control of the gas concentrations, and abnormal conditions in the fumigation cabinets (*26, 27, 78, 85, 86, 112*).

Crop Plants. The question frequently arises as to how far conclusions drawn from the results of experimental fumigations under cabinets may be applied to crop plants growing under natural field conditions in a smelter zone. In 1931 an attempt was made to supply an answer to this question by applying experimentally to a crop of barley at Summerland a series of fumigations as nearly as possible identical with those occurring at the Stroh farm plots in the smelter area at the site of a recorder station about 15 miles south of Trail. The sulfur dioxide and meteorological records taken at the Stroh farm were relayed to Summerland every few days and similar conditions of fumigation were then imposed there on a large 12 × 12 foot plot. In order to maintain temperature and humidity factors as nearly comparable as possible, a few fumigations relayed from the Stroh farm were not repeated until some later date when weather conditions at Summerland chanced to be more favorable. It was usual, however, to repeat a fumigation at Summerland about 3 days later than it actually occurred on the Stroh farm plots. The humidity was maintained as closely as possible to that obtaining in the Columbia valley area. There was a difference of only 3 days in the age of the barley at the two locations and the fumigations covered a period of about 40 days (*44*).

A rather remarkable correlation was obtained between the appearance of the first symptoms of injury on the Stroh plot barley and that at Summerland. This occurred after the plants had received approximately 124 hours of sulfur dioxide treatment, in fifteen separate fumigations; the average and maximum concentrations at Summerland were 0.29 p.p.m. and 1.05 p.p.m., respectively. The increase in sulfur content of the leaves on the experimental plot and those in the smoke zone were of the same order. Thus, when the plants were about 37 days old, the first, second, and third leaves of barley at the Stroh farm averaged 1.22% sulfur, whereas at Summerland for the same leaves the percentage was 1.37. The results demonstrated conclusively that the response of crop plants to sulfur dioxide under natural conditions is not essentially different from that found in fumigation experiments under cabinets.

A considerable number of experiments were performed to determine the most important environmental factors which influence the susceptibility of crop plants to sulfur dioxide. In establishing the effects of these factors at least two and in many cases three or more replicate experiments were made under a given set of conditions.

Concentration, Duration, and Absorption Factors. The probability of injury to the leaves of plants and the extent of leaf destruction are governed by the concentration, the time of exposure, and the rate of absorption as influenced by the environmental conditions. The rate of absorption of sulfur may be determined either by chemical analyses of leaves collected immediately before and after exposure under known conditions or, as shown by Thomas (*92*), by continuous gas analysis of the sulfur dioxide–air mixtures before and after contact with vegetation. Thomas and Hill (*100*) have developed absorption-exposure equations for the effects of short fumigations of about 60 to 100 minutes with high concentrations on alfalfa. They are of the form $C = C_0 + k/t$ where C represents the concentration, C_0 a threshold concentration to which plants can be subjected indefinitely without injury under conditions of maximum sensitivity, t the time of exposure, and k a constant related to the rate of absorption. The equation is similar to that proposed by O'Gara (*69, 100*), earlier, and it is implied that for every species of plant there is a limiting concentration which is nontoxic even after prolonged exposure. A relation of the same form would hold, probably, for low concentrations as shown by the following examples of results obtained by the author, Ledingham, and McCallum (*43, 44*).

Thus, with experiments on absorption by conifer leaves, the needles were injured only if there was an appreciable increase in sulfur content over a short time. On the other hand, much larger amounts of sulfur could be accumulated by the leaves, as shown in Table VI, without injurious effects, if the gas were absorbed more slowly over a longer time interval.

With respect to concentration-duration factors, at a relative humidity of 60 to 70% young barley plants showed the first symptoms of chlorosis after 31.5 hours of fumigation at 0.30 p.p.m. With concentrations of 0.50 to 0.60 p.p.m., the first symptoms of injury appeared in 8 hours, with 0.90 p.p.m. in 4 hours, and with 1.20 p.p.m. at 2.5 hours. In virtually saturated air (90 to 100% relative humidity), the first symptoms appeared in 17.3 hours, 4 hours, and 1.75 hours, respectively, at the above concentrations. In intermittent treatments, during a period of 20 days, barley plants were treated with gas for a total duration of 210 hours at an average concentration of 0.258 p.p.m. without injury.

In experiments with alfalfa, extremely low concentrations (0.10 to 0.16 p.p.m.) could be maintained continuously for 500 to 600 hours without causing any markings on the leaves under normal temperature and humidity conditions. With a concentration of 0.20 p.p.m. and an average relative humidity of 58%, there was no injury after 87.5 hours' exposure, whereas under similar conditions the plants showed slight markings after 66.5 hours' treatment with 0.30 p.p.m. Slight markings were produced in 17 hours' exposure to an average concentration of 0.41 p.p.m. at an average humidity of 83%.

Table VII. Absorption of Sulfur Dioxide by Alfalfa and Influence of Soil Mixture

	Sulfur Dioxide Treatment				
Time	Intake, p.p.m.	Exit, p.p.m.	Absorption, %	Temperature, ° F.	Humidity, %
Plants Turgid—Soil Moisture 12.8%					
July 30, 1936					
11:00 A.M.–noon	0.85	0.63	25.9	95	48
Noon–1:30 P.M.	0.86	0.66	23.3	95	38
1:30 P.M.–2:30 P.M.	0.92	0.74	19.6	95	36
2:30 P.M.–3:30 P.M.	1.08	0.90	16.7	95	36
3:30 P.M.–5:30 P.M.	1.17	1.00	14.6	94	66
5:30 P.M.–midnight	1.05	0.96	8.6	78	55
July 31, 1936					
Midnight–3:00 A.M.	0.89	0.83	6.7	61	60
3:00 A.M.–6:00 A.M.	0.84	0.80	4.7	62	63
6:00 A.M.–7:00 A.M.	0.87	0.74	15.0	76	58
7:00 A.M.–7:30 A.M.	1.15	0.96	16.6	88	56
7:30 A.M.–8:30 A.M.	0.84	0.72	14.4	100	51
8:30 A.M.–1:45 P.M.	0.78	0.69	11.6	100	51
Plants at Wilting Point—Soil Moisture 6.2%					
July 30, 1936					
11:00 A.M.–1 noon	0.95	0.84	11.6	90	50
Noon–1:30 P.M.	0.95	0.84	11.6	90	55
1:30 P.M.–2:30 P.M.	0.95	0.84	11.6	91	48
2:30 P.M.–3:30 P.M.	1.03	0.99	3.9	92	45
3:30 P.M.–9:00 P.M.	1.03	0.99	3.9	85	58
9:00 P.M.–midnight	0.96	0.91	5.2	72	55
July 31, 1936					
Midnight–2:00 A.M.	0.84	0.80	4.7	68	60
2:00 A.M.–7:00 A.M.	0.76	0.72	5.2	65	62
7:00 A.M.–7:30 A.M.	0.80	0.78	2.5	90	57
7:30 A.M.–9:00 A.M.	0.97	0.90	7.2	100	53
9:00 A.M.–noon	0.90	0.86	4.4	100	50

Influence of Soil Moisture. This effect was investigated by studies on dry and irrigated plots in which the soil moisture was determined at various soil depths down to 24 inches (*44*). Both barley and alfalfa plants became extremely resistant to sulfur dioxide when the soil moisture approached the wilting point. On the other hand, when the moisture was adequate for growth, variations within wide limits did not appreciably affect the susceptibility. However, even in resistant plants near the wilting point it was possible to produce markings on the leaves by raising the humidity of the dry plants by artificial means. The effects of soil moisture could be correlated with the movement of the stomata of leaves, increased resistance being imparted to the plants by closure of the stomatal openings and the resultant decrease in rate of absorption of gas. This is illustrated by the data in Table VII. Setterstrom and Zimmerman (*83*) present similar conclusions regarding soil moisture.

Relative Humidity. The relative humidity of the atmosphere below 70% is one of the most important external factors in influencing the susceptibility of plants to sulfur dioxide according to Swain (*89*), the Selby report (*33*), and others (*23*). When light and soil moisture are not limiting, high relative humidity favors the opening of the stomata and this results in a high rate of absorption of gas. Furthermore, with the soil moisture favorable for growth, a high relative humidity brings about a higher degree of leaf turgor and under such conditions plants may be quite susceptible even if the light intensity is quite low (*44*). Setterstrom and Zimmerman (*83*) state, however, that differences of 20% were necessary to cause detectable differences in resistance of greenhouse plants above 40% relative humidity.

Age. Young barley was found to be much more susceptible to injury than barley in the flower stage. Susceptibility was high when the young plants were developing leaf tissue and tillering out rapidly. The younger and middle-aged leaves were more susceptible than the older leaves. It was shown by analyses of the sulfur content of leaves before and after treatment that, for equal concentrations of gas, the rate of absorption was far less for old leaves than for young leaves (*44*).

In alfalfa, the highly functional, fully grown leaves were injured before the older or the very young rapidly expanding leaves (*31, 44, 122*). In the week following the cutting of the previous crop, the alfalfa shoots and leaves which developed subsequently were comparatively resistant to gas. At this time a considerable proportion of the carbohydrate requirements was drawn from the roots of the plant. In the advanced flower stage the resistance to sulfur dioxide increased also. The stems, bud, and cotyledons of plants were found to be extremely resistant, and this was also true of the floral portions. However, seedlings which had passed the cotyledon stage became very susceptible. Other investigators have reported similar results in regard to the age factor and its influence on susceptibility (*23, 44, 80, 83*).

Light. Since light intensity plays a very important role in plant activity, it seems reasonable to expect that plants would respond differently to sulfur dioxide, depending on the intensity of the illumination. It is known, too, from observations in industrial areas (*23, 86, 89*) that plants are more susceptible during daylight hours than at night. In the course of studies by the author on barley and alfalfa, experiments were designed to determine this effect as accurately as possible (*44*). Some of the results on alfalfa are given in Table VIII. Little difference was found in the susceptibility of barley during morning, midday, and early afternoon periods with the light intensity between 8000 and 12,000 foot-candles. However, the susceptibility was much greater in the morning than in the late afternoon and the plants were least susceptible at night.

The relative importance of the light factor in relation to the response of plants to fumigation has been assessed differently by various investigators. Some consider it to be one of the major factors (*23, 89*), while others believe it to be of minor importance (*33, 83*). It must be mentioned, however, that most species of plants receive far more light than is necessary under normal conditions of growth. A relatively small decrease in light intensity from an initial level can have no effect, therefore, on the resistance to gas.

Table VIII. Influence of Light on Susceptibility

(Series II, August 27–28, 1935)

Sulfur Dioxide Concn., P.P.M.	Time of Day	Av. Light, Foot-Candles	Absorption of Sulfur Dioxide[a], %	Injured Leaves, %	Marked Leaves, %
4.5	9:00 P.M.–10:00 P.M.	0	9.2	None	None
4.8	3:00 A.M.– 4:00 A.M.	0	9.1	None	Very slight
4.6	4:40 A.M.– 5:40 A.M.	65	11.5	None	Very slight
5.0	5:50 A.M.– 6:50 A.M.	300	15	None	16
4.8	7:30 A.M.– 8:30 A.M.	1500	25	22	42
4.6	8:45 A.M.– 9:45 A.M.	3135	32	31	51
4.9	10:30 A.M.–11:30 A.M.	4940	31	32	51
5.0	1:20 P.M.– 2:20 P.M.	3760	31	44	62
4.8	3:30 P.M.– 4:30 P.M.	1680	22	8	29
4.4	6:20 P.M.– 7:20 P.M.	50	12.5	None	Slight
4.5	9:30 P.M.–10:30 P.M.	0	9.5	None	None

[a] Figures represent percentage difference between intake and exit concentrations of sulfur dioxide; absorption by cabinet and soil at this concentration is about 5%.

Temperature. The toxic effect of sulfur dioxide increases with rising temperature, within certain limits, according to Stoklasa (*86*), and this influence is quite evident in the resistance of conifers at low temperatures. Swain (*89*) has mentioned that the four conditions which must be coincident for damage to occur to vegetation in those smelting districts where the concentrations are not high are a temperature above about 40° F., daylight, atmospheric humidity of 60% or over, and a steady prevailing wind in a given direction. During the main growing season, however, variations in temperature between 65° and 105° F. do not affect the susceptibility to any marked extent (*33, 83*).

The results of investigations of environmental factors indicate that any internal factor which causes a high rate of absorption of sulfur dioxide by leaves, such as an active rate of photosynthesis, opening of the stomata, and a high leaf turgor, will render plants susceptible to injury. The peak of absorption coincides with the period of the day or season of the year when plant activity is at a maximum, and high rates of absorption by leaves are found generally when the soil moisture is adequate for plant requirements, the stomata are fully open, and carbon dioxide assimilation is high. In a continuous fumigation, therefore, the sulfur dioxide absorption by leaves closely parallels the movement of the stomata and the high and low levels of absorption correspond to the peaks of photosynthesis and respiration, respectively (Table VII).

EFFECT OF SULFUR DIOXIDE ON CROP YIELD AND CHEMICAL COMPOSITION OF PLANTS

Although numerous investigators have stressed the injurious effects of sulfur dioxide on vegetation, their work has dealt chiefly with high concentrations of gas. Until recently, however, there has been a noticeable lack of data on the effect of concentrations lower than about 1.0 p.p.m., particularly below the level required to cause visible symptoms. As earlier work was confined mainly to field observations and experiments with concentrations which would be bound to prove destructive to plants, the belief arose that this gas is harmful in any concentration no matter how dilute. This led to the theory of invisible injury. Such views will be found principally in the works of Wieler (*112–114*) and Stoklasa (*86*). A summary of these earlier investigations was given by Bredemann and Haselhoff (*23*) and a discussion of invisible damage with arguments based on field observations has also been presented by Janson (*34*).

The author and his collaborators (*46, 47*) have studied, under natural field conditions, the effects of low, medium, and high con-

Table IX. Effect on Yield—Barley

(Invisible injury series)

No. Plots	Sulfur Dioxide Treatment: Concn., p.p.m. Min.	Max.	Av.	Duration, hr. Min.	Max.	Av.	Injured Leaves[a], Av. %	Mean Yield of Crop, % of Adjacent Controls: Plant Weights Total	Straw	Grain	Tiller Weights Total	Straw	Grain
5	0.26	0.35	0.302	15.0	210.0	70.0	0	105.2	102.8	111.0	100.0	98.0	102.8
					Standard deviation of mean			2.31	1.83	5.76	2.70	2.94	3.62
7	0.50	0.75	0.65	2.25	30.0	14.4	1	101.4	102.6	101.6	99.0	98.6	97.7
					Standard deviation of mean			1.36	2.11	1.35	1.57	1.13	2.48
12	0.78	1.00	0.924	3.2	37.5	14.15	4.3	101.6	98.8	108.0	100.2	97.8	107.7
					Standard deviation of mean			2.82	3.64	3.18	2.38	2.48	4.71
7	1.20	1.50	1.331	2.25	23.5	10.30	3.0	102.3	101.3	104.4	100.9	100.0	102.6
					Standard deviation of mean			1.66	2.07	3.18	4.07	3.63	5.40
29	..	..	0.936	..	..	22.7	1.4	102.0	100.7	105.5	99.6	98.0	102.4
					Standard deviation of mean			1.31	1.65	1.85	1.38	1.33	2.36

[a] Average leaf destruction less than 5%.

Table X. Effect on Yield—Alfalfa

(Invisible injury series)

No. Fumigated Plots	No. Control Plots	Sulfur Dioxide Treatment: Concn., p.p.m. Min.	Max.	Av.	Duration, hr. Min	Max.	Av.	Injured Leaves, Av. %	Mean Yield of Crop, % of Adjacent Controls: Green Weight Mean	Standard deviation	Standard deviation of mean	Dry Weight Mean	Standard deviation	Standard deviation of mean
							No Visible Markings							
12	12	0.10	0.30	0.167	66.5	602.0	256.2	0	100.4	7.96	2.29	103.0	7.51	2.16
11	11	0.36	0.66	0.554	4.0	16.5	8.29	0	104.8	8.10	2.44	99.6	6.92	2.08
12	12	0.76	1.15	1.008	4.0	17.0	7.60	0	101.1	2.94	0.85	100.8	7.52	2.17
29	29	0.26	2.51	0.885	4.0	17.0	7.48	0	102.6	6.43	1.19	100.2	6.96	1.29
							Alfalfa Visibly Injured by Treatment							
13	13	0.82	1.37	1.08	3.5	17.0	7.45	14.7	92.2	4.43	1.23	94.4	6.03	1.67
5	5	1.91	2.49	2.27	4.0	7.5	5.54	18.2	91.4	5.0	2.23	91.8	4.28	1.91
3	3	1.40	1.48	1.45	6.5	7.0	6.67	12.0	82.0	3.70	2.13	89.0	3.96	2.28
7	7	0.35	0.55	0.461	44.0	97.5	66.14	11.3	87.9	6.18	2.34	88.6	5.85	2.21
12	12	0.78	1.25	0.992	21.0	60.0	33.0	21.3	84.7	7.70	2.22	87.0	8.65	2.50
4	4	2.00	2.46	2.21	13.0	14.5	13.87	25.7	83.9	3.30	1.65	87.2	3.24	1.62
3	3	1.40	1.77	1.56	18.2	38.5	28.08	27.0	78.9	8.66	5.00	76.8	5.35	3.08

centrations on the growth of barley (Hannchon) and alfalfa (*M. sativa*, Grimm) with special reference to the determination of invisible injury, if such injury does, in fact, exist. The yield studies were made on 6 × 6 foot plots at various stages of growth, the weights being taken at maturity. Some of the experimental treatments consisted of single fumigations only, whereas in others the plants were treated repeatedly or continuously for long periods. A number of plots of alfalfa were treated continuously with low concentrations throughout the growth of a crop.

The fumigated and control plots were selected at the time of each experiment, the control in most cases being situated adjacent to the fumigated plot and covered with a cabinet for the same length of time. Consideration was given to the possible effect of shadows cast by the cabinet or equipment on one plot or the other, and at times it was found advisable to have the control plot in an adjoining row in order to eliminate such effects. Because certain areas of the experimental field had to be left bare to provide sufficient space for the fumigation equipment and automatic sulfur dioxide and carbon dioxide analyzers, a completely random arrangement of control and treated plots was not possible. Differences which developed after treatment between fumigated and control plots could be reasonably attributed to the effect of fumigation. However, in the analysis of the harvest data conclusions have been based only on the results of as many experiments as possible within a series and not on single experiments. The average yield for each series of experiments, the deviations from this average, and the standard deviation of the mean were calculated as a percentage of the controls.

Barley. In treatments under various conditions of exposure over a relatively wide range of concentrations in which the fumigations were not carried beyond the point where the first symptoms of incipient injury appeared on the leaves or where the concentrations were too low to cause visible markings, it was found, as shown in Table IX, that there was no effect on the yield with fumigations which caused less than 5% of leaf destruction. This table summarizes the results on 60 fumigated barley plots; some of the treatments given these plots were 0.26 p.p.m. for 210 hours, 0.30 p.p.m. for 72.5 hours, 0.80 p.p.m. for 22 hours, and 1.20 p.p.m. for 15.4 hours.

With injurious fumigations involving concentrations in the range of 0.78 to 2.00 p.p.m., where an average leaf injury of 20.3% was produced in a series of 17 plots, the average weight of straw was 92.0% and the average weight of grain 95.5%, with standard errors of 2.7 and 2.8%, respectively. The treated plants showed a marked ability to recover from the effects of sulfur dioxide. Injurious fumigations during the flowering period of barley caused a reduction in the yield of grain but not in that of the straw. During the flower-and-milk stage the leaves show a considerable decrease in normal physiological activity with a decline in the rate of vegetative growth. A reduction in the weight of straw occurred only in severe exposures where the damaged leaves were shed after treatment.

Alfalfa. The question of a possible decrease in yield or subliminal effect after treatment with low concentrations was studied even more intensively with alfalfa as the plant material. Alfalfa is more sensitive to sulfur dioxide than barley although both plants are among the most susceptible known species. However, after the primary leaves have unfolded, alfalfa shows practically uniform susceptibility throughout its entire growth period.

A considerable number of alfalfa plots were treated with low concentrations for long periods in the range of 0.10 to 0.30 p.p.m. and for shorter periods with higher concentrations; the experiments were stopped before a measurable amount of leaf destruction had occurred. In the case of the lowest concentrations, however, the treatment was continued throughout the life of a crop. The mean yields are shown in Table X for 64 treated plots where there was no appreciable injury to the leaves and no reduction in yield. Among the treatments in this series were an average concentration of 0.10 p.p.m. for 504 hours, 0.11 p.p.m. for 602 hours, 0.14 p.p.m. for 525 hours, and 0.16 p.p.m. for 562 hours, also 0.20 p.p.m. for 88 hours and 0.30 p.p.m. for 66.5 hours. In another invisible injury series, where plots were given shorter treatments with concentrations ranging from 0.26 p.p.m. to 2.51 p.p.m., without appreciable leaf injury resulting therefrom, there was also no effect on the yield. A reduction in yield was evident only in fumigations causing appreciable amounts of leaf destruction.

Among other factors investigated were the effects of gas conditions on the subsequent untreated crops and also the influence on the height growth of plants. It was concluded that with concentrations in the range of 0.10 to 0.30 p.p.m. there was little likelihood of any effect on the yield under normal growing conditions

Table XI. Chemical Composition

(Comparative average analyses of barley grain from fumigated and control plots at maturity)

No. of Experiments	Averages for Fumigated Barley Plots: Sulfur Dioxide Treatment, Concn., p.p.m.	Sulfur Dioxide Treatment, Duration, hr.	Age of plants at start of treatment, days	Leaf injury, %	Nitrogen, %	Reducing sugars and sucrose, %	Starch[a], %	Averages for Control Barley Plots: Nitrogen, %	Reducing sugars and sucrose, %	Starch[a], %
Series I[b]										
3	0.276	96.9	24–40	0	2.06	2.12	61.3	2.24	2.20	62.0
6	0.517	10.5	26–32	2	2.48	2.22	62.3	2.36	2.47	62.5
8	0.965	14.5	51–57	2	2.05	2.24	60.3	2.03	2.37	60.9
3	1.04	7.4	24–39	5	2.21	1.97	63.9	2.23	2.29	64.0
Series II[b]										
6	1.34	15.6	20–37	24	2.61	2.36	60.5	2.46	2.34	62.6
4	3.25	16.2	54–56	16	2.37	2.62	59.7	2.39	2.73	62.4
2	2.24 5.07	3.0 3.5	19 41	16 70	2.04	2.44	51.9	2.31	2.41	58.8

[a] Acid-hydrolyzable polysaccharides.
[b] Mean difference in starch content between control and fumigated plots in Series II = 3.1%, $t = 2.41$, $n = 22$, P approximately equal to 0.02; correlation between leaf injury and reduction in starch content of grain (Series I and II), $r = +0.838$, P less than 0.02.

Table XII. Composition of Alfalfa Leaves Treated with Low Concentrations of Sulfur Dioxide

Plot	Sulfur Dioxide Treatment	Leaf Injury, %	Moisture, %	Total Sulfur, %	Organic and Ammoniacal Nitrogen, %	Reducing Sugars, %	Sucrose, %	Acid-Hydrolyzable Polysaccharides, %	Acid-Hydrolyzable Polysaccharides Plus Sugars, %
I-11	0.10 p.p.m., 196 hr.	0	83.5	1.13	5.11	0.62	2.30	6.93	9.85
I-10	Control cabinet	...	83.4	0.71	5.04	0.65	1.99	6.87	9.51
C-11	0.10 p.p.m., 504 hr.	0	81.2	1.28	5.20	1.00	2.16	7.24	10.40
C-12	Control cabinet	...	80.9	0.60	5.07	0.71	2.25	8.87	11.83
G-12	0.11 p.p.m., 602 hr.	0	81.6	1.77	4.84	0.77	1.83	7.75	10.35
G-13	Control cabinet	...	83.9	0.65	5.03	0.55	1.40	6.53	8.48
G-8	0.12 p.p.m., 240 hr.	Slight	81.5	1.48	5.13	1.52	2.91	8.49	12.92
G-7	Control cabinet	...	80.7	0.71	5.38	1.00	2.31	9.54	12.85
G-16	0.16 p.p.m., 562.5 hr.	0	83.0	1.89	4.98	0.80	1.26	6.57	8.63
G-15	Control cabinet	...	82.7	0.81	4.97	0.44	0.86	7.78	9.08
A-5	0.17 p.p.m., 112.5 hr.	Slight	80.0	0.96	5.30	2.27	2.83	6.87	11.97
A-4	Control cabinet	...	80.2	0.61	5.17	1.55	2.62	8.13	12.30
A-3	0.18 p.p.m., 136 hr.	Slight	80.4	1.17	5.11	1.54	3.18	8.13	12.85
A-4	Control cabinet	...	79.8	0.68	5.17	0.96	2.90	7.17	11.03
H-5	0.24 p.p.m., 160 hr.	Slight	83.3	1.46	4.97	1.56	2.89	5.34	9.79
H-4	Control cabinet	...	82.1	0.70	5.20	...	...	4.98	...
C-17	0.37 p.p.m., 378 hr.	8	79.2	1.71	4.67	3.77	3.70	8.01	15.48
D-16	Control cabinet	...	79.3	0.60	4.89	2.44	4.28	8.74	15.46
A-1	0.40 p.p.m., 171 hr.	8	78.6	1.90	5.17	0.92	2.26	6.18	9.36
A-2	Control cabinet	...	76.6	0.70	4.82	0.79	2.07	9.96	12.82
Average of fumigated leaves			81.23	1.475	5.048				11.16
Average of control leaves			80.96	0.677	5.074				11.48

even when, after prolonged exposure, slight markings might be produced occasionally. No evidence was obtained to indicate the existence of any invisible effect on the yield.

Chemical Composition. The yield studies were supplemented by analyses of the sulfur, protein, sugars, and acid-hydrolyzable polysaccharides of leaves and grain (*47*). Where the extent of leaf injury to barley was less than 5%, no significant differences were observed in the protein and carbohydrate content of the grain, as shown in Table XI. Fumigations with more injurious concentrations of sulfur dioxide resulted in a significant decrease in acid-hydrolyzable polysaccharides, but no consistent trend in protein content was evident.

The chemical data for a series of experiments on alfalfa with average concentrations between 0.10 and 0.40 p.p.m., where the leaf injury was negligible, indicated no significant differences in either moisture content, protein content, or total sugars plus starch in the leaves of the fumigated plots in comparison with the controls (Table XII). However, leaves damaged severely by treatment with high concentrations of gas showed a considerable decrease in moisture content, sucrose and starch, a decrease in alcohol-soluble nitrogen, and an increase in insoluble nitrogen. The apparently normal fumigated leaves, in this case, were not appreciably different from the controls in regard to the moisture content and the nitrogen fractions but showed a considerable decrease in the amount of polysaccharides (*47*).

The earliest quantitative studies on the growth of plants under the influence of sulfur dioxide were performed by the Selby Smelter Commission (*33*) who developed a technique for fumigating plants which, with certain modifications, has since become recognized as the proper approach to this problem. They concluded from extensive field plot studies on barley, involving short treatments with high concentrations, repeated frequently, that there was no reduction in yield when the leaves were not injured appreciably. A decrease in yield was always correlated with a definite amount of leaf destruction. O'Gara performed similar experiments with alfalfa (*68*) in 1915; these indicated that there was no significant effect on yield from a considerable number of short fumigations as long as the treatments did not produce any more than traces of markings. Subsequently Hill and Thomas (*31*) showed, in an extensive study of the influence of leaf destruction by high concentrations on the yield of alfalfa, that the reduction in yield is proportional to the percentage of leaf area destroyed, that this decrease could be duplicated by clipping off from normal plants an equivalent leaf area, and that the gas must produce visible effects to decrease the yield.

Swain and Johnson (*90*) investigated the effect of exposures of 3 to 6 hours daily for 28 days, with low nonmarking concentrations, on the development of wheat plants grown in nutrient solutions under almost ideal conditions of light, humidity, and nutrient supply. There was no sign of any injurious action on the rate of growth or yield. The average concentrations varied, in the different experiments, from 0.18 to 0.36 p.p.m. Setterstrom, Zimmerman, and Crocker (*84*) also studied the effect of low, nontoxic concentrations on the yield of alfalfa grown in sand and watered with nutrient solutions in a greenhouse. Water supply, nutrients and sulfur content of nutrient supply, and age of plants were varied systematically, and the treated plants were grown for periods up to 25 days in a sulfur dioxide atmosphere containing

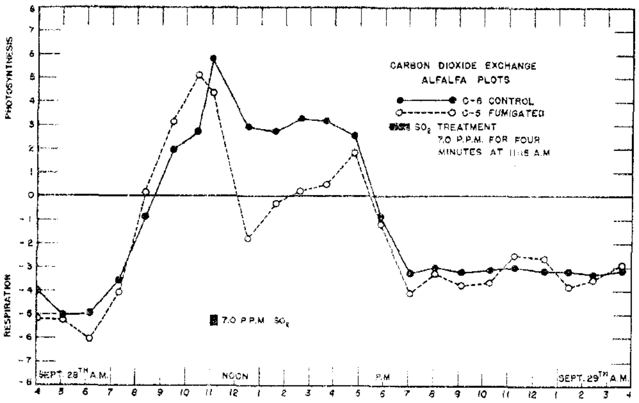

Figure 8. Inhibition of Photosynthesis in Alfalfa

Extremely short exposure to 7.0 p.p.m. of sulfur dioxide and subsequent recovery from this treatment which caused no leaf destruction

0.10 or 0.20 p.p.m. The yield was not decreased under any of the conditions employed. But under some conditions there was a significant increase in yield. Deficiencies in sulfur content of the nutrient supply could be made up by absorption of sulfur dioxide from the air. Plants grown with a deficient water supply responded more readily to stimulation by sulfur dioxide than did those grown with an ample supply of water. The treatments had no significant effect on the nitrogen content of the alfalfa.

Quite recently Thomas *et al.* (*97, 99*) have reported the results of extensive investigations on plants grown in large sand-culture beds in air-conditioned greenhouses. They found that prolonged treatment with 0.10 p.p.m. improved the yield of sulfur-deficient plots of alfalfa, whereas the yield of treated plots supplied adequately with sulfate was unaffected. The absorbed sulfur dioxide was changed principally to sulfate, but a small amount of organic, labile sulfur was produced (*98*). The distribution of the sulfur in cereals was followed by tracer experiments utilizing S^{35} and radioautographs (*21, 96*). A high initial absorption of sulfur dioxide by the leaves was followed by a lowering of the concentration as the sulfur was distributed throughout the plant. During ripening a considerable portion of the sulfur was translocated to the grain, with conversion of most of the sulfur dioxide to organic forms. The distribution of the sulfur in the plant, after sulfur dioxide treatment, was not different, noticeably, from the distribution in untreated plants.

Finally, in direct feeding experiments with animals, Widtsoe (*109*), many years ago, found that there was no effect on the relative milk production, weight of butter fat, and weight of animals, when cows were fed alfalfa hay exposed to sulfur dioxide from smelters in the vicinity of Murray, Utah. This was confirmed in more recent work by Cunningham and Addington (*9*) who used alfalfa in a field about 3 miles from a smelter where the atmospheric sulfur dioxide conditions were known and the leaflets had been marked on four occasions during the growth of the crop.

INFLUENCE OF SULFUR DIOXIDE ON STOMATAL BEHAVIOR, PHOTOSYNTHESIS, AND RESPIRATION

The absorption of energy from sunlight by chlorophyll and the assimilation of carbon dioxide from air are the fundamental processes for the synthesis of organic matter by autotrophic plants. The energy thereby stored in the plant is released during respiration for the production of other complex substances. Therefore, a study of the rate of carbon dioxide assimilation and respiration is of the utmost importance in the evaluation of the effect on vegetation of concentrations of sulfur dioxide usually found in the atmosphere.

Some indication of the specific action of sulfur dioxide in high concentrations on the photosynthetic mechanism of plants may be gleaned from a consideration of their behavior in the presence and absence of light. In water plants—for example, *Elodea canadensis*, which absorbs sulfur dioxide through the epidermis—much higher concentrations are required for injury in darkness than in light (*74*). Plant organisms which do not contain chlorophyll—for example, flower parts—are notably more resistant to sulfur dioxide. This has also been shown to be true of buds and cotyledons and very young leaves in which the chlorophyll has not been fully elaborated. In variegated leaves the white portions may remain alive after sulfur dioxide treatment which has injured the green parts (*65, 67*). Therefore, sulfur dioxide in sufficient concentration is capable of acting specifically on the chlorophyll when carbon dioxide assimilation is in evidence.

In view of the fact that measurements of the rate of carbon dioxide exchange would provide the most sensitive means of determining the invisible effect, if any, on plants in the presence of sulfur dioxide, a study of this problem was undertaken by the author and his associates (*41, 45, 48*) at Summerland, B. C. The relative rates of carbon dioxide assimilation and respiration of similar plants in test and control cabinets were measured continuously by analytical autometers designed by Thomas (*101*). Experiments were made with high concentrations of short duration and with lower concentrations applied intermittently or continuously during the growth of a crop of alfalfa, the gas conditions in many of these experiments were such that no visible injury was apparent on the plant.

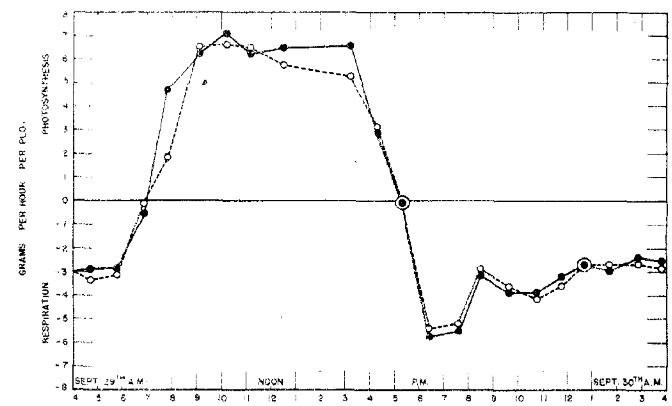

Figure 9. Carbon Dioxide Assimilation and Respiration of Alfalfa Treated with 7.0 P.P.M. for 4 Minutes on Previous Day

Since leaf stomata are known to play a major role in the gaseous exchange in plants (*7*), it seemed reasonable to assume that their behavior would also be a factor in relation to this problem. There is little, if any, information available concerning the specific action on stomatal behavior of sulfur dioxide at various concentrations and times of exposure. Zimmerman and Crocker (*122*) have postulated their probable relation to the decrease in susceptibility shown by plants growing in the dark or suffering from lack of water. Therefore, studies were made of the stomatal behavior of alfalfa by three different methods: direct microscopic measurements; the absolute-alcohol-strip method (*54*); and by use of automatic porometers (*45, 50*) attached to living plants.

High Concentrations. The influence of high concentrations, above 1.0 p.p.m., was immediately detectable in a lowering of the rate of photosynthesis. Two experiments with concentrations of 7.00 p.p.m. and 1.25 p.p.m. of 4 and 30 minutes, respectively, where no visible injury appeared on the leaf tissue, are typical of the effect observed. With 7 p.p.m. the photosynthesis was inhibited and for a few hours respiration predominated. During the remaining hours of daylight the treated plants gradually recovered their normal rate of assimilation as shown in Figure 8.

Table XIII. Effect of Sulfur Dioxide on Carbon Dioxide Exchange of Alfalfa

(Intermittent treatments for 25.4 hours at 0.31 to 0.38 p.p.m.)

1936	Sulfur Dioxide Treatment[a] Duration, hr.	Av. concn., p.p.m.	Max. concn., p.p.m.	CO_2 Assimilated Daily, Grams: Fumigated plot E-13	Control plot F-14	CO_2 Respired Nightly, Grams: Fumigated plot E-13	Control plot F-14
Aug. 22	...	...	...	17.85	15.20	1.50	2.60
23	...	...	...	36.05	35.30	16.50	14.90
24	2.0	0.32	0.37	43.75	44.45	18.20	15.50
25	2.0	0.34	0.36	65.00	67.25	12.00	14.30
26	2.1	0.31	0.39	44.90	51.70	35.30	34.25
27	11.0	0.38	0.43	53.40	46.20	32.10	20.50
28	6.3	0.36	0.38	41.60	35.60	30.40	30.40
29	...	...	...	36.45	35.75	18.70	23.90
30	2.0	0.33	0.35	43.00	50.90	19.10	20.10
31	...	...	...	52.30	49.50	...	...
Total assimilation and respiration, Aug. 22 to 31				434.30	431.85	183.80	176.45
Net assimilation, Aug. 22 to 31				250.50	255.40	...	...

[a] Treatment Aug. 24 from 9:45 to 11:45 A.M.; Aug. 25 from 9:30 to 11:30 A.M.; Aug. 26 from 10:40 A.M. to 12:50 P.M.; Aug. 27 from 9:15 P.M. to 8:00 A.M., Aug. 28; Aug. 28 from 4:00 to 10:30 P.M.; Aug. 30 from 9:15 to 11:15 A.M.

The levels of photosynthesis and respiration on the following day are shown in Figure 9. In the short fumigation with 1.25 p.p.m., an inhibition of assimilation was again noticeable during the presence of the gas, but subsequently there was a rapid recovery from this effect. In other experiments with highly injurious concentrations in which a considerable amount of leaf destruction ensued there was also a marked reduction in the apparent carbon dioxide assimilation followed by an increase in the night respiration.

Observations on stomatal movement were made on both injured and uninjured leaves during and after exposure to concentrations of about 1.0 p.p.m. or higher. It was found that irrespective of whether or not injury had occurred on the leaves, a much higher proportion of the stomata was closed or partially closed on leaves from the treated plants than on those taken from the controls. But a few hours after removal of the gas the closed stomata on unaffected leaves began to recover their normal movement. The time required for recovery of normal stomatal behavior and photosynthesis from the narcotic influence of sulfur dioxide in undamaged leaves was almost proportional to the concentration in the range above about 1 p.p.m.

Medium Concentrations. With decreasing concentration of gas, the inhibiting effect on photosynthesis became smaller and, finally, in the concentration region of 0.44 to 0.50 p.p.m., it reached the vanishing point. Since no injury, either to the chlorophyll or the protoplasm of the plant cells, was produced in these experiments, this action on the chlorophyll mechanism is probably equivalent to the effect produced by a sudden reduction in light intensity.

In order to confirm the absence of any detectable effect on photosynthesis in the region of 0.40 p.p.m., a series of experiments was carried out with concentrations in the range of 0.30 to 0.40 p.p.m., the maximum not exceeding 0.43 p.p.m. The plants were given six treatments with gas, four being carried out in full sunlight and two under conditions of low light intensity. The total duration of these treatments was 25.4 hours. No appreciable effect was detected on either photosynthesis or night respiration, as shown in Table XIII.

Only a slight reduction in the number of fully open stomata was observed during treatment with 0.89 p.p.m. in daylight and none when the concentration was reduced to 0.76 p.p.m. Continuous fumigation at an average concentration of 0.40 p.p.m. had no influence on the march of the stomata until acute and chlorotic symptoms appeared on the leaves.

Low Concentrations. With low concentrations of gas, there was no effect on photosynthesis or respiration even though the gas was present continuously throughout the growth of a crop of alfalfa. The rates of carbon dioxide exchange during a long treatment of 504 hours, under conditions of comparatively high humidity and high gas absorption, are given in Table XIV. The temperature, humidity, and light intensity, as well as photosynthesis and respiration, were recorded continuously throughout the period of the test. No acute symptoms of injury appeared at any time and the number of chlorotic and senescent leaves toward the end of the experiment, when the plants had reached maturity, was not any greater in the fumigated than in the control plot. The net assimilation for the treated plants was 101.5% of that of the control. Other experiments confirmed the fact that low concentrations of sulfur dioxide have no effect whatever on photosynthesis or stomatal behavior if there is no visible leaf destruction.

In fact, in a narrow range of concentration, between 0.10 and 0.20 p.p.m., some stimulation of photosynthetic activity was detected. Thus, in a continuous exposure to 0.17 p.p.m. for 66 hours, it was found that although prior to this treatment the total apparent assimilation of the plants was 99.3% of the control plants, during the sulfur dioxide treatment the rate of assimilation increased to 108% of that of the controls.

The earlier literature on the influence of sulfur dioxide on photosynthesis and respiration has been reviewed by Bredemann (*23*), but in view of the abnormally high concentrations dealt with in most of that work, there is no point in mentioning it here. However, Noack (*67*) in experiments on the water moss (Fontinalis) found a decrease in carbon dioxide assimilation under constant light intensity after a treatment of 24 hours in darkness with 5×10^{-4}% sodium bisulfite solution. Wehner (*107*), on the other hand, has demonstrated a stimulation of the assimilation of Fontinalis and some land plants in sulfurous acid solutions in a dilution of 5×10^{-5} to 10^{-6}%. More dilute concentrations were without effect. Noack has also considered the possibility of a stimulating effect by sulfur dioxide in sufficiently dilute concentrations (*66*).

In 1930 Heilbronn (*29*) investigated the influence of industrial air on assimilation by plants in two plant chambers maintained under equal concentrations of carbon dioxide. In one chamber the plants were treated with a normal industrial atmosphere, while in the other similar plants were subjected to purified air. He found evidence of greater and more rapid growth in the chamber containing industrial air. However, these experiments have been criticized by Bredemann (*23*) on various grounds.

Table XIV. Effect of Sulfur Dioxide on Carbon Dioxide Exchange of Alfalfa

(Continuous treatment for 504 hours)

1935	Sulfur Dioxide Treatment: Av. concn., p.p.m.	Max. concn., p.p.m.	CO_2 Assimilated Daily, Grams: Fumigated plot C-11	Control plot C-12	CO_2 Respired Nightly, Grams: Fumigated plot C-11	Control plot C-12
Aug. 13			31.9	29.1	15.5	20.7
14	0.107	0.19	61.7	65.9	17.8	11.4
15	0.151	0.20	66.2	72.9	33.4	42.1
16	0.081	0.14	62.3	63.3	31.7	34.8
17	0.074	0.15	47.2	47.9	14.1	20.3
18	0.065	0.07	66.9	64.3	9.1	8.4
19	0.088	0.11	68.0	72.0	...	...
20	0.084	0.12	63.5	63.3	11.6	8.6
21	0.072	0.12	75.5	75.1	6.8	12.5
22	0.051	0.08	68.6	72.6	8.2	10.0
23	0.056	0.08	32.4	37.4	31.9	29.4
24	0.082	0.16	28.1	38.2	46.4	40.8
25	0.075	0.10	10.4	14.8	30.4	31.6
26	0.067	0.10	29.8	31.6	32.7	32.5
27	0.118	0.21	24.2	23.6	20.8	29.4
28	0.074	0.19	20.3	19.4	36.3	42.6
29	0.046	0.11	56.9	62.6	20.0	20.1
30	0.052	0.14	27.3	23.4	35.6	40.6
31	0.098	0.12	48.1	51.9	32.7	35.3
Sept. 1	0.213	0.44	49.9	56.0	22.6	33.3
2	0.154	0.44	46.5	41.5	25.9	35.7
3	0.099	0.17	52.1	58.2	32.4	33.1
4	0.063	0.09	49.5	52.0	29.1	29.1
Total assimilation and respiration, Aug. 13–Sept. 4			1087.3	1137.0	545.0	602.3
Net assimilation, Aug. 13–Sept. 4			542.3	534.7	...	...

A notable contribution to this subject was made in 1937 by Thomas and Hill (*102*), who had originated the best technique for the continuous measurement of photosynthesis and respiration of plants under natural environmental conditions (*101*). Their results were in substantial accord with those obtained by the author and his associates at Summerland, B. C.; both investigations were conducted at about the same time. They found that short fumigations with high concentrations of sulfur dioxide, which were discontinued before an appreciable amount of leaf destruction was produced, caused a large reduction in the rate of photosynthesis of alfalfa, but a recovery of photosynthesis was evident within an hour after treatment. The effects of such fumigations could be observed in a somewhat lowered rate of apparent assimilation for about 2 days after the treatment. Thereafter the rate became normal or greater than normal. A number of short fumigations, with concentrations ranging from 0.70 to 1.26 p.p.m., each caused a definite decrease in the photosynthetic level. Immediately after the fumigations were discontinued, however, the activity rose to a normal or greater than normal level. The decreased assimilation that had occurred during fumigation was largely counterbalanced by increased assimilation afterwards. There was no effect during continuous treatments with 0.24 p.p.m. for about 3 days, with 0.19 p.p.m. for 11 days, and with 0.14 p.p.m. for at least 39 days. A slight stimulation of apparent assimilation was noted at the low concentrations.

CONCLUSION

The recent studies on photosynthesis and respiration of plants in the presence of sulfur dioxide indicate in detail the effect of a wide range of concentration on alfalfa. The behavior of this species may be taken as typical of the reaction of the plants most susceptible to this gas. In none of these investigations with low concentrations, in the absence of visible injury, did the treatment decrease the yield, photosynthesis, or respiration, or interfere with the march of the stomata, or deteriorate the chemical composition, protoplasm, or other plant structure. A reduction in yield or rate of growth was correlated with a definite amount of leaf destruction or loss of foliage in fumigations of sufficient duration at concentrations above 0.25 p.p.m.

It is hoped that the "invisible injury" theory has now been disposed of, once and for all time, and will not be resurrected again in problems involving sulfur dioxide damage to plant life.

From the results discussed in this paper it is evident that sulfur dioxide concentrations up to approximately 0.30 p.p.m. may be present in the atmosphere of agricultural areas during the growing season and not prove detrimental to vegetation as long as the exposures, under existing environmental and internal plant factors, are not prolonged sufficiently to induce appreciable leaf injury at these concentrations. In fact, within the range of 0.10 to 0.20 p.p.m. such low concentrations may actually exert a beneficial effect, especially on sulfur-deficient plants. Even when the concentrations are somewhat higher than 0.50 p.p.m., the effect on photosynthesis is only temporary and very much like that of a passing cloud causing a sudden drop in light intensity, unless the exposure is long enough to injure the leaf tissue. At high levels of gas concentration the absorption of gas by the leaves proceeds so rapidly that injurious effects on the tissue are soon noticeable, particularly when the stomata are open and soil moisture, light, and humidity are conducive to a high rate of absorption. Concentrations in excess of 0.50 p.p.m. should be limited to periods of darkness, low humidity, drought, and low temperatures.

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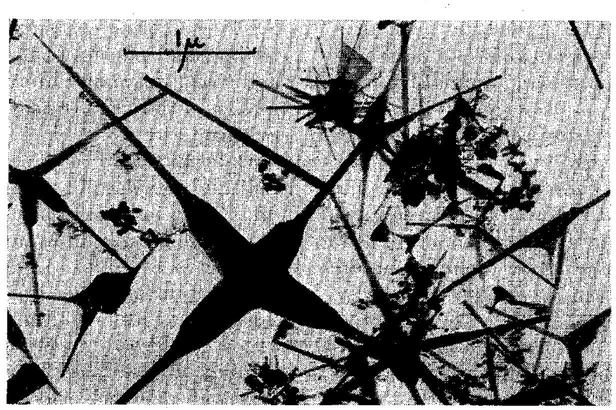
A monthly average of 340 tons of dust per square mile fell on Chicago during the first 5 months of 1949; the monthly average in 1948 was 699 tons and in 1947, 768 tons. The Chicago Dust Abatement Department uses data from the analytical chemistry laboratory of the Armour Research Foundation, Illinois Institute of Technology, as a check on the city's smoke and dust abatement program and as an indication of areas which need attention. Chicago maintains 25 dust-collecting stations distributed so that they cover the entire city. Dust is collected in battery jars, placed usually on the tops of four-story buildings where actual dustfall is collected rather than the transfer of dust close to ground level. The amount of dust that falls on Chicago varies inversely with the temperature and depends greatly on the amount of coal and oil used to heat homes and buildings. A cold wind not only makes it more difficult to heat a building, but it also agitates the dust from the ground into the air. Considerably more than 50% of the dust captured in the Loop during the winter months is noncombustible, largely fly ash and inorganic minerals from the soil. At other stations, particularly in outlying sections of the city, the greater percentage of the dust is soluble material.



Zinc Oxide Smoke (×25,000)