

Elizabeth Iha

Hello,

My name is Elizabeth Iha and I live in Bonner County, Idaho - the county next to the proposed smelter. I care about this proposal because I care about the environment and the health of my family. Idaho DEQ needs a equal seat at the table with WA DOE for determination of this permit.

ENVIRONMENTAL IMPACTS

1. In 2008 Washington State Legislature enacted E2SHB 2815, codified in the Public Health and Safety Chapter 70.235 which specifies limits to greenhouse gas (GHG) emissions in

Washington. RCW 70.235.020 details the following statutory GHG limits for Washington State:

"(1)(a) The state shall limit emissions of greenhouse gases to achieve the following emission reductions for Washington state:

- i. By 2020, reduce overall emissions of greenhouse gases in the state to 1990 levels;
- ii. By 2035, reduce overall emissions of greenhouse gases in the state to twenty five percent below 1990 levels;
- iii. By 2050, the state will do its part to reach global climate stabilization levels by reducing overall emissions to fifty percent below 1990 levels, or seventy percent below the state's expected emissions that year."

How possibly can WA state reach these goals by saying yes to a major polluter in the vulnerable Upper Columbia region?

2. The air quality in the region for the proposed smelter is already hit hard during fire season, please do proper study of the region during fire season for particulates and then use that data as baseline in determination of permit.

3. What will happen to the waste hydrochloric acid, nitric acid and the caustic pot-ash used in the last step of the process? Will it be released into our rivers, lakes and streams? Please see this article: https://www.pv-magazine.com/magazine-archive/the-trouble-with-silicon_10001055/ which mentions "In streams, rivers and lakes, this chemical acts as a fertilizer, nourishing algae and killing fish."

4. Consider the areas many air inversions that create "socked in" air with no place to go.

5. Idaho Department of Environmental Quality needs to have an equal place at the table with WA DOE in determination for permit.

ECONOMIC IMPACTS

1. Trees are injured by sulphur dioxide. Please see attached, "Injury to Northwestern Forest Trees By Sulfur Dioxide from Smelters" by the US Department of Agriculture. This regions economy has been and continues to be driven by trees and consequently, the health of trees. Per idahoforests.org, over 29,000 people work in Idaho's forest sector. Please consider this in your determination for permit.

2. This region is driven by many "clean mountain air" economic factors; tourism, recreation, hunting, fishing, mountain sports, etc. A smelter in this area would clearly have a negative impact on these economic factors and should be declined for permit.

HEALTH

1. Production of Silicon Metal and Alloys Is Associated With Accelerated Decline in Lung Function: A 5-Year Prospective Study Among 3924 Employees in Norwegian Smelters

https://journals.lww.com/joem/Abstract/2007/09000/Production_of_Silicon_Metal_and_Alloys_Is.12.aspx. Please read this. These are not the kind of jobs we want.

SEPA PROCESS

1. This proposal could negatively impact 3 states; Washington, Idaho and Montana. It's a proposal by a Canadian company. Due to these factors, I request that WA DOE request the involvement of the National EPA and adhere to a NEPA process instead of a SEPA process.

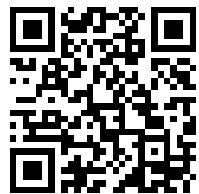
And lastly, NO FACILITY. Not here, not anywhere. The time is now to stop repeating the past. The time is now to take care of our environment and in doing so, take care of humanity.

Thank you for your consideration,
Elizabeth Iha

This is a reproduction of a library book that was digitized by Google as part of an ongoing effort to preserve the information in books and make it universally accessible.

Google™ books

<https://books.google.com>



A1.36:

no. 1117

Injury to

Northwestern Forest Trees

By

Sulfur Dioxide from Smelters



Technical Bulletin No. 1117

U. S. Department of Agriculture

Forest Service

UNIVERSITY OF VIRGINIA LIBRARY



X030347167

Digitized by Google



CONTENTS

	Page		Page
Introduction.....	1	Investigations in the upper Colum- bia River Valley, Wash.—Con.	
Sulfur dioxide injury and associated factors.....	2	Geographical distribution of sul- fur dioxide injury.....	20
Investigations in the vicinity of Anaconda, Mont.....	4	Relative susceptibility of trees, shrubs, and herbs.....	24
Description of the area.....	4	Reproduction of timber.....	28
General observations of injured timber.....	6	Mortality of ponderosa pine and Douglas-fir.....	30
Reproduction of the timber.....	7	Height increment of ponderosa pine.....	33
Diameter increment.....	7	Diameter increment of ponderosa pine.....	40
Injury by fungi.....	10	Timber recovery.....	44
Investigations in the upper Colum- bia River Valley, Wash.....	10	Summary.....	46
Description of the area.....	12	Literature cited.....	48
Dissemination of sulfur dioxide.....	14		
Diagnosis of sulfur dioxide and other kinds of injury.....	17		

AUG 4

iv

*Injury to Northwestern Forest Trees by Sulfur Dioxide from Smelters*¹

By THEODORE C. SCHEFFER and GEORGE G. HEDGCOCK,² *pathologists,*
Forest Service

INTRODUCTION

Sulfur dioxide discharged from smelters has been the major cause of gas injury to forest trees. Injury and killing of trees and agricultural plants by gases emanating from large industrial centers and from smelters has been mentioned in the literature for at least 80 years. A bibliography of reports on the subject prior to 1908 was published in 1913 (*11*).³ Publications dealing in significant part with gas injury to timber in North America are comparatively few (*6-10, 12, 13, 15-17*) and they cover mainly damage caused by smelters. By far the most comprehensive of these is one issued in 1939 by the National Research Council of Canada (*12*); this incorporates the findings of a number of investigators on areas in northeastern Washington affected by the smelter at Trail, British Columbia. One of the investigators, Katz (*10*), later summarized parts of this and additional work dealing with sulfur dioxide injury to trees and crop plants, and included an extensive bibliography on the subject. Shortly afterward, Thomas (*15*) reported on damage to plants by various industrial gases, and devoted considerable space to literature review. A compilation of 97 reports of air pollution was published in 1952 (*18*).

This report presents previously unpublished investigations by pathologists of the Division of Forest Disease Research on smelter injury to forest trees in Washington and Montana. Most of the observations were made from 1928 to 1936 on coniferous timber in that portion of the upper Columbia River Valley lying in Stevens County, Wash.; they were made in connection with the same general investigation on which the 1939 Canadian publication (*12*) was based. The remainder were made about 20 years earlier, on timberlands near Anaconda, Mont. The work was prompted by currently large amounts of timber damage near large smelters on the respective areas. The field work was planned and carried out by the late George G. Hedgcock; the writer served as his field assistant in 1929.

The primary objectives were (1) to ascertain the symptoms of smelter fume injury, (2) to determine how these might be distinguished from similar forms of injury caused by other agencies,

¹ Submitted for publication January 3, 1955.

² Deceased 1946.

³ *Italic numbers in parentheses refer to Literature Cited, p. 48.*

(3) to ascertain the relative susceptibility of different tree species to the injury, and (4) to appraise the timber damage with respect to reduced growth rate and reproduction and to increased mortality. Some of these features were considered broadly in relation to the proximity of the timber to the smelter, to the local and general topography, and to the climatic and other environmental factors. As these factors are not quantitatively the same in any two places, the investigations were essentially case studies.

During the past several years increasing attention has been given to the subject of air pollution (18). This bulletin contains information on one type of air pollution and its effects on forest trees and shrubs. No comparable studies have been made in this country.

SULFUR DIOXIDE INJURY AND ASSOCIATED FACTORS

Most of the smelters located in timbered areas treat relatively large tonnages of high-sulfur ores, and their stack effluents, when not controlled, are concentrated in such great amounts that injury to trees can be produced over comparatively large areas. The harmful constituents of smelter smoke are largely sulfur dioxide gas created by the combustion of sulfur in sulfide minerals, sulfuric acid formed from sulfur trioxide, and fumes of fine solid particles containing lead and arsenic compounds and acid sulfates (4). In some cases appreciable amounts of carbon monoxide and sulfur trioxide (liquid) may be present. Sulfur dioxide, however, is the only constituent that is liberated in sufficient quantity to cause more than comparatively local damage (20). Before the adoption of regulatory measures, smoke containing 1.5 percent and more of sulfur dioxide was frequently discharged from the stacks at some smelters. As little as one part per million (0.0001 percent) will injure certain plants severely.

Although smelter smoke generally is released from high stacks, it tends to diffuse rather slowly and the sulfur dioxide constituent, therefore, may reach the ground in harmful concentrations even after a considerable length of time. The descent and surface accumulation of sulfur dioxide is favored by its weight, which is a little more than twice that of air. Disturbances of the thermal balance of the air at times bring about a particularly rapid descent of the gas (17). This type of behavior will be discussed later. Other factors being equal, the distance from the smelter at which repeated injury occurs varies with the amount of sulfur dioxide discharged, the height above ground at which it is discharged, the wind velocity, the location of the vegetation in relation to the smelter, and the prevailing wind direction. Topographic features, such as deep valleys, which tend to confine the gas stream, naturally would extend the borders of injury beyond those in a more level terrain. It has been reported (1) that with high stacks the maximum concentration of sulfur dioxide in the smoke stream at ground level is farther from the stack than it is with low stacks, but this maximum concentration is smaller than in the case of the low stacks. Judging from curves presented by Wells (20), stack height determines sulfur dioxide concentration at ground level for only a few miles from the smelter.

The mechanism of sulfur dioxide injury to plants has received relatively little attention in the literature. Work by Brizi (2), Haselhoff and Lindau (5), and others suggests that on reaching the interior of the leaf, which seems usually necessary for injury to occur, the sulfur dioxide is partially converted to sulfurous and even sulfuric acid. Acted on by these chemicals, the cells lose their capacity to hold and to translocate water and as a result become flaccid and dry out. Critically impaired water conduction is evidenced particularly in broad-leaved plants, in which the tissues between the veins generally are the first to shrink and die. In addition to the desiccation brought about by sulfur dioxide fumigation, the chlorophyll of leaf cells is destroyed, possibly by the strong reducing properties of sulfur dioxide and sulfurous acid. This is indicated by an early loss of green color in the affected areas, even while the leaves are still turgid. In the last phase of injury, defoliation occurs.

The severity of injury incurred by plants exposed to a given dosage of sulfur dioxide varies with different species. Also, for a given species it is determined by the stage of maturity of the plant and by certain environmental conditions. Particular attention has been given to the nongenetic variables of susceptibility by Setterstrom and Zimmerman (14), and Katz and Ledingham in the report by the National Research Council of Canada (12). Their findings, and those of others reported by them, point to the generalization that the incidence and degree of injury is determined chiefly by the rate at which sulfur dioxide is taken up by the leaf—not the total amount taken up, and that this rate is greater as conditions favoring carbon dioxide assimilation (photosynthesis) are improved. The most influential of these conditions is the amount of stomatal opening, which is governed by the turgor of the leaf, particularly in the guard cells. Although photosynthetic activity itself contributes to guard-cell turgor, there is some evidence that it may additionally promote sulfur dioxide susceptibility. Plants apparently are not injured unless the leaf tissues are affected sufficiently to produce visible markings.

The following criteria have been summarized from findings by Setterstrom and Zimmerman (14). Although derived almost entirely from studies on crop plants, there is evidence that they are substantially applicable to trees also.

Temperature.—At 40° F. and below, plants are much more resistant to sulfur dioxide than at higher temperatures. In the usual range of temperatures higher than 40° F., variation in resistance with differences in temperature is small.

Relative humidity.—Susceptibility to sulfur dioxide injury increases as the relative humidity increases, the trend being more pronounced in the lower than in the higher relative humidity ranges. Wetting of leaf surfaces alone has little effect on susceptibility.

Soil moisture.—Minor variations, at the time of fumigation, in soil moisture in the range adequate for growth have no marked effect on susceptibility. Plants grown with an ample supply of moisture prior to fumigation are more susceptible than those grown with an inadequate supply, even though soil moistures at the time of fumigation are the same.

Light.—Susceptibility increases significantly with increased light at the time of fumigation, up to about 3,000 foot-candles. Plants grown in heavy shade prior to fumigation, on the other hand, are more susceptible to injury than those grown without shade.

Soil fertility.—Plants grown in poor soil are more susceptible to injury than plants grown in good soil.

Age.—Mature plants are more susceptible than young plants. Also, middle-aged leaves, which are the most functional ones, are more susceptible than relatively young or old leaves.

Continuity of fumigation.—Exposure to sulfur dioxide does not make a plant more susceptible to injury with subsequent exposure provided sufficient time elapses for recovery between fumigations. Discontinuous exposure at short intervals, however, may result in the same amount of injury as continuous exposure to the same concentration of sulfur dioxide. In either case, concentrations of sulfur dioxide less than 0.20 part per million apparently may be tolerated indefinitely without injury.

From the foregoing it is apparent that plants are most likely to be injured by sulfur dioxide when there is a favorable growing temperature, high relative humidity, bright weather, and protracted wind from the direction of the sulfur dioxide source. These factors in combination may have a slight to moderate additive effect, but their primary influence is more analogous to that of links in a chain; a critical deficiency in any one of them may preclude injury despite the adequacy of the others for it.

INVESTIGATIONS IN THE VICINITY OF ANACONDA, MONT.

Sulfur dioxide injury to trees in the vicinity of Anaconda, Mont., was investigated in 1910 and 1911. The scope of this study was comparatively small, the main purpose being to ascertain the approximate limits and the general degree of injury to the various timber species on the Deerlodge National Forest. The information gathered contains many elements of general or potentially general application, but is in considerable measure specific for the area and for the time.

Description of the Area

The Washoe smelter, source of the injury, is located close to Anaconda, Mont., near the southern and on the western edge of a broad stretch of the Clark Fork River Valley. Its position in relation to some of the major features of the surrounding topography is shown in figure 1. The broad portion of the Clark Fork Valley extends about 5 miles south of the smelter and about 30 miles north of it. Nearest the river, at elevations below 5,000 feet, the valley is mostly from 4 to 6 miles wide. Above elevations of 5,000 to 5,400 feet, the valley slopes begin to rise sharply and within a few miles reach elevations of 7,000 to 9,000 feet. The valley proper might be thought of as terminating at about the 6,000-foot level, where it is mostly from 10 to 12 miles wide. The slopes to the east and south terminate at the Continental Divide and to the west at the Flint Creek Range and the

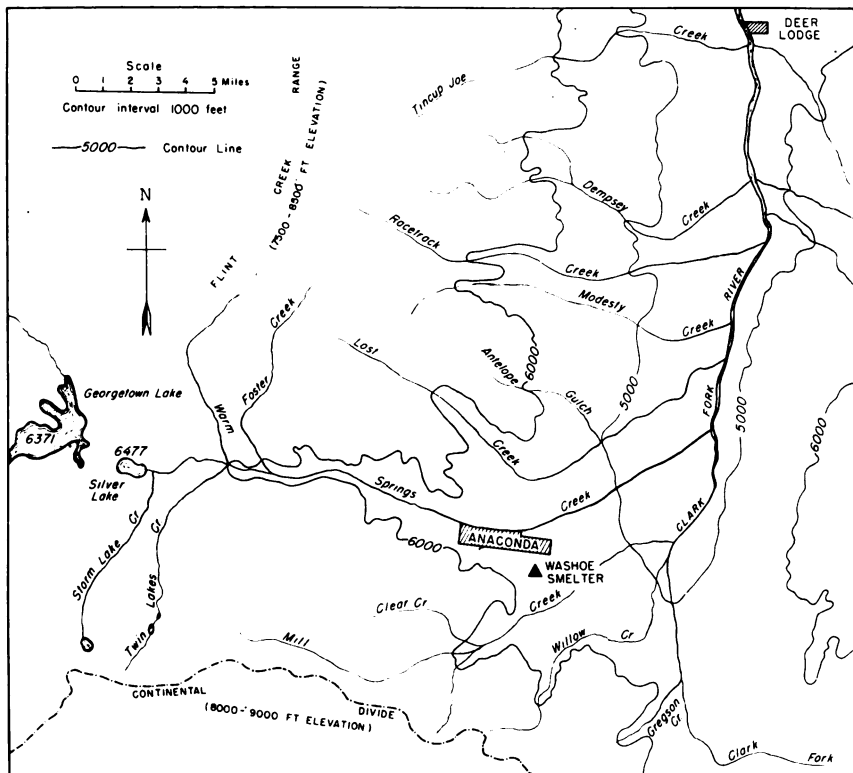


FIGURE 1.—General topographical features of the area affected by the smelter at Anaconda, Mont.

long saddle south of it, running between Georgetown Lake and Silver Lake. Numerous streams drain into Clark Fork River through rather deep clefts in the slopes to the west and southwest. The investigation centered on this general drainage area.

There was little timber in the valley proper. To reach the main body of timber, therefore, the smelter fumes generally had to move laterally from the main valley into tributary valleys or directly across comparatively high ridges. In a few instances the smoke could move directly into the tributary valleys, such as the valleys of Warm Springs Creek and Lost Creek (fig. 1), both of which have outlets near the smelter.

The smelter stands on a hill that rises about 600 feet above its base (elevation 6,000 feet). The stack also is nearly 600 feet high, so that the smoke is discharged at an elevation of approximately 7,200 feet. The smoke stream levels off several hundred feet still higher, and thus is in a position to extend rather freely over a large surrounding area. The smelter, erected in 1902, was preceded by a smaller structure, which was established in 1884 for preliminary smelting of ores and enlarged in 1892 for complete smelting.

The prevailing local winds are from the south during all but the spring months, when they are from the northwest. All directions are regularly represented, however. Precipitation ranges from 15 to 20 inches per year; thus the area might be described as semiarid. Midsummer temperatures average about 60° to 65° F. and midwinter temperatures about 20° to 25° F. at the lower elevations. Daytime temperatures during the summer often are in the nineties. In general, precipitation and temperature are of a continental character.

The principal species of trees are lodgepole pine,⁴ Douglas-fir, Engelmann spruce, subalpine fir, and limber pine (*Pinus flexilis* James)—all conifers. Minor coniferous species are Rocky Mountain juniper (*Juniperus scopulorum* Sarg.), common juniper (*Juniperus communis* L.), and, rarely, ponderosa pine. Deciduous species, all of minor importance, are quaking aspen, lanceleaf cottonwood (*Populus* × *acuminata* Rydb.), thinleaf alder, water birch, and several species of willows.

General Observations of Injured Timber

Based on amounts and frequencies of foliage thinning at different distances from the smelter, the order of susceptibility of timber species to sulfur dioxide liberated by the Washoe smelter was as follows, beginning with the most susceptible:

- | | |
|---------------------|---------------------------|
| 1. Subalpine fir | 5. Ponderosa pine |
| 2. Douglas-fir | 6. Limber pine |
| 3. Lodgepole pine | 7. Rocky Mountain juniper |
| 4. Engelmann spruce | 8. Common juniper |

The last three species showed practically no injury. Douglas-fir exhibited about as much foliage injury as subalpine fir but was not killed as extensively near Anaconda, Mont.

General smelter injury could be distinguished from winter injury partly by differences in the relative susceptibility of the trees to the two kinds of damage. The order of susceptibility of species to the red-belt form of winter injury was:

- | | |
|-------------------|---------------------------|
| 1. Ponderosa pine | 5. Engelmann spruce |
| 2. Douglas-fir | 6. Subalpine fir |
| 3. Lodgepole pine | 7. Rocky Mountain juniper |
| 4. Limber pine | 8. Common juniper |

The most marked differences in order of susceptibility to smelter injury and to winter injury occur with the subalpine fir and ponderosa pine.

Apparently because the smoke stream reached comparatively great heights above the surrounding terrain, injury to the forests was widespread in all directions, and the smelter was near the geographical center of the area of worst injury. In terms of an approximate radius of injury, in 1910 and 1911 all of the major tree species were either dead or dying as far as 5 to 8 miles from the smelter. Engelmann spruce showed no evidence of injury beyond 15 miles, lodgepole pine beyond 18 miles, Douglas-fir beyond 20 miles, and subalpine fir beyond 22 miles.

⁴ Scientific names of most of the trees mentioned in this bulletin are given in table 5, p. 25.

With reference to features shown in figure 1, observations by Haywood (7) would place the approximate limits of timber injury a year or two earlier than these observations somewhere near Tincup Joe Creek on the north, near Georgetown Lake on the west, and on the south, about 2 miles south of the Continental Divide where it comes closest to the south fork of Mill Creek. The extension of injury was least in the southward direction, probably because of the higher elevations of the mountains to the south and because of the prevalence of wind movement from the south. The easterly extension of injury could not be reliably ascertained because of the complication introduced by smoke from smelters at Butte, Mont., about 22 miles south-east of Anaconda.

Although the amount of injury was generally less with increasing distance from the smelter, the decrease was not uniform. The growing conditions of the forest, of course, were not uniform. Moreover, smoke from the smelter was channeled to some extent through the creek valleys and, at higher levels, was deflected from a straight course by mountains and high ridges. Such deflection was observed on several days in four different localities. As a result, certain slopes were exposed to critical dosages of sulfur dioxide and others were not.

Reproduction of the Timber

Within 5 miles of Anaconda, Mont., there was little or no restocking of logged or burned-over areas. Cones of surviving trees were to a large extent abortive; hence they may not have contained viable seeds. Farther out a limited number of seedlings had become established but were able to survive no longer than 1 or 2 years.

Near the border zone of injury some reproduction was observed, especially lodgepole pine, which was abundant. As a rule, however, reproduction was comparatively slight where the foliage of the older trees showed injury. In 1911, stands of lodgepole pine and Douglas-fir southwest of Anaconda, Mont., were dying (fig. 2); in 1931 these areas were denuded and eroding. Well outside the zone of injury, northerly slopes that had been logged over or burned usually were becoming restocked fairly promptly and uniformly. No large areas destitute of seedling trees were seen in this area, although on many southerly slopes the stocking was retarded and quite irregular because of naturally unfavorable conditions attendant to such exposure.

Diameter Increment

A small number of trees were felled for the purpose of observing current and prior rates of diameter increment. The trees were mostly of sapling or small-pole size, and all were free of visible fire, insect, or fungus damage. Some were growing well within the zone of smelter injury and others were growing near the border of this zone, as evidenced by relative ages of green foliage retained by the trees. Also, trees from comparable stands of timber outside the zones of injury were measured to determine normal rates of increment. Certain of these outside trees were on an area that in 1908 and 1909 had



FIGURE 2.—Dead and dying lodgepole pine and Douglas-fir trees, southwest of Anaconda, Mont., 1911. This region was bare and badly eroded and gullied by 1931.

suffered considerable winter injury of the red-belt type. The measurements were made on disks taken at approximately breast height in the trees.

Species given most attention in this particular phase of the study were subalpine fir, lodgepole pine, and Douglas-fir. The average diameter increments in various years of the sample trees of these species are plotted in figure 3. The differences in growth rate shown for trees in the different zones, although indicative, are not reliable evidence of a sulfur dioxide effect. That is because of uncertainty as to the natural differences to be expected, had sulfur dioxide not been present. There are two additional lines of evidence of growth retardation, however. These are (1) a persistent decline in growth rate in the zone of smelter injury during periods in which the overall rate in the outside zone tended to increase or remain about the same, and (2) the comparatively smooth growth curves for the injury zone as contrasted with the irregular curves for the outside zone. The irregularity of annual rings reflects year-to-year growth response to fluctuations in precipitation whereas regular growth under the same conditions denotes the presence of a growth-inhibiting agency.

Thus, retardation of diameter growth of all three species is indicated for all years from 1892 through 1910 in the zone of injury. In the cases of the subalpine fir and the Douglas-fir, the two local species most sensitive to sulfur dioxide, the degree of retardation apparently was greater in the years subsequent to 1900, or thereabout, than in earlier years. This may have been connected with the installation of the new smelter in 1902. The situation with trees on the border of the zone of injury is not so well defined. Characteristics of the growth curves, intermediate between those for trees in the zone of injury and those for trees outside, suggest the occurrence of some intermediate degree of retardation. Judging by the relative smoothness of the

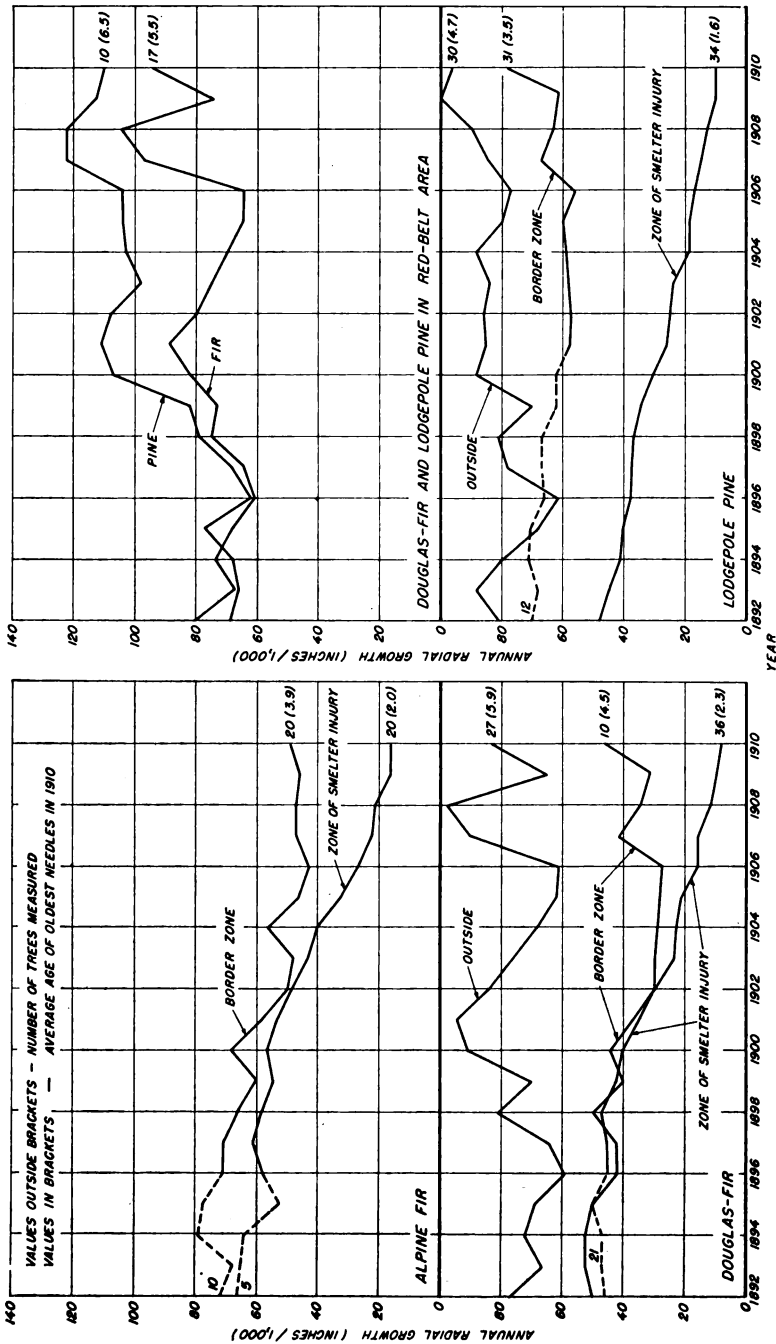


FIGURE 3.—Annual diameter increment, in terms of radius enlargement, of trees inside and outside the Anaconda, Mont., zone of sulfur dioxide injury, and of two of the species in an area subject to red-belt type of winter injury. Number of trees sampled is indicated by numerals adjacent to the broken and continuous lines.

curves, retardation also occurred in the border zone as early as 1892, at least with lodgepole pine and Douglas-fir.

For the trees in the red-belt area, from the same criteria and from the general similarity of their growth curves to corresponding ones for the outside zone, there was no marked reduction of diameter growth due to winter injury.

Injury by Fungi

A number of species of fungi were parasitic on needles or twigs of coniferous trees. Examination of several hundred trees in and outside the area of smelter injury indicated that most of such fungi were nowhere prevalent and the frequency of occurrence was about the same in both areas. Certain rusts, *Melampsorella cerastii* Schroet., *Peridermium coloradense* Arth. & Kern, and species of *Phragmidium*, *Melampsora*, and *Gymnosporangium*, were almost absent from the smelter zone but were rather abundant in surrounding areas.

Dwarf mistletoe is rather common on Douglas-fir and lodgepole pine east of the Pacific coast. However, there was less dwarf mistletoe in and adjacent to the Anaconda area than at other places.

The prevalence of heart rot was ascertained in 180 lodgepole pine trees and in 480 Douglas-fir trees 100 years or more old, growing in the Anaconda area. About 7 percent of the pines and 72 percent of the Douglas-firs were infected in various degrees by *Polyporus schweinitzii* Fr. or by *Fomes pini* (Fr.) Karst. These proportions were comparable to those found in Montana forests outside the area.

INVESTIGATIONS IN THE UPPER COLUMBIA RIVER VALLEY, WASH.

The upper Columbia River Valley, Wash., investigations were made from 1928 through 1936, chiefly in the main valley and larger tributaries between the Canadian border and points about 65 miles southward. Hedcock visited a small number of additional areas eastward and westward in the same general region, to become oriented to tree and shrub conditions where virtual absence of sulfur dioxide would be unquestioned. The investigations in the upper Columbia River Valley were much more comprehensive than the earlier ones at Anaconda, Mont. Moreover, the geographical features differed significantly. Whereas the Anaconda smoke stream attained a sufficient relative height to spread widely in most directions, that of the Columbia River Valley was held to a large extent between high valley slopes. As a consequence, the area of injury was much elongated and the extent of damage varied markedly with elevation.

The following information was sought: (1) The relative prevalence of injury by sulfur dioxide, winter, drought, insects, and fungi. (2) The geographical distribution of sulfur dioxide injury. (3) The relative susceptibility to sulfur dioxide of various trees, forest shrubs, and associated herbaceous plants. (4) The effect of the sulfur dioxide on the reproduction and mortality of trees in different parts of the area. (5) The effect of the sulfur dioxide on the height growth and diameter increment of representative trees in different parts of the area.

Data were taken quantitatively wherever practicable, and on trees that would permit comparisons of the sulfur dioxide injury at different places with the least complication by differences in natural factors. To determine the effect of sulfur dioxide on trees, evidence of injury was compared with a background of data obtained at similar places outside the area of injury. Locations used for this purpose were largely in the main river valley south of Kettle Falls, Wash., to a distance of about 25 miles (fig. 4) and to a limited extent along the

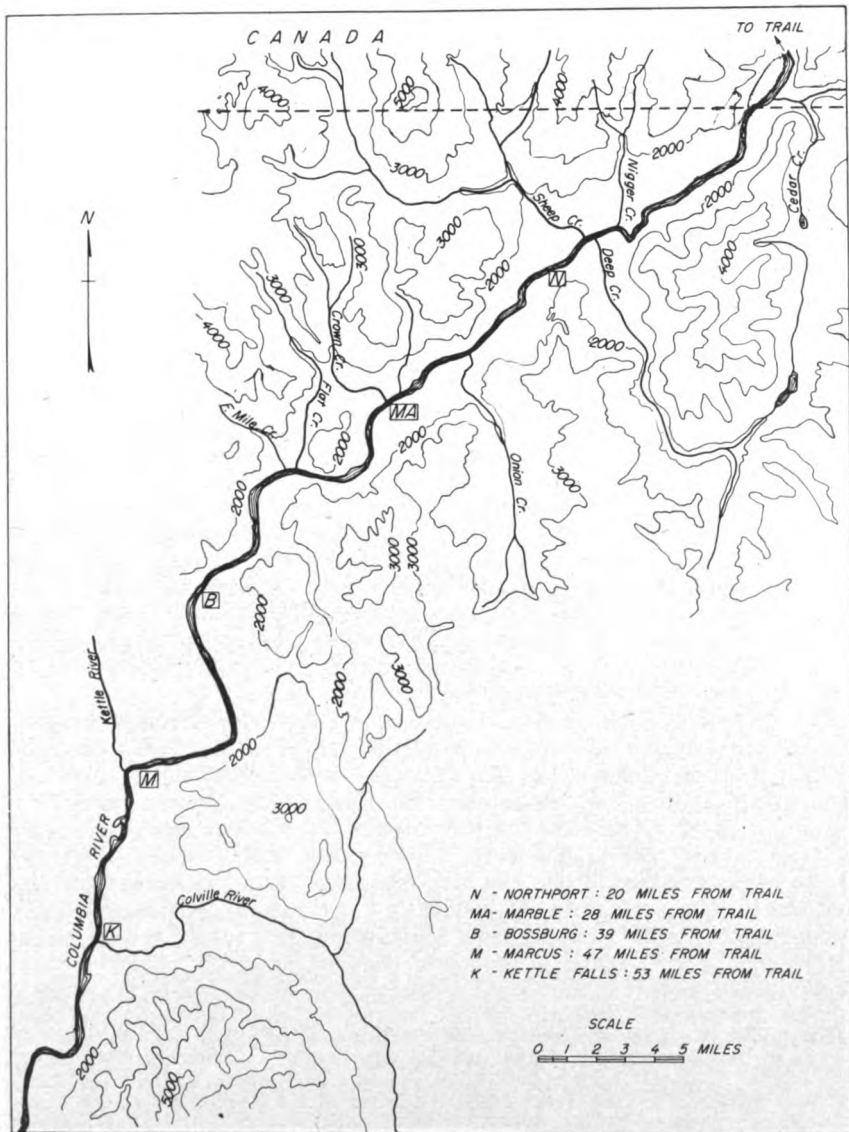


FIGURE 4.—Topographic features of the upper Columbia River Valley, Wash., area of investigations.

upper reaches of the tributary valleys. Control sites were selected sufficiently near the area of injury to have a similar climate. These sites were restricted to alluvial soils since this was the type of soil on which the quantitatively appraised trees in the area of injury were growing. Some differences in precipitation and soil fertility were unavoidable and had to be taken into account in making comparisons.

To further base the comparisons on uniform growing conditions, trees or groups of trees to be compared were selected for similar characteristics in such respects as stage of maturity, degree of dominance, freedom from crowding, and freedom from fire injury.

The boundaries of injury occurred at various elevations, on a variety of sites, and in timber of different kinds and ages. The chief criterion of injury was the typical leaf markings caused by sulfur dioxide; these were determined for forest shrubs as well as trees. The relative abundance of injury manifested by leaf markings and the proportion of certain trees with less than a normal quota of foliage served to delineate broad zones of injury according to its severity.

For convenience in mapping, all records were classified according to the quarter section on which they were obtained.

Description of the Area⁵

Topography

The smelter is located in a gorge of the Columbia River at Trail, British Columbia, about 11 miles north of the border. From Trail, the river swings eastward and then southwestward to form a large arc terminating near the border. After a brief deflection southward it continues southwesterly, without important deviation for approximately 15 miles to Marble, about 28 miles by river from Trail. It then makes a series of large bends covering an additional distance of approximately 21 miles to Marcus, about 48 miles by river from Trail; its direction over the remainder of the study area is generally south. Most of these and certain subsequently described topographic features are shown in figure 4.

This portion of the Columbia River Valley cuts through mountainous terrain, the slopes rising rather steeply but rarely precipitously from flatlands along the river to elevations of 3,000 to more than 4,000 feet within a few miles. Adjacent to the river and occupying much of the area below the 2,000-foot level are a series of terraces or benchlands. The modal width of the valley at 2,000 feet, exclusive of places at tributary inlets, is about 2 miles. At an altitude of 3,000 feet the northern 25 miles of the valley varies in width from about 2½ miles to 4 miles; progressing southward, the valley at the same

⁵ A considerable part of the geographical and related descriptions of the area is based on material contained in U. S. Department of State Publication 1649 (17) and in a U. S. Department of Agriculture report of a soil and agricultural survey of the areas (19). Most of the topographic details were provided by a 1942 U. S. Forest Service map of the western half of the Kaniksu National Forest. Some of the land features near the river at the time of the survey were subsequently changed by the rise in the Columbia River following the building of the Grand Coulee Dam.

altitude broadens considerably. In general, the valley is comparatively narrow and deep, and thus at times would act as a flume, confining and guiding the passage of sulfur dioxide released in it.

Soil

The soils of the river benches and wider portions of the tributary creek valleys consist largely of alluvial deposits formed by glacial outwash and river and lake sedimentation. The character varies from fine sandy and silt loams to coarser deposits of sandy and gravelly texture. The organic content is low for the most part and drainage is excessive. These soils are productive enough for limited types of farming. Some of the vegetative cover is subject to drought during the latter part of the growing season, mainly because of the excessive subdrainage.

Above the benchlands and farther back from the river and tributary streams, certain areas of land are hilly but, nevertheless, moderate enough in contour for pasturage and limited crop growing. The soils here are largely derived from weathered glacial till. They are well drained for the most part and have low to good moisture-holding capacity.

Interspersed with the hilly land but more prevalent at higher elevations are areas of essentially mountainous character. The slopes are steep and often stony, and rock outcrop is usually abundant. Except for the rock outcrop and stony sections, the soil is deep enough to support a heavy growth of timber.

Climate

The climate of the Columbia River Valley in the portion investigated is relatively dry. The mean annual precipitation ranges from less than 16 inches in the southern part to somewhat more than 18 inches in the northern part. The corresponding number of days per year in which the precipitation is 0.01 inch or more is about 90 and 115. A little more than 40 percent of the precipitation occurs during the months of April through September. Total annual snowfalls range from about 40 to nearly 60 inches.

The average temperature over the area is about 68° F. in July and about 22° F. in January. Midsummer temperatures during the day commonly exceed 90° F.; the nights are comparatively cool. Days without killing frosts usually are limited to a period of about 3 months, between early June and early September.

Air Movements

The direction of the surface wind is, in general, from the northeast down the river valley, but this varies at different times of day and in different seasons. During the growing season the air generally is in active movement whereas during the colder months relatively calm periods of several days are common. Moreover, the wind direction during the day is generally from the north until late in the afternoon and then from the south. This variation also may occur during the nongrowing season, but with less frequency and regularity.

Timber

Much of the area is forested, chiefly with coniferous species. Ponderosa pine, Douglas-fir, and western larch are the species of chief commercial importance. At the time of the study most of the mature trees had been cut except on the higher mountain slopes and at scattered places in the valleys. The remaining timber was generally less than 100 years old.

Ponderosa pine, with some admixture of Douglas-fir, western larch, and lodgepole pine, predominates on the lower slopes and river benches. At higher elevations and farther up the tributary valleys, Douglas-fir and larch are the dominant species, particularly in the northern part of the watershed where there is more rainfall. By and large, the stands here are comparatively thick. Some western redcedar, western hemlock, and Pacific yew also occur at the higher elevations, chiefly in draws and moist places on north and east slopes. Additional minority species on the higher slopes are western white pine, grand fir, subalpine fir, and Engelmann spruce, and on the drier, rockier sites, western juniper.

Broad-leaved, deciduous trees are relatively few and unimportant on the area; these will be mentioned in discussing species susceptibility to sulfur dioxide.

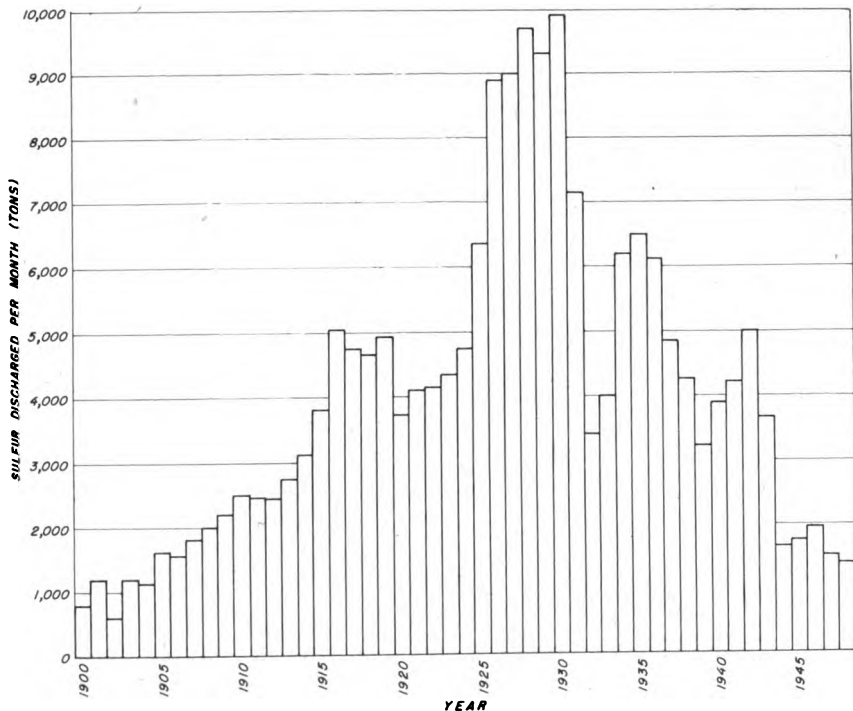
Dissemination of Sulfur Dioxide

The area has been affected by two smelters. One, a small plant, located at Northport, Wash., in the river valley about 10 miles south of the Canadian border, was operated intermittently from 1896 to 1908 and continuously from 1916 to 1921. About 2,100 tons of sulfur per month were released during the first period and about 900 during the latter period.⁶ These are relatively small sulfur outputs and the damage caused by them covered a rather restricted area.

The other smelter, located at Trail, B. C., was started in 1896 also, and has been in continuous operation. It is one of the largest non-ferrous ore-processing plants in the world. The quantities of sulfur emitted from its stacks from 1900 to 1948 are shown in figure 5.⁷ The monthly output in 1900 was about 800 tons and by 1925 it had increased to approximately 6,300 tons. Peak outputs, averaging about 9,300 tons monthly, occurred during the next 5 years (1926-30), and in this period damage to vegetation was reported. In 1931 the discharge of sulfur was sharply curtailed by the institution of sulfur-recovery measures to about 7,100 tons per month, and, with considerable irregularity, this downward trend was maintained until in 1948 the amount was only about 1,400 tons. In addition to these sulfur curtailments, in 1933 the smelter adopted the practice of regulating the sulfur discharge so that the larger amounts would coincide with wind direction and other atmospheric conditions least favorable for sulfur dioxide injury (12). The influence of these regulatory measures in limiting sulfur dioxide at points southward in the river valley to concentrations mostly less than 0.50 part per million is shown by figure 6.

⁶ One ton of sulfur is equivalent to approximately 2 tons of sulfur dioxide.

⁷ Yearly records of sulfur discharge were furnished by the Consolidated Mining & Smelting Co. of Canada.



M 89744 F

FIGURE 5.—Average monthly discharge of sulfur from stacks of the smelter at Trail, British Columbia. (Courtesy of Consolidated Mining & Smelting Co. of Canada.)

Late in 1925, and early in 1927, in the period of maximum sulfur discharge, two 409-foot stacks were erected. The gases from these stacks rise about 400 feet to an elevation of approximately 2,400 feet, or 1,100 feet above the Columbia River, before leveling off.⁸ During the growing season, the sulfur dioxide in this high gas stream regularly tends to descend twice daily, a peak amount reaching the ground at about 9 to 10 o'clock in the morning and again, usually with shorter duration, in the early evening. These descents are caused by disturbances of the thermal balance of the air, the first created by the morning sun and the second by the setting of the sun. The concentration of sulfur dioxide during this daytime type of fumigation usually increases very rapidly to a maximum in a few minutes and then drops off at a diminishing rate until only traces remain after 2 or 3 hours. Moreover, because of the prior accumulation and extension of the sulfur dioxide in the upper air, the fumigations occur almost simultaneously at widely separated points.

During the nongrowing season or at other times in which there is not the daytime type of fumigation, the sulfur dioxide drifts along the valley with the wind, diffusing more or less uniformly as it ad-

⁸ Information regarding the movement of sulfur dioxide from the smelter is given in greater detail in U. S. Department of State Publication 1649 (17).

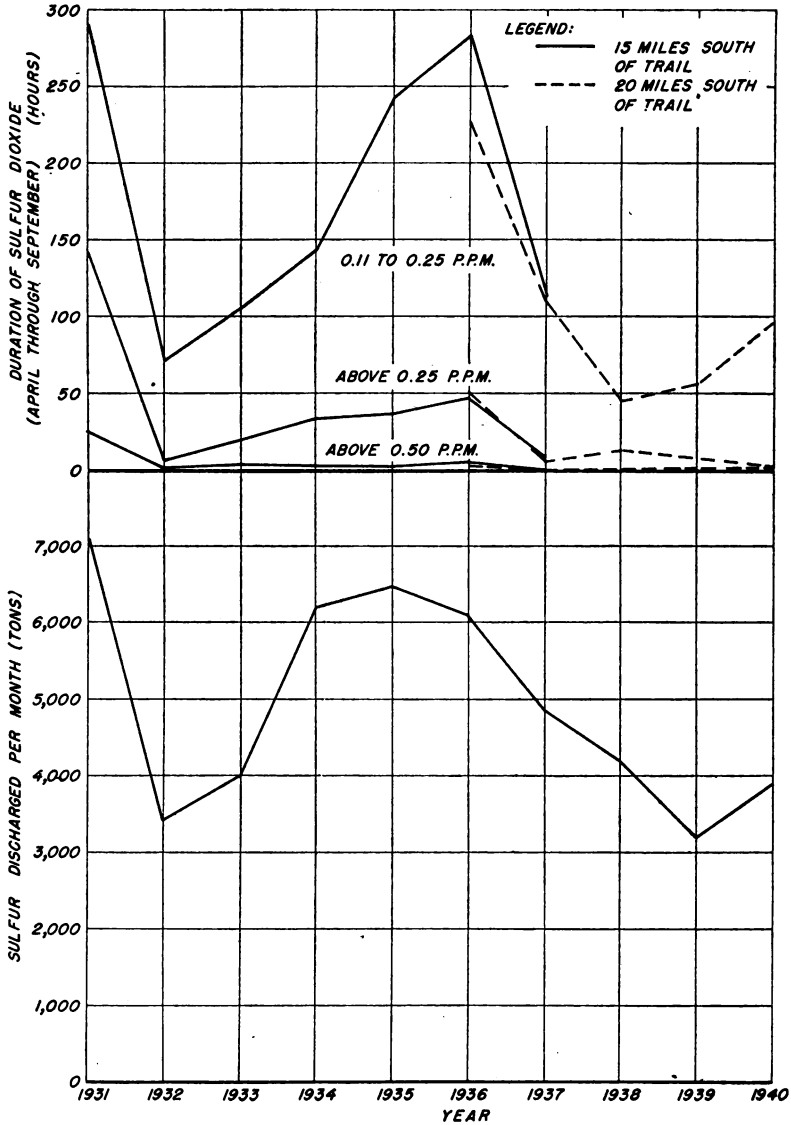


FIGURE 6.—Average rates of sulfur discharge from the smelter stacks and corresponding summer occurrence of sulfur dioxide at points south of Trail, British Columbia, following curtailment of sulfur output in 1931 and associated regulatory measures instituted in 1933. (Based on data by Katz (12) and others (17).)

vances. The fumigations under this condition occur in succession as the gas moves from place to place increasingly distant from the smelter.

The concentration of sulfur dioxide decreases very rapidly from Trail, British Columbia, to a point about 16 miles downstream. Far-

ther to the south, the concentration is lower and decreases more gradually. An example of the rapid dilution is shown in table 1, based on data obtained by Katz (12).

A paramount factor affecting the rate of diffusion of sulfur dioxide is the turbulence or gustiness of the air that is created by eddy currents. Ordinarily, with good turbulence, the sulfur dioxide is rapidly diffused upward above the sides of the valley.

TABLE 1.—Average and maximum concentrations of sulfur dioxide at different distances south of the smelter at Trail, British Columbia

Distance south of the smelter by river (miles)	Data from automatic recorders for the period May through September 1937 ¹	
	Average sulfur dioxide concentration	Maximum sulfur dioxide concentration
6.....	0.0380	1.22
15.....	.0118	.31
20.....	.0103	.28

¹ Data by Katz (12).

Diagnosis of Sulfur Dioxide and Other Kinds of Injury

To survey the effects of the release of sulfur dioxide from the smelter, particularly on the areas of marginal influence, it was necessary to determine the kinds of injury that might be confused with that caused by sulfur dioxide. Winter and drought injury are most likely to be confused with sulfur dioxide injury, since all of these are manifested by a reddening or browning of the foliage. Insects and fungi present a further complication by causing a decline in vigor of trees attacked by them.

Sulfur Dioxide Injury

Pine trees subjected to repeated injury by sulfur dioxide lose their needles in a succession beginning with the oldest ones and ending with the youngest.⁹ Depending on the severity and frequency of fumigation, the youngest needles and some others may survive and continue to function. Moreover, the needles persist to a greater age on the younger branches, those in the upper crown and on young trees, than they do on the older branches. As a result, trees that are not killed, especially the older ones, acquire a thin, scrawny appearance, with

⁹ The resistance of the young foliage is attributable to its stomatal condition. Blüsgen (3) found that stomata become effectively movable only in the mature leaf. In the young leaves they are closed, or open only under especially favorable conditions. Although the middle-aged needles may be injured most, the oldest ones tend to be cast first.

foliage concentrated in the tops. This symptom of injury is a striking one and was not apparent with injuries of other kinds to pines on the area.

Coniferous needles affected by moderate dosages of sulfur dioxide become chlorotic and yellowish in color before they are shed prematurely. With large dosages, on the other hand, they tend to acquire first a water-soaked appearance and shortly thereafter a reddish or brownish color, varying in intensity. Often the color is rather pronounced, especially where the foliage is exposed to direct sunlight. Needles frequently are not discolored over their entire length; the area of discoloration may be either solid or discontinuous in the form of bands. The banded character is more typical of injury occurring in the winter.

Leaves of injured broadleaf trees and forest shrubs, if not killed entirely, become desiccated and turn brown between the veins. Although marginal damage also may take place, the intercostal pattern generally predominates. This characteristic was rather specific for sulfur dioxide injury on the area and where conspicuous amounts were found it was considered to be evidence of contact with sulfur dioxide. Shrubs, some of which were markedly susceptible to the gas, were particularly useful in diagnosing sulfur dioxide injury because they were prevalent almost everywhere.

Winter and Drought Injury

Winter injury and drought injury are both caused to a large extent by a deficiency of water in transpiring leaves; it is not surprising, therefore, that their appearance resembles in some respects the desiccating effects of sulfur dioxide. Nevertheless, there are a number of differentiating features that in total provide a reasonably satisfactory means of distinguishing among the three kinds of injury. Such features pertaining to needle-bearing trees are listed in table 2.

A large area in Washington and Idaho having essentially the same climate as the upper Columbia River Valley was surveyed in 1930 for winter injury. Ponderosa pines averaging not more than 1 percent of the total stand were found to have suffered winter injury in the winter of 1929-30. Practically all of these recovered in the following year. There was no evidence of a greater amount of winter injury on the smelter area. A similar survey in 1935 revealed no winter injury either outside or inside the smelter area.

Drought injury was largely limited to the smaller trees, especially of lodgepole pine. Most of this took place in 1929 and 1934. Damage specifically attributable to drought injury was most readily appraised by comparison with the trees on the outside areas. This was believed to be a conservative procedure inasmuch as in most years the area affected by sulfur dioxide had the advantage in rainfall.

Injury by Fungi and Insects

Damage to the timber by insects did not resemble sulfur dioxide injury except superficially. Epidemic infestations of Douglas-fir by the tussock moth (*Hemerocampa pseudotsugata* McD.) occurred at a few places, mostly outside the sulfur dioxide affected area. Bark

TABLE 2.—*Some distinguishing features of sulfur dioxide, winter, and drought injury to needle-bearing trees*¹

Diagnostic feature	Kind of injury		
	Sulfur dioxide	Winter	Drought
Needle discoloration.....	Reddish or brownish when strongly affected.	Reddish or brownish, but generally less than by sulfur dioxide.	Reddish or brownish, but generally less than by sulfur dioxide.
Extent of discoloration of individual needles.	Often incomplete; in the form of bands.	Needles usually entirely discolored.	Needles usually entirely discolored.
Seasonal occurrence of discoloration.	Any time in mild weather.....	Spring.....	Late summer to early winter.
Maturity of affected needles.	Middle-aged and older needles affected first.	Older needles affected first.....	Youngest needles affected first.
Maturity of affected trees.	Excepting seedlings, older trees affected most.	Young trees affected most.....	Young trees affected most.
Character of injury to branches.	Branches die progressively from bottom of tree to the top, and needles from the inner parts of branches to the ends.	Lower branches are not injured if protected by snow.	Branches die progressively from top of tree to the bottom and needles from the branch tips inward.
Orientation of crown injury.	Circumferential differences in crown injury are small or absent.	The side of the crown most exposed to sun and warm wind is affected most.	Circumferential differences are usually small, or, if present, are related to differences in ground moisture.
Killed trees.....	Dying relatively gradual; deficiency of needles at death.	Dying rapid; normal quota of needles at death.	Dying rapid; normal quota of needles at death.
Geographic distribution of injury.	Severity of injury is related to distance from smelter and to topographic features tending to confine sulfur dioxide.	Injury tends to be distributed in bands roughly paralleling contour lines.	Injury is most likely to occur where topography and soil are unsuited to the accumulation and retention of ground moisture.

¹ The specific features may be regarded as common but not invariable.

beetles (*Dendroctonus* spp.) and engraver beetles (*Ips* spp.) were observed on many of the larger ponderosa pines. The attacks apparently followed weakening of the trees and were not the primary cause of the weakening. This is typical except where infestations have built up to epidemic proportions.

Most parasitic fungi were no more prevalent on the area of sulfur dioxide injury than on outside areas. In fact, a number of fungi parasitic on needles and leaves appeared to be retarded or inhibited where sulfur dioxide was present, especially where the injury to forest trees and shrubs was greatest. This was noted chiefly with species of *Cronartium*, *Coleosporium*, *Melampsora*, *Peridermium*, *Pucciniastrum*, *Puccinia*, *Lophodermium*, *Hypoderma*, and *Hypodermella*. Two rusts, *Melampsora albertensis* Arth. on quaking aspen and *M. occidentalis* Jacks. on black cottonwood, were not found in the zone of greatest sulfur dioxide injury, but occurred sparsely where there was moderate injury and more abundantly where the injury was least. The same was true of *Pucciniastrum pustulatum* (Pers.) Diet. on grand and subalpine firs, *Coleosporium solidaginis* (Schw.) Thuem. on lodgepole pine, *Cronartium harknessii* Meinecke, *C. comandrae* Pk., and *Lophodermium pinastri* (Schrad.) Chev. on ponderosa and lodgepole pines. The larch needle parasite, *Hypodermella laricis* Tub., was found to some extent on outside areas but not at all on the area affected by sulfur dioxide.

Armillaria mellea (Fr.) Quel., on the roots and root collar of the pines, was most prevalent inside the area of sulfur dioxide injury. This was not surprising since this fungus, like bark beetles, occurs more frequently on weakened than on healthy trees.

Geographical Distribution of Sulfur Dioxide Injury

Three zones of injury were recognized and quantitatively delimited through 1929, 1930 (year of maximum sulfur discharge), and 1931. This was done according to the average percentage of injury to ponderosa pine and Douglas-fir trees and to forest shrubs. The percentage of injury to the trees was measured by the relative numbers of trees on sample plots that were dead or dying, or in poor condition. The condition of a tree was assessed by criteria that will be described in a following section. The percentage of injury to shrubs was measured by the percentage of leaf tissue conspicuously marked or killed. Injury as thus appraised in the three zones was 60 to 100 percent for zone 1, 30 to 60 percent for zone 2, and 1 to 30 percent for zone 3. Chief among the indicator shrubs was Pacific ninebark (*Physocarpus capitatus* (Pursh) Kuntze, because it was present nearly everywhere and because it was outstandingly susceptible to sulfur dioxide.

Although all degrees of injury were present in each zone, there were substantial differences in average severity. Zone 1, which was relatively low in the valley and nearest the smelter, contained in large part trees with what might be called acute injury. The acute injury was characterized by quick killing of leaves or needles and conspicuous marking of the foliage (fig. 7). Zone 2 extended higher up and farther south in the valley, and contained trees with injury of intermediate severity. Zone 3, extending still higher and farther south,

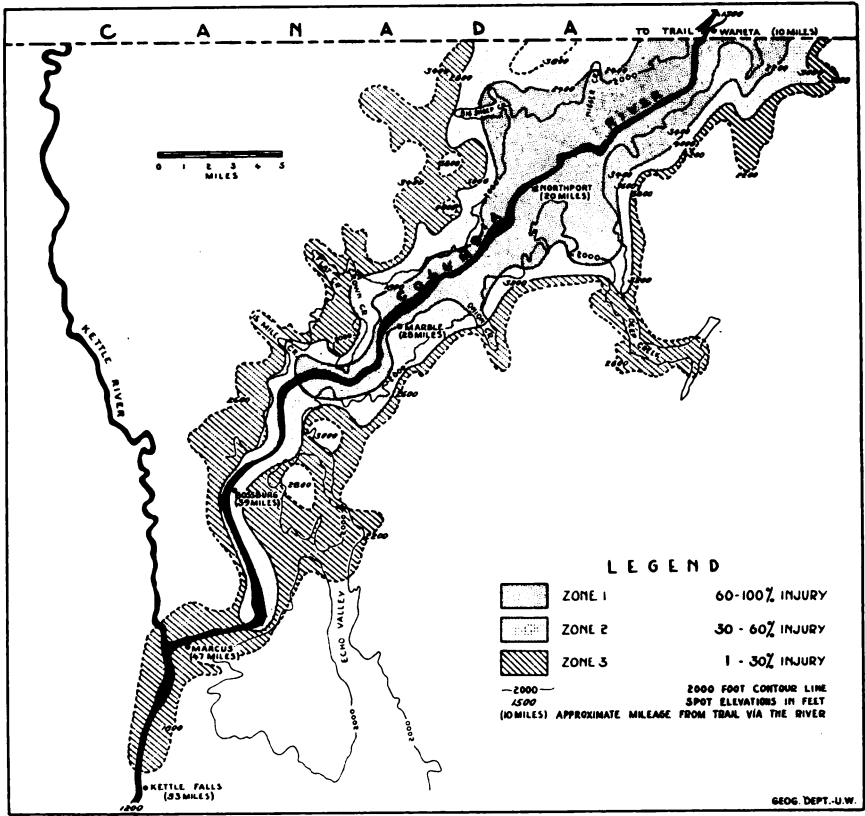


FIGURE 7.—Typical ponderosa pine trees in zone 1 in 1930, showing crown killing and thinning by sulfur dioxide. The site is a river bench about 28 miles south, by river, of the smelter at Trail, British Columbia.

to the limit of injury, contained trees with relatively slight markings and suffering from slow but progressive deterioration.

A map showing the areas occupied by zones 1, 2, and 3 in 1931 is given in figure 8. Further references in this report to zones of injury relate to the zones as located in 1931, even though, as in later years, the actual amounts of injury had diminished in the respective zones.

The area outside of zone 3, which hereafter will be referred to as the outside zone, contained uninjured trees that served as controls for estimating damage in the other zones. As already mentioned, the



M 8:912 F

FIGURE 8.—Zones of sulfur dioxide injury in the upper Columbia River Valley, Wash., in 1931, and associated topographic features of chief significance.

outside zone was largely in the main river valley south of Kettle Falls to a distance of about 25 miles and to a limited extent in the upper reaches of tributary valleys.

In 1931 the southern limit of injury (zone 3) was approximately 52 miles along the course of the river from the smelter. Corresponding limits for zones 1 and 2, respectively, were about 33 miles and 45 miles. The width of the observed area of injury, i. e., across the valley, was highly variable because of the influence of pronounced variations in the topography. It ranged in triangular fashion from an average of about 10 miles in the northern part to about 1½ miles near its southern extremity. As might be expected, the tributary valleys had areas of injury extending farthest from the river. For example, the penetration up Deep Creek was about 10 miles. It may be significant that, insofar as they can be compared, the outer limits of injury delineated in figure 8 conform rather well with the limits of abnormal accumulations of sulfur in foliage, indicated in data presented by Katz and McCallum (12).

Much of the territory covered by injury in zone 1 is less than 2,000 feet in elevation and consists of the river terraces and benchlands

referred to in describing the area. The maximum elevation in zone 1 was about 3,400 feet; this was east of the river and about 3 miles south of the international boundary line, where downriver winds are deflected by a turn in the valley toward slopes that are among the steepest on the area. The maximum elevation in zone 3 was about 4,200 feet. The 2,000-foot contour of elevation and spot elevations shown in figure 8 aid in determining the distribution of injury with reference to elevation and the major drainage features.

The area of injury was progressively greater in 1928, 1929, and 1930, as observed in its extension farther down the main valley and farther up the tributary valleys. In 1931 the area of injury decreased; however, the decrease in area was significant only in zone 3. This recession is attributable to a sharp decrease (approximately 28 percent) in 1931 of sulfur released from the smelter stacks. There were further decreases in subsequent years.

No attempt was made to define the limits of injury after 1931. That the limits generally receded, however, is indirectly indicated in the successive amounts of acute injury in the years 1931 to 1935. These are shown in table 3, which summarizes observations made on 11 species of broadleaf trees and shrubs. The corresponding amounts of sulfur discharged (also shown in table 3) partially explain the falling off. The absence of correlation between injury and sulfur discharge after 1932 is attributable to the regulatory measures adopted in 1933 (12).

Although the limits of injury receded, and the amounts of injury in the different zones declined after 1930, what appeared to be slightly abnormal sulfur contents in the foliage were found in 1936 as far as 53 miles down the river valley from Trail, British Columbia (table 4). In the same period, injured plants were observed no farther than 30 to 40 miles down the valley, well inside the southern extremity of zone 2. As a byproduct of these observations, there is the further evidence that the rate of sulfur accumulation and not the total amount in foliage determines the incidence and extent of injury.

TABLE 3.—*Acute sulfur dioxide injury of broadleaf trees and shrubs in different years*

Year	Average sulfur discharge per month	Injured plants ¹		
		Zone 1	Zone 2	Zone 3
	<i>Tons</i>	<i>Percent</i>	<i>Percent</i>	<i>Percent</i>
1931	7, 100	49	21	21
1932	3, 400	19	8	5
1933	4, 000	18	3	3
1934	6, 200	21	5	2
1935	6, 500	19	7	4

¹ Based on observations on Douglas maple, thinleaf alder, saskatoon serviceberry, water birch, California hazel, creambush rockspirea, Pacific ninebark, Lewis mockorange, Columbia snowberry, and wild rose and willow species. Scientific names are given in tables 5 and 6.

TABLE 4.—*Sulfur content of foliage in 1936 at various distances south of the smelter*¹

Distance from the smelter (miles)	Zone	Average sulfur content, by tree or shrub species							
		Ponderosa pine	Western larch	Water birch	Douglas maple	Pacific nine-bark	California hazel	Service-berry	Lewis mock-orange
		Per-cent	Per-cent	Per-cent	Per-cent	Per-cent	Per-cent	Per-cent	Per-cent
12	1	0.27	0.75	0.73	1.15	0.54	0.93	0.80	0.59
17	1	.26	.52		.83	.38		.56	
25	1	.23		.58	1.08	.22	.43	.48	
28	1	² .20	.38				.41	.41	
39	2	² .18	.46	² .40			² .52	² .40	
47	3	² .16			² .52	² .25	² .36	² .40	² .21
53	(³)	² .08				² .13	² .18	² .15	
65	(³)	² .10	² .13	² .25	² .33	² .07	² .05	² .15	² .15

¹ Chemical analyses were made by the Bureau of Chemistry and Soils, U. S. Department of Agriculture.

² Sulfur content not accompanied by apparent injury.

³ Outside.

Relative Susceptibility of Trees, Shrubs, and Herbs

Observations were made of the relative susceptibility to foliage injury of the more common trees and shrubs and to a limited extent of the associated herbs. The order of susceptibility of the trees, measured by the amount of acute injury, is shown in table 5. The order for shrubs and herbs is given in table 6.

Western larch is placed low in the listing of susceptibility because of its ultimate resistance to killing. Actually, when its needles are young it is perhaps the most susceptible of all the plants that were critically observed. Because of this, larch trees were damaged in localities and in years when other timber species were not much affected. Nevertheless, the larch had a generally greater survival capacity than many of the others because its mature needles, like the first-year needles of most other conifers, are very tolerant to sulfur dioxide; moreover, the needles are shed each fall, so that injury or premature shedding affects the tree only through a single growing season.

The order of susceptibility of the tree species shows little agreement with that reported by Katz and associates (12). This is due partly to a difference in the criterion of susceptibility and partly to the fact that the relative amounts of injury to many species tended to differ considerably with location, year, and injury zone.

From limited observations the relative susceptibility of the evergreen coniferous trees appeared to be about the same during the dormant season and during the growing season. Data on this point were gathered in 1935, at a place where there had been four exposures to fumes from the smelter, ranging in duration from 12 to 145 hours and in average sulfur dioxide concentration from 0.30 to 97 p. p. m.

TABLE 5.—*Approximate order of susceptibility, beginning with the most susceptible, of trees in the upper Columbia River Valley to acute sulfur dioxide injury*

Conifers	Broadleaf trees ¹
Grand fir <i>Abies grandis</i> (Dougl.) Lindl.	Thinleaf alder <i>Alnus tenuifolia</i> Nutt. (24).
Subalpine fir <i>Abies lasiocarpa</i> (Hook.) Nutt.	Western paper birch <i>Betula papyrifera</i> var. <i>commutata</i> (Reg.) Fern (20).
Western redcedar <i>Thuja plicata</i> Donn.	Sitka mountain-ash <i>Sorbus sitchensis</i> Roem.
Western hemlock <i>Tsuga heterophylla</i> (Raf.) Sarg.	Water birch <i>Betula occidentalis</i> Hook.
Douglas-fir <i>Pseudotsuga menziesii</i> (Mirb.) Franco.	Douglas maple <i>Acer glabrum</i> var. <i>douglasii</i> (Hook.) Dipp. (15).
Western white pine <i>Pinus monticola</i> Dougl.	Bitter cherry <i>Prunus emarginata</i> Dougl.
Ponderosa pine <i>Pinus ponderosa</i> Laws.	Common chokecherry <i>Prunus virginiana</i> L.
Lodgepole pine <i>Pinus contorta</i> Dougl.	Blueberry elder <i>Sambucus glauca</i> Nutt.
Western larch <i>Larix occidentalis</i> Nutt.	Willow <i>Salix</i> spp. (11).
Engelmann spruce <i>Picea engelmannii</i> Parry.	Columbia hawthorn <i>Crataegus columbiana</i> Howell.
Western juniper <i>Juniperus occidentalis</i> Hook.	Black cottonwood <i>Populus trichocarpa</i> Torr. & Gray.
Pacific yew <i>Taxus brevifolia</i> Nutt.	Black hawthorn <i>Crataegus douglasii</i> Lindl.
	Quaking aspen <i>Populus tremuloides</i> Michx.

¹ Numbers in parentheses are the average percentages of trees with acute injury in zone 1 during the years 1932 to 1936.

during the preceding fall and winter. These fumigations were coupled with daytime temperatures above the critical 40° F.

The possibility of sulfur dioxide injury to coniferous trees during winter months has been a point of some disagreement. It has been observed in the vicinity of several smelters, however, where it was manifested by overwinter increases in amount of injury. Although relatively much less damaging generally than injury during the spring and summer, injury during the winter results in the death of trees if repeated in successive years. The stomata, through which the injurious amounts of sulfur dioxide must enter the leaves, are not always completely closed in winter (3). Circumstantial evidence of injury during the winter is found in the greater damage incurred by the evergreen species than by the larch and broadleaf species, which do not retain their leaves in winter.¹⁰ Only the broadleaf species, including many with relatively low tolerance to sulfur dioxide, remained alive near the smelter.

¹⁰ Sulfur dioxide may enter the stomatal openings in stems as well as in leaves; but in stems they are normally much less numerous than in leaves and therefore are a relatively minor avenue of gas entrance.

TABLE 6.—Approximate order of susceptibility, beginning with the most susceptible, of forest shrubs and herbs in the upper Columbia River Valley to acute sulfur dioxide injury

Shrubs ¹	Herbs
Pacific ninebark <i>Physocarpus capitatus</i> (Pursh) Kuntze (28).	Fireweed <i>Epilobium angustifolium</i> L.
Creambush rockspirea <i>Holodiscus discolor</i> (Pursh) Maxim. (25).	Sticky geranium <i>Geranium viscosissimum</i> Fisch. & Mey.
Lewis mockorange <i>Philadelphus lewisii</i> Pursh (23).	Queencup beadlily <i>Clintonia uniflora</i> (Schult.) Kunth.
Wild rose <i>Rosa</i> spp. (23).	Pacific sanicle <i>Sanicula graveolens</i> Poepp., syn. <i>S. nevadensis</i> var. <i>septentrionalis</i> (Greene) Math.
Saskatoon serviceberry <i>Amelanchier alnifolia</i> Nutt. (21).	Dogbane <i>Apocynum</i> sp.
California hazel <i>Corylus cornuta</i> var. <i>californica</i> (A. DC.) Sharp (18).	Slim solomonplume <i>Smilacina stellata</i> (L.) Desf., syn. <i>S. sessilifolia</i> Nutt.
Shinyleaf spirea <i>Spiraea lucida</i> Dougl.	Prickly lettuce <i>Lactuca scariola</i> L.
Red raspberry <i>Rubus strigosus</i> Michx.	Running mallow <i>Malva rotundifolia</i> L.
Whitebark raspberry <i>Rubus leucodermis</i> Dougl.	Wartberry fairybells <i>Disporum trachycarpum</i> (S. Wats.) Benth. & Hook.
Western thimbleberry <i>Rubus parviflorus</i> Nutt.	Bracted strawberry <i>Fragaria bracteata</i> Heller.
Sticky currant <i>Ribes viscosissimum</i> Pursh.	Showy aster <i>Aster conspicuus</i> Lindl.
Columbia snowberry <i>Symphoricarpos rivularis</i> Suksd., syn. <i>S. albus</i> var. <i>laevigatus</i> (Fern.) Blake (15).	Buckhorn plantain ("ribgrass") <i>Plantago lanceolata</i> L.
Redstem ceanothus <i>Ceanothus sanguineus</i> Pursh.	
Snowbrush ceanothus <i>Ceanothus velutinus</i> Dougl.	
Redosier dogwood <i>Cornus stolonifera</i> Michx.	
Smooth sumac <i>Rhus glabra</i> L.	
Western poisonivy <i>Toxicodendron radicans</i> var. <i>rydbergii</i> (Small) Kelsey & Dayton, syn. <i>T. rydbergii</i> Small.	

¹ Numbers in parentheses are the average percentages of shrubs with acute injury in zone 1 during the years 1932 to 1936.

A small number of trees and shrubs were compared for susceptibility to sulfur dioxide as artificially applied under controlled conditions. A portable fumigation chamber was used to regulate the gas dosage. The work was done in cooperation with the U. S. Bureau of Agricultural and Industrial Chemistry. The fumigations were made on 9 plots located at Wenatchee, Wash., far removed from the smelter. The plants were moved to the plots 20 months beforehand, and were well established at the time of fumigation. Two nominal sulfur dioxide concentrations were used, 1/2 part per million (actually 0.53 to .60 p. p. m.) and 1 part per million (actually 1.00 to 1.08

p. p. m.). The fumigating was done late in June, over a period of 12 days. The leaves at this time were not all mature, particularly those of the trees, and the mature ones were still relatively succulent.

Results are given in table 7. The relative order of resistance indicated for the shrubs is similar to that observed for the same plants in the field (table 6). This, however, was not true of the trees, which showed considerably less consistency than the shrubs with respect to the effect of length of fumigation. Probably this can be attributed to the more variable maturity of the tree foliage.

TABLE 7.—*Acute injury to small transplants produced by artificial fumigation*

INJURED LEAVES					
Shrub or tree	Sulfur-dioxide treatment ¹				
	$\frac{1}{2}$ part per million—7 hours	1 part per million			
		4 hours	5 hours	6 hours	7 hours
	Percent	Percent	Percent	Percent	Percent
Pacific ninebark.....	50 (33)	38 (8)	79 (2)	75 (9)	94 (2)
Creambush rockspirea.....	72 (2)	25 (6)	-----	62 (3)	-----
Saskatoon serviceberry.....	8 (1)	15 (3)	65 (1)	42 (8)	94 (3)
California hazel.....	0 (2)	10 (3)	-----	38 (2)	-----
Lewis mockorange.....	0 (11)	-----	2 (1)	4 (3)	-----
Columbia snowberry.....	0 (22)	0 (3)	0 (1)	2 (11)	9 (3)
INJURED SHOOTS					
Western larch.....	35 (4)	100 (1)	86 (2)	-----	-----
Lodgepole pine.....	0 (2)	33 (2)	0 (2)	-----	50 (2)
Western white pine.....	-----	-----	-----	-----	33 (3)
Ponderosa pine.....	100 (1)	-----	0 (2)	100 (1)	20 (5)
Douglas-fir.....	-----	1 (4)	0 (4)	66 (3)	0 (6)

¹ The number of plants fumigated in each case is shown in parentheses.

During the fumigations with 1 part per million of sulfur dioxide, notes were taken of the earliest appearance of acute injury. Compared on this basis, the ninebark, rockspirea, and western larch showed injury at the end of 3 hours. (in other experiments a comparable result was obtained with alfalfa), Douglas-fir and serviceberry at the end of 3½ to 4 hours, lodgepole pine at the end of 5 hours, and ponderosa pine, white pine, and snowberry at the end of 6 hours.

Injury was noted in all of the plants that were fumigated for 6 hours with 1 part per million of the gas, and some were injured by 4 hours' fumigation (table 7). Only the more sensitive plants were injured by ½ part per million maintained for 7 hours. As a rule, plants that showed little or no injury immediately after harmful fumigation developed acute markings from 2 to 4 days later. The minimum concentration of sulfur dioxide required to injure coniferous

trees appears to be not less than about 0.25 parts per million. Katz and McCallum (12) found that during the growing season pine and Douglas-fir could tolerate this amount continuously for 450 hours.

In a few supplementary tests on ponderosa pine seedlings grown from seed planted in the same season, the seedlings were found to be much more susceptible to sulfur dioxide than the larger, transplanted trees. For example, 80 percent of the seedlings were severely injured by a 4-hour treatment with 1 part per million of sulfur dioxide, as contrasted with 20 percent of the larger trees when treated at the same concentration for 7 hours.

Reproduction of Timber

One of the more conspicuous features of the areas affected by sulfur dioxide was the scarcity of timber reproduction. This appeared to be a result both of reductions in cone crops and of nonsurvival of seedlings that became established. General observations and notes taken from 1928 to 1933, and Forest Service data on cone production in parts of the Colville National Forest bordering the injury zones, indicated that in zone 1 there were only a few cones borne by western larch, lodgepole pine, and Douglas-fir, whereas on areas outside the injury zones cone production by these species was fair to good. During the same period, ponderosa pine in zone 1 produced virtually no cones. In zones 2 and 3 the cone crops of all species were progressively better but still inferior to those in the outside areas.

No reproduction was observed in zones 1, 2, or 3, except for a slight amount of western larch in zone 3.

In 1936, when the rate of sulfur dioxide dissemination was between 60 and 70 percent of the peak rates prevailing 5 to 10 years earlier, formal appraisals of ponderosa pine cone production were made on more than 100 plots distributed largely in the main river valley. The data are summarized in table 8. A better cone crop was produced in zones 1, 2, and 3 in 1936 than in 1932 and 1933. Nevertheless, the trend of increasing production from zone 1 to the outside remained, indicating that sulfur dioxide had continued to be detrimental to

TABLE 8.—*Cone production by ponderosa pine trees of bearing age in 1936*¹

Zone	Plots examined	Trees examined	Trees with—		
			No cones	Few cones	Many cones
	<i>Number</i>	<i>Number</i>	<i>Percent</i>	<i>Percent</i>	<i>Percent</i>
1.....	33	7,670	81	13	6
2.....	20	5,274	46	20	34
3.....	30	5,142	27	17	56
Outside.....	30	7,742	16	17	67

¹ Average monthly rates of sulfur discharge by the smelter during the period of cone maturation were 6,200 tons in 1934, 6,500 tons in 1935, and 6,100 tons in 1936.

cone crops even in zone 3. Cone production in 1936 and in other years was, of course, not necessarily evidence of harmful effects in the particular years of the observations. Where two or more years are required for the cones to mature, as in the case of ponderosa pine, the maturation process might well be arrested in the first year.

To determine the effect of sulfur dioxide in the different zones on seedlings that might be produced, despite a scarcity of cones, seedling plots were established with ponderosa pine and Douglas-fir seed gathered outside the area of injury. Some of the screens on the plots were damaged and the stocking depleted by rodents. Additional depletion resulted from drought, particularly in zone 3 and in the outside zone. Observations made on the remaining stock, in 1932 and 1933, 1 to 2 years after seeding, are shown in table 9. Sulfur dioxide injury was distinguished by the reddish or brown color produced in the needles, in contrast with the straw color produced by drought.

TABLE 9.—*Injury to 1- to 2-year-old trees grown from seed planted in the different zones*

Year and zone	Ponderosa pine			Douglas-fir		
	Plots	Trees	Trees either injured or killed	Plots	Trees	Trees either injured or killed
1932:	<i>Number</i>	<i>Number</i>	<i>Percent</i>	<i>Number</i>	<i>Number</i>	<i>Percent</i>
1-----	10	77	90	10	489	84
2-----	3	13	31	3	139	14
3-----	(¹)	(¹)	(¹)	2	171	0
Outside----	(¹)	(¹)	(¹)	3	136	0
1933:						
1-----	8	948	98	8	486	78
2-----	4	45	45	4	76	37
3-----	1	24	0	1	9	0
Outside----	4	63	0	4	33	0

¹ Plots damaged by rodents.

The percentages of injured or killed seedlings indicate that seedling mortality as well as poor cone crops could account for the lack of reproduction in zone 1 in 1932 and 1933. This was partially the case for zone 2 also, but not for zone 3, in which there was no clear evidence of seedling injury by sulfur dioxide. Seedling mortality in zones 2 and 3 presumably was a larger factor in earlier years, however, because of larger prevailing amounts of sulfur dioxide. In 1932 and 1933, for example, the amounts liberated by the smelter were less than half (3,400 and 4,000 tons per month) those liberated in the years 1926 to 1930 (8,900 to 9,900 tons per month), when the most extensive general injury occurred.

In 1948, considerable, though somewhat spotty, reproduction of ponderosa pine was found in the northern half of zone 1. Some of the young trees were as old as 15 years, but most of them were not more

than 6 years old. The dates of seed germination denoted by these two ages were near the time when the sulfur output was being restricted to no more than 6,500 tons per month and supplemental regulatory measures were first instituted (1933), and when it was first curtailed to less than 2,000 tons per month (1944).

Mortality of Ponderosa Pine and Douglas-fir

The relative numbers of dead trees and of trees in various states of decadence were ascertained in 1929-35 for ponderosa pine and Douglas-fir in the different zones. To accomplish this, census plots were measured and staked out in selected places more or less uniformly distributed along the main river valley and in the larger tributary inlets. Plots observed in the same series of years and dealt with as a unit will be called a census group. As a census group consisted of the same plots from year to year, it furnished a good basis for discerning trends of reduced vigor and mortality.

In order that the condition of trees in the respective zones might be comparable except for amounts of sulfur dioxide injury, census groups to be compared were selected for uniformity of tree size and growing conditions. All of the trees were young, and they occurred in scattered open stands where there could be no suppression by crowding. Localities with trees damaged by fire were not included. The soil in all cases was alluvial in type, as described for the river benches.

In classifying the trees, the ponderosa pines with scant and largely discolored foliage were called *dying*; those with one year's foliage still green were designated as *poor*; those with more than 1 but less than 2 full years' green foliage as *fair*; and those with 2 or more years' green foliage as *good*. The Douglas-firs with scant and largely discolored foliage were designated as *dying*, those with more than 1 but less than 2 full years' green foliage as *poor*, those with 2 to 3 years' green foliage as *fair*, and those with more than 3 years' green foliage as *good*.

The bases for the census data on ponderosa pine saplings are shown in table 10. The data are presented in figures 9 and 10. In all three of the zones affected by sulfur dioxide, the proportion of dead ponderosa pine saplings increased rather markedly from 1929 to 1931 (fig. 9). It continued to increase thereafter but at a declining rate until in 1935 dying of trees on the census plots had virtually ceased. In general, the proportion of dead trees, and, to a lesser extent, the rate of dying, was greatest in zone 1 and least in zone 3. Judging from the small proportion of dead trees at all times in the outside zone, practically all of the mortality may be attributed to the influence of sulfur dioxide.

The identity of individual trees was not recorded; therefore, the rate of deterioration of trees of a particular *initial* vigor class (e. g., in 1929) could not be determined. The net change from period to period in the proportion of the total stand made up by a vigor class could, of course, be ascertained. Such information for the sapling ponderosa pines located in zone 1 is presented in figure 10. It is apparent from the curve values that some of the poor trees passed

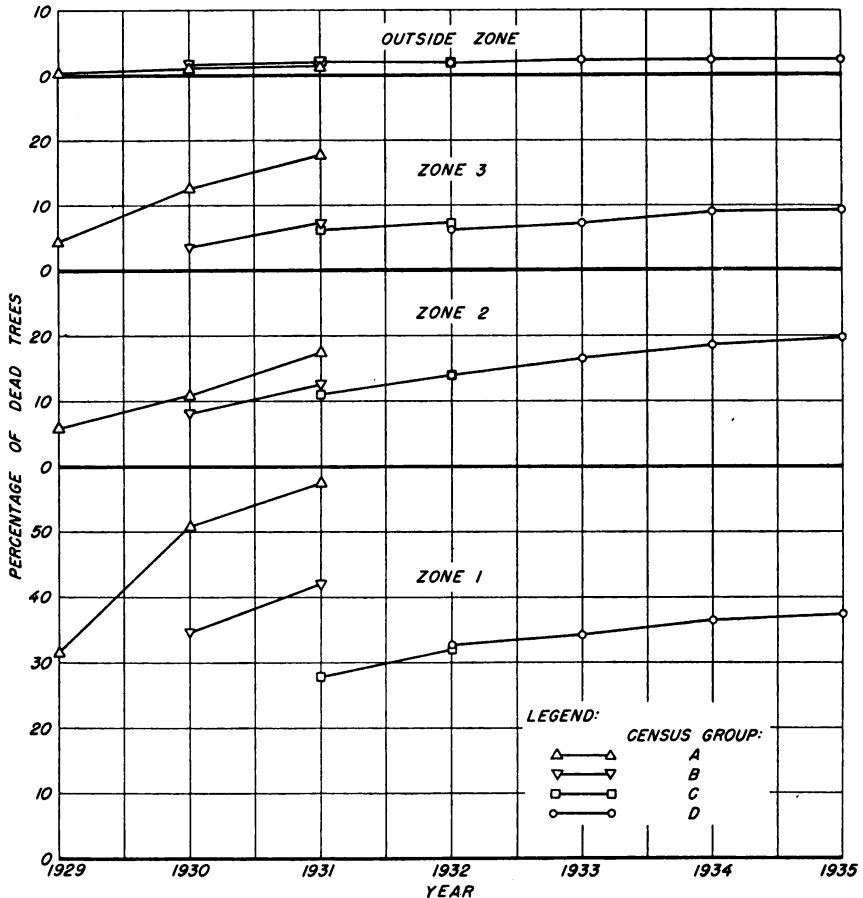


FIGURE 9.—Relative numbers of dead ponderosa pine saplings observed in the different injury zones in the upper Columbia River Valley, Wash., 1929 through 1935.

rapidly through the dying class and were killed within a single year, especially in 1930 and 1931. This is indicated by the inadequate number of dying trees in some of the years to account for the increase in number of dead ones in a following year.

During the period of lesser injury, after 1931, little change took place in the proportions of poor and dying trees, whereas the proportion of fair trees decreased at about the same rate as the proportion of dead trees increased. Such a balance of classes suggests that during this period the rate of retrogression of trees from each of these classes to the one next lower was nearly the same.

The proportion of good trees remained virtually constant throughout the entire period except for a decline in 1930 from the 1929 level. In fact, there was little room for further decline since the residual stand of good trees in 1930 was less than 2 percent. Of chief significance here is the persistence in good condition of these few trees.

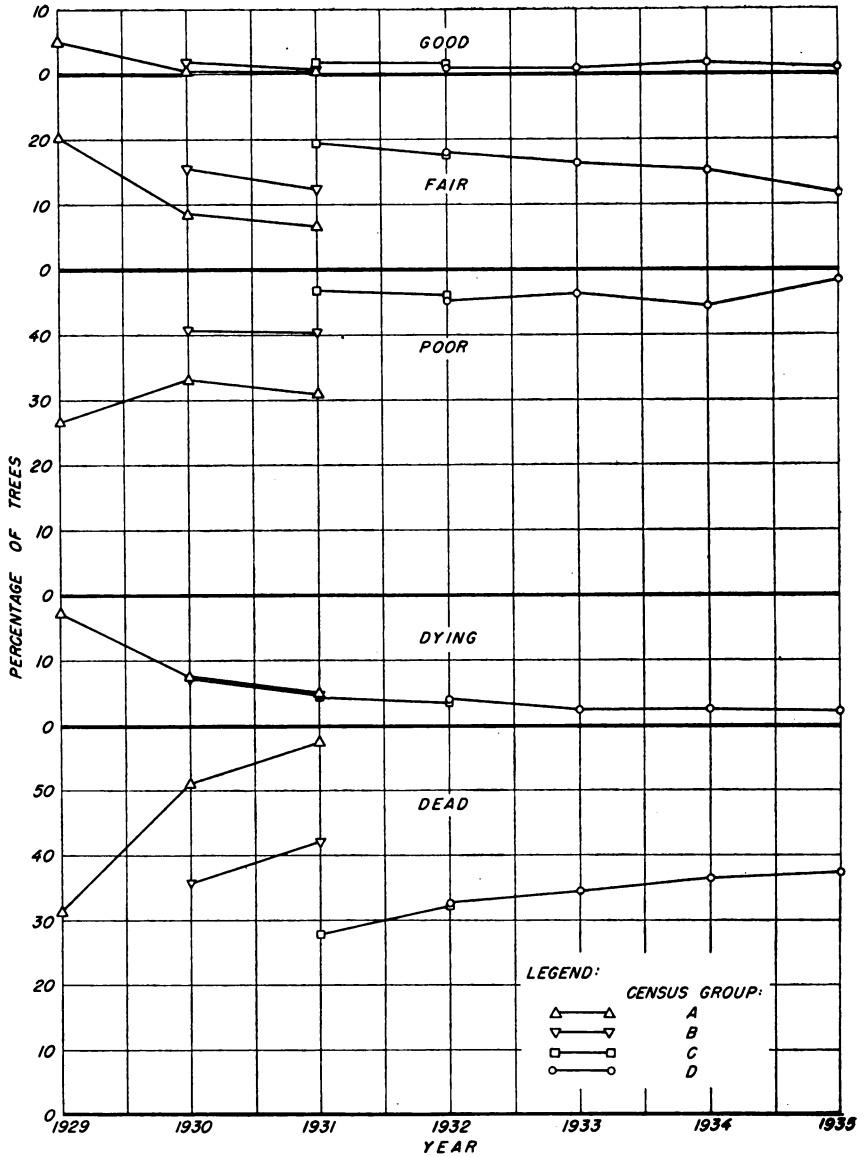


FIGURE 10.—Relative numbers of ponderosa pine saplings in various conditions of vigor observed in zone 1 in the upper Columbia River Valley, Wash., 1929 through 1935.

Such trees were not confined to a few plots that might have been protected by peculiarities of the environment but, rather, they occurred on two-thirds of the plots. It must be concluded, therefore, that they were morphologically or physiologically constituted to tolerate greater exposure to sulfur dioxide than the other trees.

TABLE 10.—*Basis of census for appraising the condition of ponderosa pine saplings*

Census group	Years in which records were taken	Injury zone	Plots	Acres	Trees
			Number	Number	Number
A.-----	1929 to 1931.-----	1	8	23.75	557
		2	8	10.75	527
		3	6	3.50	392
B.-----	1930 and 1931.-----	(¹)	7	2.00	426
		1	19	27.75	1,443
		2	19	15.50	1,535
C.-----	1931 and 1932.-----	3	18	8.75	1,303
		(¹)	20	8.00	1,624
		1	14	6.25	1,290
D.-----	1932 to 1935.-----	2	16	6.25	1,304
		3	17	8.00	1,225
		(¹)	19	7.25	1,507
		1	12	5.75	1,093
		2	15	5.50	1,234
		3	16	7.50	1,172
		(¹)	18	7.00	1,372

¹ Outside.

Similar data were obtained for larger (10 to 18 inches d. b. h.) ponderosa pine trees, but only in 1930 and 1931 (table 11). The vigor and mortality situation of the larger trees appears in essential features to have been much like that of the saplings in the same years (census group B, figures 9 and 10). Apparently ponderosa pines become more tolerant of sulfur dioxide as they grow out of the seedling stage, but beyond the sapling stage they do not continue to become more tolerant.

The census data for the Douglas-fir saplings are given in table 12. In 1929 the proportion of dead firs in the respective zones was comparable to that found in census group A of the ponderosa pine saplings (fig. 9). Further losses in zone 3, in 1930 and 1931, were much the same, but in zones 1 and 2 losses were much higher for Douglas-fir than for ponderosa pine. For example, in 1931, 90 percent of the firs were dead in zone 1 and 40 percent in zone 2, in contrast with corresponding values of 57 and 17 percent for the sapling ponderosa pine. These larger mortality figures for the fir are reflected in a lower representation of trees in the good, fair, and poor classes and a somewhat higher representation in the dying class.

In zone 1 there were no sapling firs in good condition in 1930 and 1931, and no more than 2 percent in fair condition. Thus individual trees especially tolerant to sulfur dioxide were not present among the Douglas-firs as they were among the pines.

Height Increment of Ponderosa Pine

One of the more precisely measurable variables of the pine trees was their annual height increment. This was denoted for the current year's growth by the length of the terminal shoot at the end of the growing season and for preceding years by the lengths of the inter-

TABLE 11.—*Ponderosa pine trees (10 to 18 inches d. b. h.) in various conditions of vigor in 1930 and 1931*

Condition of trees	Year	Trees injured ¹			
		Zone 1	Zone 2	Zone 3	Outside zone
		<i>Percent</i>	<i>Percent</i>	<i>Percent</i>	<i>Percent</i>
Good.....	1930	2	5	11	40
	1931	1	4	11	36
Fair.....	1930	16	47	59	50
	1931	14	34	49	52
Poor.....	1930	42	34	22	7
	1931	36	40	27	9
Dying.....	1930	8	4	3	1
	1931	6	6	4	1
Dead.....	1930	32	10	5	2
	1931	43	16	9	2

¹ Numerical basis of records:

Zone:	Plots	Acres	Trees
1.....	24	480	2,628
2.....	12	225	1,136
3.....	26	375	2,120
Outside.....	24	320	2,011

TABLE 12.—*Douglas-fir saplings in various conditions of vigor, 1929-31*

Condition of trees	Year	Trees injured ¹			
		Zone 1	Zone 2	Zone 3	Outside zone
		<i>Percent</i>	<i>Percent</i>	<i>Percent</i>	<i>Percent</i>
Good.....	1929	2	22	46	67
	1930	0	10	32	66
	1931	0	7	18	71
Fair.....	1929	9	32	34	25
	1930	2	24	36	24
	1931	0+	16	36	18
Poor.....	1929	17	28	11	5
	1930	9	29	17	6
	1931	6	31	23	6
Dying.....	1929	40	9	6	1
	1930	10	7	6	1
	1931	4	6	5	0+
Dead.....	1929	32	9	3	2
	1930	79	30	9	3
	1931	90	40	18	5

¹ Numerical basis of records:

Zone:	Plots	Acres	Trees
1.....	6	19.50	476
2.....	6	11.25	443
3.....	6	5.00	331
Outside.....	6	2.00	461

nodes between the whorls of lateral branches. Advantage was taken of the opportunity thus presented to investigate the effect of sulfur dioxide in the different zones on the height increment of ponderosa pine trees.

Measurements were made of a large number of trees scattered throughout the same general area included in the mortality and vigor census. Also, the same precautions were taken to select trees of comparable character and with similar environments. Trees injured by causes other than sulfur dioxide were rejected. Crooked trees were rejected except for the first group that was measured, in 1930. Trees of two nominal classes were considered: Sapling trees, ranging in height from 8 to 20 feet, and pole-size trees, ranging in height from 25 to 50 feet. In all cases, the measured trees were dominants in their group. For measuring, the small trees were pulled over with a pole hook; the larger ones had to be felled. All data were obtained in the fall. Basic information on these trees is given in table 13. The pole-size trees were measured in two different years, for growth in the years 1923 through 1931. The sapling trees were measured in 7 different years, for growth in the years 1926 through 1936.

The average measurements of annual height increment of pole-size and sapling trees in each zone are given in figure 11. Also included are the amounts of sulfur discharged by the smelter and the January-August precipitation in zone 1 and in the outside zone.¹¹

In drawing conclusions from the curves of figure 11, the height-increment trends in the outside zone are taken as the pattern of growth that might be expected in the other zones were it not for modifications induced by the presence of sulfur dioxide. Moreover, such modifications are considered to be a reflection of injury occurring in the year preceding the one in which the modification appeared. These assumptions seem reasonable in view of two features of the outside height-increment and the precipitation curves:

1. The trends of precipitation were similar in zone 1 and in the outside zone; therefore, zonal differences in trends of height increment are not attributable to precipitation differences.

2. Variations in precipitation from year to year usually were attended by similar and equally pronounced variations in height increment in the outside zone. The changes in height increment lagged one year behind the changes in precipitation. This is typical and is due to the dependence of tree height growth on foodstuffs stored up in the previous season. For the same reason, a lag in the effect of sulfur dioxide on height growth would be expected, particularly inasmuch as the young shoots are comparatively tolerant of sulfur dioxide.

Height growth in the years 1923 through 1926 was for the most part as irregular, and therefore as responsive to precipitation differences, in zones 1, 2, and 3 as in the outside zone. Thus there is no evidence in these growth responses that sulfur dioxide had been pres-

¹¹ The precipitation shown for zone 1 is the average of amounts reported by the U. S. Weather Bureau for Laurier (Kettle River Valley) and Northport, Wash. That for the outside zone is the average for Kettle Falls and Colville, Wash. Precipitation totals are limited to the period January through August because of evidence obtained by Lathe and McCallum (12) that growth on the area is most closely correlated with precipitation in that period.

TABLE 13.—*Basis of the height-increment measurements on ponderosa pine trees*

POLE-SIZE TREES							
Observation group	Years covered by the records	Zone of injury	Plots	Trees	Condition of the trees		
					Poor	Fair	Good
			Number	Number	Per-cent	Per-cent	Per-cent
A.....	1923 to 1930.....	1	22	132	36	60	4
		2	17	102	25	62	13
		3	21	126	11	68	21
		(¹)	20	120	5	49	46
B.....	1924 to 1931.....	1	27	135	29	64	7
		2	25	125	12	56	32
		3	24	120	6	34	60
		(¹)	24	120	2	22	76
SAPLING TREES							
C (crooked trees included).	1926 to 1930.....	1	32	640	60	36	4
		2	17	340	34	54	12
		3	14	280	18	43	39
		(¹)	17	340	4	29	67
D.....	1926 to 1931.....	1	31	620	50	46	4
		2	17	340	14	47	39
		3	25	500	9	32	59
		(¹)	23	460	1	26	73
E.....	1926 to 1932.....	1	33	660	41	48	11
		2	18	360	14	54	32
		3	24	480	10	40	50
		(¹)	25	500	1	24	75
F.....	1927 to 1933.....	1	25	500	48	41	11
		2	25	500	21	43	36
		3	25	500	6	34	60
		(¹)	25	500	4	18	78
G.....	1928 to 1934.....	1	25	500	43	45	12
		2	25	500	18	51	31
		3	25	500	10	38	52
		(¹)	25	500	4	28	68
H.....	1929 to 1935.....	1	20	400	29	40	11
		2	20	400	7	44	29
		3	20	400	2	27	51
		(¹)	20	400	1	16	83
I.....	1930 to 1936.....	1	20	400	13	51	36
		2	20	400	3	40	57
		3	20	400	0	24	76
		(¹)	25	500	0	7	93

¹ Outside.

ent in quantities sufficient to appreciably affect height growth in any of the zones prior to 1926. A sulfur dioxide effect in years subsequent to 1926 is indicated, however, since height growth in zones 1, 2, and 3 showed generally less response to precipitation differences as compared with the response in the outside zone. Moreover, the growth

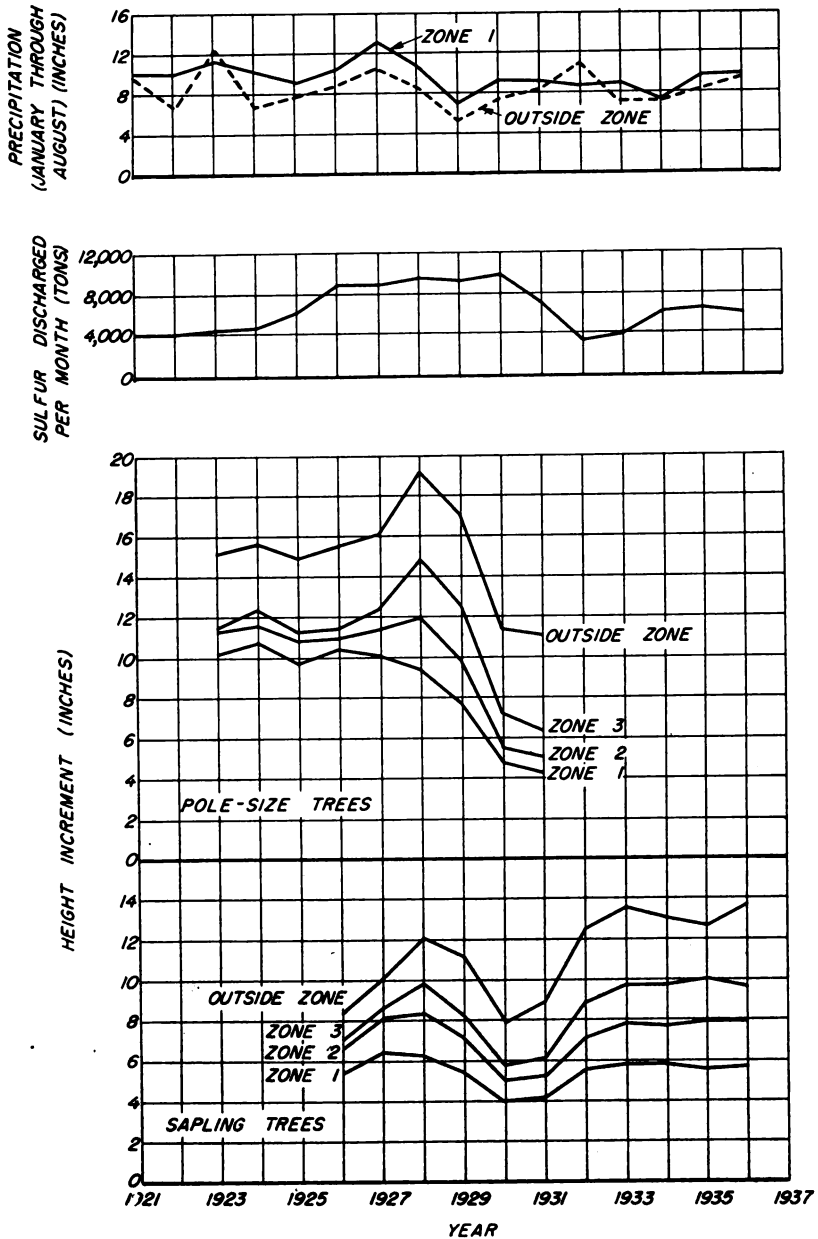


FIGURE 11.—Annual height increment of ponderosa pine trees in the upper Columbia River Valley, Wash.

response tended to diminish progressively from zone 3 to zone 1. This is particularly apparent in the relative growth responses immediately following the precipitation extremes occurring in 1927 and 1929. The influence of sulfur dioxide on height growth appears to have con-

tinued in zones 1, 2, and 3 until at least 1932, but the situation thereafter is obscure in the growth trends.

The effect of the sulfur dioxide on height growth in the different zones and years cannot be directly appraised from figure 11. Height increments shown in the figure, although progressively smaller from the outside zone to zone 1, are only partially influenced by sulfur dioxide. A complicating factor in this connection was a tendency for the river valley soil to be progressively more fertile from zone 1 to the outside zone farther down the river. For a quantitative appraisal of any retardation in height increment caused by sulfur dioxide, it would be necessary to eliminate from the comparisons of growth-rate level between zones 1, 2, and 3 and the outside zone all differences attributable to natural environmental factors. An attempt to do this was made by adjusting the level of growth for the outside zone so that it coincided on the average with the level for the respective injury zones during the years 1923 to 1926, which seemed to be free of significant sulfur dioxide influence. Separate adjustments were made for the pole-size and the sapling trees; in the latter case the adjustments could be made only on the basis of growth in 1926. The results are shown in figure 12. The curves for zones 1, 2, and 3 are exactly like those of figure 11, but those for the outside zone are reduced in level by a uniform percentage in each case, to bring about the coincidence just mentioned. The curve for the pole-size trees in the outside zone, for example, was lowered from its true level by 33.5 percent at all points to bring it into coincidence with the 1923-26 portion of the curve for the pole-size trees of zone 1. The corresponding reduction for the sapling trees was 35.7 percent.

From the comparisons thus afforded it might be concluded that there was definite retardation of height growth in 1928 (resulting from injury in 1927) in zones 1 and 2, and about a year later in zone 3, and that the retardation continued without appreciable abatement in all three zones through 1936, the last year of height-growth observations. As would be expected, the retardation was greatest in zone 1 and least in zone 3.

Average reductions in annual height increment of sapling and pole-size trees from 1928 to 1936 are indicated to be about 2.5 inches in zone 1, 2.0 inches in zone 2, and 1.1 inches in zone 3. The only evidence of correlation between the amount of retardation by sulfur dioxide and the rate of sulfur dioxide discharge was the retardation, within a year or two after the sulfur dioxide discharge was increased to nearly 9,000 tons per month (1926). There was no correlation between retardation and precipitation. The slowest growth during the investigations occurred in 1930 and 1931, following the beginning of a dry period in 1928 and semidrought conditions in 1929. The indicated degree of retardation in 1930, nevertheless, was similar to that following wetter years.

Retardation of height increment persisted as late as 1936 in spite of reduced sulfur dioxide discharge by the smelter and the institution of full regulatory measures in 1933. This conclusion is reached from the fact that a considerable margin of error can be assumed in the placement of the curve for the outside zone in figure 12 without eliminating the indication of such persistence of retardation in at least

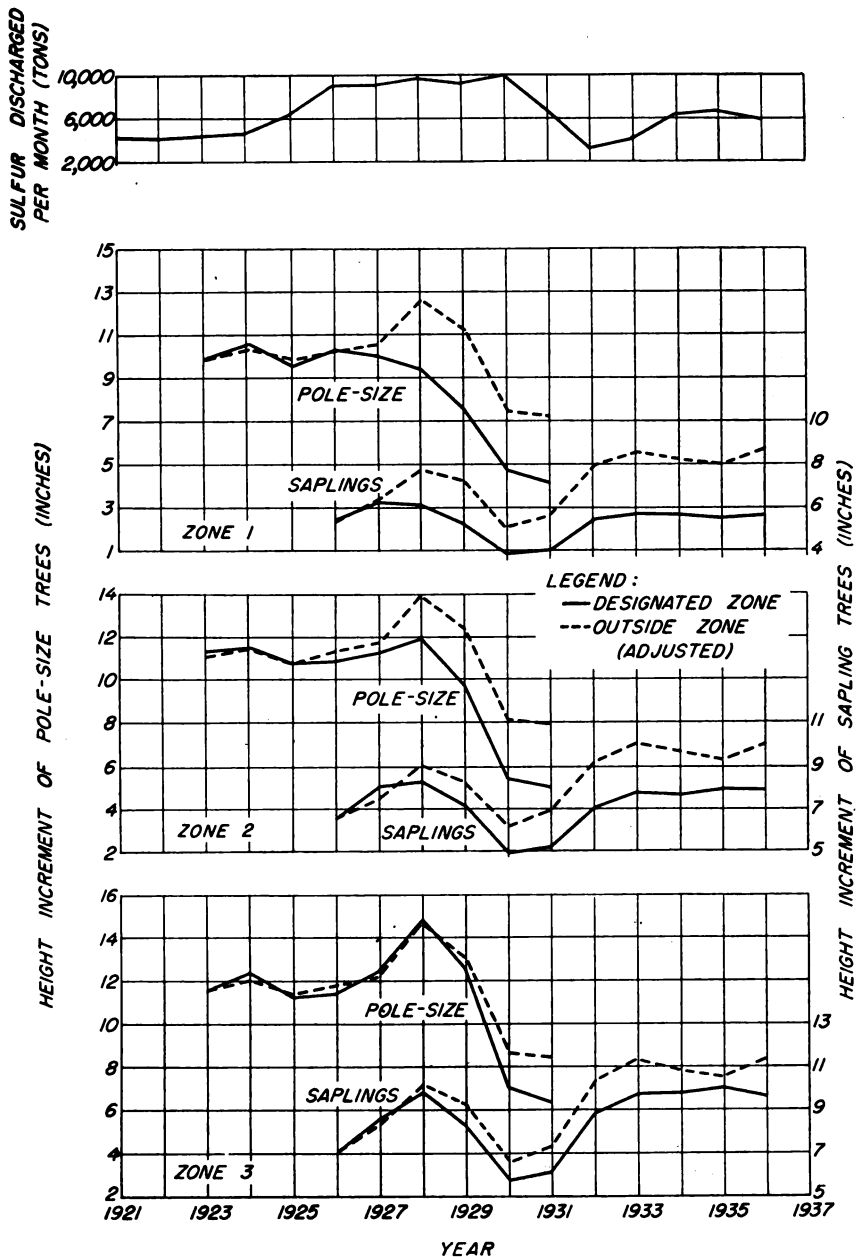


FIGURE 12.—Comparison of annual height increment of ponderosa pine trees in the upper Columbia River Valley, Wash., and theoretical increment that could have been expected without sulfur dioxide injury. Theoretical increment was formulated by adjusting the actual increment for the outside zone shown in figure 11. Each curve of expected increment is proportional to the corresponding curve of actual increment shown in figure 11 for the outside zone.

zones 1 and 2. The prolonged effect on height increment may be explainable by the fact that as compared with the outside zone, a significantly greater percentage of the measured trees were still in no better than fair condition (i. e., with no more than 1 to 2 years' green foliage) in 1934, 1935, and 1936 (table 13). Needle casting 3 or more years prematurely would reduce the food manufacturing capacity of the trees. The length of time over which needle losses of this sort might affect height growth would depend on the relative contribution to growth made by the older needles.

Diameter Increment of Ponderosa Pine

Another rather precisely measurable variable of the tree growth was that of diameter increment. Since the annual increment is shown by growth rings where there are distinct growing seasons, it was possible to ascertain the increment in any specific year preceding the current one.

Ponderosa pine trees were used for this phase of the study also. These were located on the same general areas as the trees measured for height growth, and they were selected with the same precautions to minimize insofar as practicable the influence of variables of environment, other than sulfur dioxide. Except for a relatively small number, the trees were felled and the measurements were made on disks sawed from the trunk at a height of about 2 feet. The disks were taken after the end of the growing season or, if not, the current year's growth was not measured. Measurements were made with the aid of a compound microscope equipped with a sliding stage and wheel micrometer. The face of the disk to be measured was sanded smooth. Where disks were not used, samples were taken in the form of cores obtained with a Swedish increment borer. The cores were smoothed along one side with a microtome knife.

Two sets of diameter-increment measurements were obtained, using disks for the first and increment cores for the second. The basis of the first set is given in table 14. The measurements themselves are summarized in figure 13. The curves for the outside zone in figure 13 have been adjusted as previously described for their adjustment in connection with the height-increment study. Here also the purpose of the adjustment was to delineate growth rates that might have occurred in zones 1, 2, and 3, respectively, had sulfur dioxide not been present. Accordingly, it was made so as to bring the paired curves of figure 13 to the same average level for the period 1916 through 1924, when the rate of sulfur discharge did not appreciably exceed 5,000 tons per month.¹² The measured trees in the outside zone had grown faster than the others; therefore, the adjustment in each case was downward. The reduction factors were as follows: Zone 1, 32.5 percent; zone 2, 28.4 percent; and zone 3, 21.9 percent. This diminished the indicated growth superiority of the outside trees and thereby should furnish conservative evidence of suppressed diameter growth.

¹² This level of sulfur output was approximately half the peak amounts discharged a few years later. As has been noted, it did not significantly affect height increment.

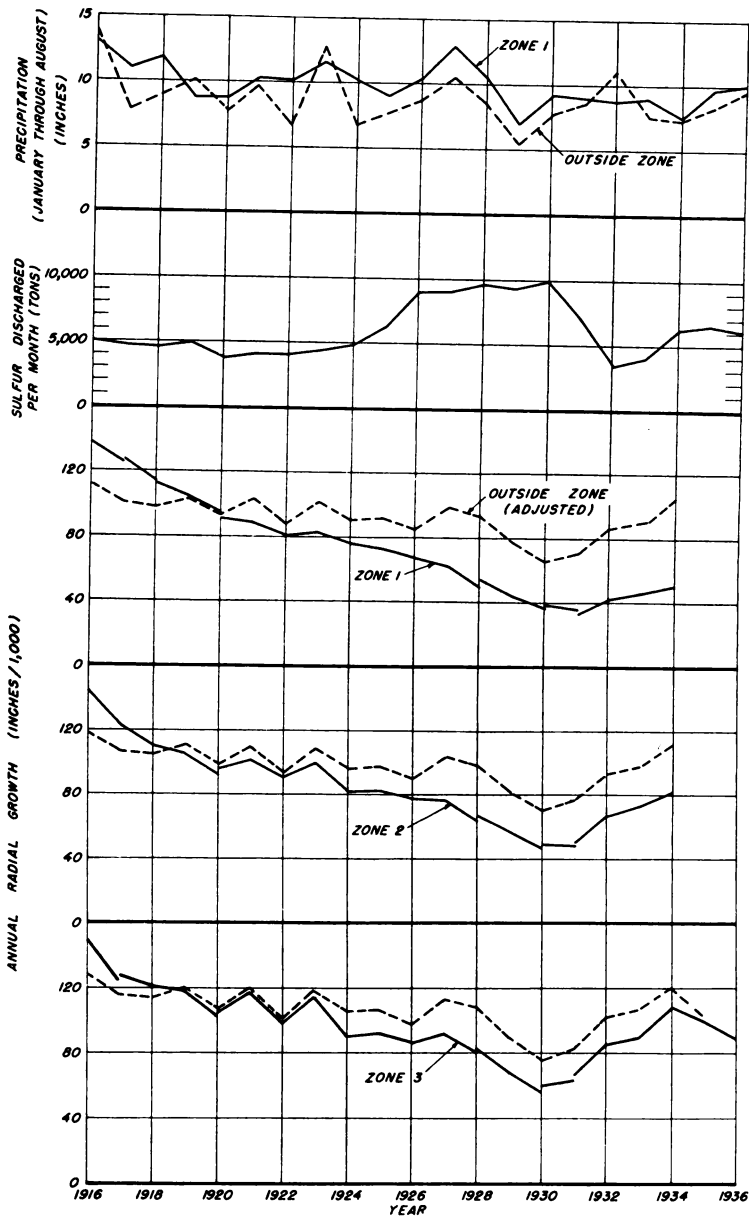


FIGURE 13.—Comparison of annual radial growth of ponderosa pine trees in the upper Columbia River Valley, Wash., and theoretical growth that could have been expected without sulfur dioxide injury. The theoretical growth was formulated by adjusting the actual growth for the outside zone. Each curve of expected increment is proportional to the curve of actual increment (not shown) for the outside zone, as explained in the text. Segments of the interrupted curves for zones 1, 2, and 3 represent averages based on different groups or groupings of trees; groupings were arranged so that the same series of growth years were represented by all trees in the group.

TABLE 14.—Basis of the first set of diameter-increment measurements on ponderosa pine trees

Zone	Year of study	Plots Number	Trees Number	Average diameter at breast height Inches	Average height Feet	Average age at stump Years	Average percentage of trees—		
							Poor ¹ Percent	Fair ¹ Percent	Good ¹ Percent
1.	1929	14	84	5.1	31	29	63	35	2
	1930	22	132	5.9	33	34	36	60	4
	1931	27	135	5.3	28	32	29	64	7
	1934	12	240	8.0	---	---	45	53	2
Total or average		---	591	² 6.5	---	---	41	55	4
2.	1929	14	84	5.1	30	27	35	49	16
	1930	17	102	6.3	34	33	25	62	13
	1931	25	125	6.3	32	34	11	55	32
	1934	12	240	7.7	---	---	23	56	21
Total or average		---	551	² 6.7	---	---	23	56	21
3.	1929	8	48	5.5	30	31	21	65	14
	1930	21	126	6.5	33	32	10	68	22
	1931	24	120	6.8	34	32	5	37	58
	1934	12	240	8.1	---	---	7	46	47
	1936	20	400	9.5	---	---	0	0	100
Total or average		---	934	² 8.2	---	---	5	29	66
Outside.	1929	15	90	6.3	32	29	0	50	50
	1930	20	120	6.7	33	28	5	49	46
	1931	24	120	6.8	30	30	2	22	76
	1934	12	240	8.3	---	---	3	27	70
	1936	26	520	9.9	---	---	0	0	100
Total or average		---	1,090	² 8.6	---	---	1	18	81

¹ Poor—1 year's foliage still green; fair—more than 1 but less than 2 full years' green foliage; good—2 or more years' green foliage.
² Weighted according to number of trees in the respective plots.

In figure 13, the diameter growth of ponderosa pine trees in zone 1 appears to have been retarded by sulfur dioxide as early as 1921. This is suggested not only by the relatively slower growth in 1921 but also by the failure of the zone 1 trees to respond to increased precipitation in 1921. Similarly, both the differences in growth rate and a comparative lack of response to fluctuations in precipitation indicate a continuing retardation of diameter growth in zone 1 until as late as 1934, when the last measurements of this set of trees were made. A marked retardation occurred in 1927, presumably as a result of the greater than 40 percent increase in sulfur dioxide discharge beginning in 1926.

Allowing for a margin of error in the adjusted location of the outside zone curve, indications are that retardation of diameter growth in zones 2 and 3 commenced not later than 1924 and that it continued in these zones also through 1934. Little or no retardation is indicated for zone 3 in 1935. As in zone 1, the amount of retardation increased sharply in 1927.

The retardation in general was greatest in zone 1 and least in zone 3. In terms of radial growth, the average indicated reductions in zones 1, 2, and 3 during the years 1924 through 1934 were 0.033 inch, 0.022 inch, and 0.017 inch respectively. The corresponding percentage reductions are 38, 24, 17.

There is considerable similarity between the diameter growth curves in figure 13 and corresponding portions of the height growth curves in figure 12. Perhaps the major point of difference is the appreciably earlier inception of retardation indicated for the diameter growth. The significance of this difference, if it is a real one, is not clear. It may be traceable to the fact that the height increment of pines depends in large measure on reserve foods stored during the previous year whereas diameter increment apparently is influenced by both the reserve foods and those currently produced.¹³

In 1949 Scheffer observed that trees even in the northernmost part of zone 1 were in excellent condition and apparently had not been importantly affected by sulfur dioxide since 1940 at the latest. A few additional trees in the different zones were measured for diameter increment, and growth rates in the years 1940-48 rather than those in years preceding injury were used as a basis for adjusting the outside-zone growth rates to serve as an approximation of normal increments to be expected in earlier years in zones 1, 2, and 3. The method of adjustment was similar to that used in the previous studies of height growth and diameter growth. Since the two bases of comparison were nearly 2 decades apart, evidence of growth suppression given in one case is essentially independent of that given in the other.

This second set of growth measurements is summarized in figure 14. If allowance is made for the small number of trees represented, the trends shown by figure 14 for zones 1 and 2 are similar in most respects to those in figure 13. In both instances, indications of retarded growth first appear in 1924 or shortly before, and they continue through 1934. The retardation indicated by the second set of measurements for zone 3 is considerably smaller than that indicated by

¹³ Information furnished by Division of Silvicultural Relations, Forest Products Laboratory.

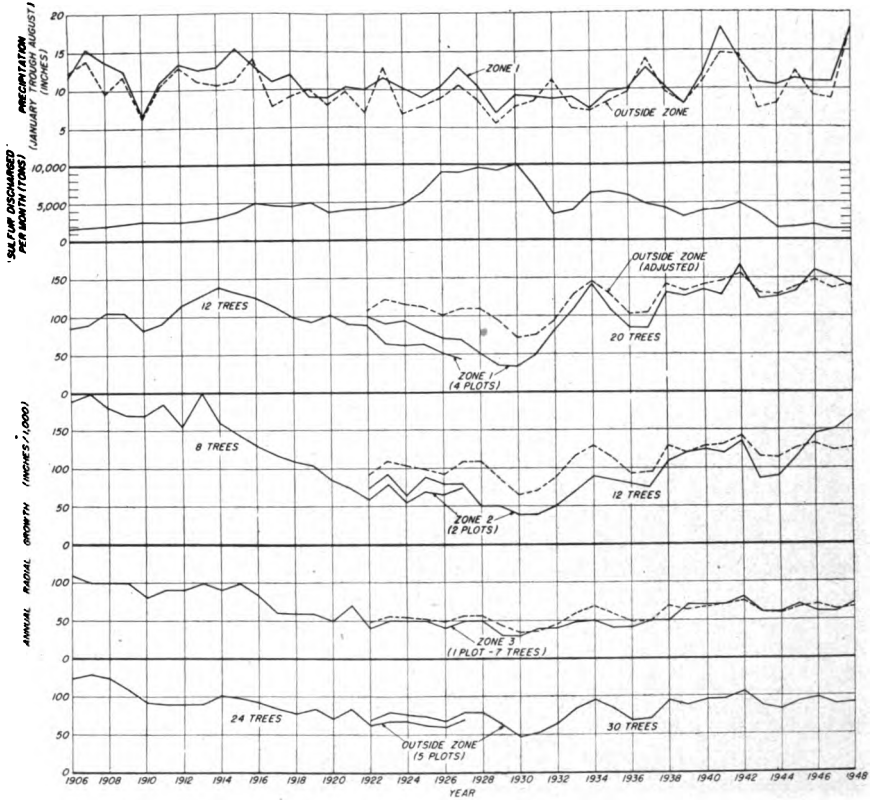


FIGURE 14.—Supplementary measurements of radial growth of ponderosa pine trees in the upper Columbia River Valley, Wash., and expected growth (outside zone, adjusted) had there been no suppression by sulfur dioxide.

the much more comprehensive first set, but the period of apparent retardation is about the same. Judged by the combined evidence for the three zones, important retardation probably ceased permanently in 1937 or 1938.

Timber Recovery

In the summer of 1949, there was no evidence of current sulfur dioxide injury, even to ninebark, the especially sensitive indicator plant. Moreover, there was no evidence of significant injury in recent years. Ponderosa pine trees only about 14 miles from Trail, British Columbia, for example, had green needles persisting to ages of 4 and 5 years, and in some instances 6 years. Functional branches were present comparatively low on the trunks. These lower branches are among the first to be killed by sulfur dioxide. Young reproduction of various ages was additional evidence of healthy surroundings for some years past. Typical representatives of both young and mature, full-crowned ponderosa pine trees as they appeared in zone 1 in 1949

are shown in figure 15. These contrast sharply with the thin-crowned ponderosa pines, same zone but 10 miles farther from the smelter, in 1930, as shown in figure 7.

There is no indication in the diameter increments of the last several years (fig. 14), or in the above-mentioned features, of any residual deleterious effect of sulfur accumulations in the soil. This accords with the findings of Katz and others (12) that increases in acidity, decreases in base saturation, and increases in the sulfate content of the soil were negligible at distances greater than 10 or 12 miles from the smelter.

It was not clear what permanent effect, if any, the tree mortality caused by sulfur dioxide may have had on the character of the timber stands. In 1949 the density of stocking and appearance in other respects of stands in zone 3 were not conspicuously different from those in the outside zone. On the other hand, the stands in zone 1 were plainly thinner for the most part than in the other zones, partly a result of fires as well as of killing by sulfur dioxide, but there were no denuded areas so large as to preclude fairly uniform natural re-stocking if fires are kept out in the future.

On the whole, it appeared that the reestablished stands are likely to be comparable to the original ones in species representation except possibly for parts of the river valley slopes in zone 1. Here sizable but irregularly distributed areas had been taken over by western larch, presumably because of a larger original proportion of parent trees on the slopes and because of the superior capacity of this species to survive exposure to sulfur dioxide.



M 98395 F

FIGURE 15.—Representative ponderosa pine trees in zone 1 in 1949. The full crowns are typical of trees that have been healthy for several years. The site is a river bench about 18 miles south, by river, of the smelter at Trail, British Columbia.

No abnormal erosion of timbered areas was observed in 1949. Any tendencies toward erosion 15 to 25 years earlier, therefore, must not have been so aggravated as to preclude subsequent natural correction by vegetative cover.

SUMMARY

Studies were made of two northwestern timbered areas affected by sulfur dioxide from large ore smelters. One area, which was much more intensively studied than the other, is in the Columbia River Valley, Wash., just south of the Canadian border. The smelter of chief significance on the area, at Trail, British Columbia, had been in operation since 1896. The other area is in the general vicinity of Anaconda, Mont., where a smelter was established in 1884. Most of the observations were on coniferous timber, which is the major constituent of the forest stands.

The underlying factors of sulfur dioxide injury and of the dissemination of the gas are described. As these factors are not quantitatively the same in different places, the investigations were essentially case studies. The incidence and distribution of injury were observed in relation to the proximity of the timber to the smelters, to the local and general topography, and to climatic and other environmental features.

The chief external criterion of injury was the typical leaf markings. A shrub, Pacific ninebark, was particularly sensitive to sulfur dioxide and aided considerably in establishing limits of influential amounts of the gas. The relative abundance of injury manifested by leaf markings and the proportion of trees with reduced foliage and vigor served to delineate broad zones of injury.

Winter injury was a minor complicating factor in diagnosing sulfur dioxide injury, which it resembles. Distinguishing features are described. Insect attack on the timber was not noticeably influenced by the presence of sulfur dioxide except perhaps for some increase in bark beetle infestation of larger trees that had been reduced in vigor by the gas. Parasitic fungus diseases were no more prevalent in sulfur dioxide affected areas than in outside areas. In fact, a number of needle and leaf fungi, including several rusts, appeared to be retarded or inhibited, especially where the injury by sulfur dioxide to trees and shrubs was greatest. Heart rot of Douglas-fir and of lodgepole pine trees was of about equal prevalence on areas of injury and outside.

Sulfur assays of foliage corroborated earlier findings by others that the presence of sulfur dioxide results in abnormal accumulations of sulfur; but it is apparently the rate, not the amount, of sulfur entry into the leaf that determines the occurrence of injury.

Tables are presented listing the order of observed susceptibility of forest trees, shrubs, and herbs to sulfur dioxide injury. The evergreen coniferous trees generally are more severely injured by smelter fumes than trees that do not retain their foliage in winter. Winter fumigations may injure evergreen species, particularly when temperatures are above 40° F. Although tending to be much less damaging than warm weather fumigations, they will result in death if repeated in successive years. The order of susceptibility of evergreen conifers

in the dormant season appeared to be much like that in the growing season. Western larch is perhaps the most susceptible to sulfur dioxide of all the observed trees when its needles are young. Nevertheless it has superior survival capacity because its mature needles are relatively tolerant, and, being deciduous, it is affected by foliage injury only through the growing season in which the injury occurs.

In experiments with artificial fumigation, small transplants of conifers were injured by exposures of 6 and 7 hours to 1 part per million of sulfur dioxide, and some were injured by 4 hours' exposure. Seven hours' exposure to $\frac{1}{2}$ part per million did not affect all species. The lowest concentration of sulfur dioxide that will injure conifers appears to be no less than 0.25 part per million. In supplementary experiments on first-year ponderosa pine seedlings, the seedlings were much more susceptible to injury than the larger transplanted trees.

In areas affected by sulfur dioxide, reproduction was scarce. This appeared to be a result of reduced cone crops and susceptibility and increased mortality of seedlings.

Ponderosa pine saplings were reduced in vigor and killed in increasing numbers during years of heaviest fumigation. Considerably larger trees (i. e., about pole size) were similarly affected, indicating that increased maturity beyond the sapling stage was not accompanied by greater tolerance to sulfur dioxide. A small percentage of sapling and larger ponderosa pines remained in good condition through the years of heaviest fumigation, suggesting that these individuals may have possessed certain morphological or physiological protective features not present in most of the trees. On areas of moderate sulfur dioxide injury, Douglas-fir saplings were reduced in vigor and killed in proportions comparable to those of the ponderosa pine saplings. Nearer the smelter, however, the Douglas-firs were much more severely affected than the pines, and, unlike the pines, no individuals remained in good condition.

Height increment, indicated by the spacing between branch whorls, and diameter increment, indicated by the width of growth rings, were decreased in ponderosa pines where there was visible injury to the trees. The amount of retardation was ascertained by comparing growth rates of the affected trees with rates of comparably situated trees outside the areas of injury, after first making allowances for differences in rate arising from natural differences in the environment. There was about a year's delay in the effect on height increment of sulfur dioxide and precipitation influence; diameter increment tended to be altered in the same year in which these factors changed considerably. Under the influence of progressively larger amounts of sulfur dioxide in the atmosphere, diameter growth seemed to be retarded a few years earlier than height growth.

Eighteen years after the initiation of sulfur dioxide abatement measures in the Columbia River Valley area of study, and about 12 years after the last indication of suppressed diameter growth of the trees, there was no evidence of current injury to timber or plants. Moreover, the appearance of individual trees did not suggest any residual effect of earlier damage; all trees had a normal quota of needles and active branches. Likewise, there was no indication in the diameter increments of the past several years of any residual

deleterious effect of sulfur accumulations in the soil. Timber stands in zones of greatest injury were still thinner than elsewhere but there were no denuded areas so large as to preclude fairly prompt restocking. Young reproduction of various ages was already established at many places. The composition of the stands did not appear to be greatly altered except for an increased proportion of larch reproduction on some areas.

LITERATURE CITED

- (1) ANONYMOUS.
1917. THE SULPHUR DIOXIDE PROBLEM. *Metall. and Chem. Engin.* 17 (12) : 1-4.
- (2) BRIZI, U.
1904. SULLE ALTERAZIONI PRODOTTE ALLE PIANTE COLTIVATE DALLE PRINCIPALI EMANAZIONI GASOSE DEGLI STABILIMENTE INDUSTRIALI. *Review by Solle in Zeitschrift für Pflanzenkrankheiten* 14: 160-163.
- (3) BÜSGEN, M.
1929. THE STRUCTURE AND LIFE OF FOREST TREES. Ed. 3, rev. and enl. by E. Münch. (Eng. trans. by Thomas Thompson.) 436 pp. London.
- (4) FULTON, CHARLES H.
1915. METALLURGICAL SMOKE. *U. S. Bur. Mines Bul.* 84, 94 pp., illus.
- (5) HASELHOFF, E., AND LINDAU, G.
1903. DIE BESCHÄDIGUNG DER VEGETATION DURCH RAUCH. *HANDBUCH ZUR ERKENNUNG UND BEURTEILUNG VON HAUCHSCHÄDEN.* 412 pp. Cebträder Borntraeger, Berlin.
- (6) HAYWOOD, J. K.
1905. INJURY TO VEGETATION BY SMELTER FUMES. *U. S. Dept. Agr. Bur. Chem. Bul.* 89, 23 pp., illus.
- (7) _____
1910. INJURY TO VEGETATION AND ANIMAL LIFE BY SMELTER WASTES. *U. S. Dept. Agr. Bur. Chem. Bul.* 113, 63 pp., illus.
- (8) HEDGCOCK, G. G.
1912. WINTER KILLING AND SMELTER INJURY IN THE FORESTS OF MONTANA. *Torreya* 12: 25-30.
- (9) _____
1914. INJURIES BY SMELTER SMOKE IN SOUTHEASTERN TENNESSEE. *Wash. Acad. Sci. Jour.* 4: 70-71.
- (10) KATZ, MORRIS.
1949. SULFUR DIOXIDE IN THE ATMOSPHERE AND ITS RELATION TO PLANT LIFE. *Indus. and Engin. Chem.* 41 (11) : 2450-2465.
- (11) McCLELLAND, ELLWOOD H.
1913. BIBLIOGRAPHY OF SMOKE AND SMOKE PREVENTION. Mellon Institute of Industrial Research and School of Specific Industries, Smoke Investigation Bul. 2, 164 pp.
- (12) NATIONAL RESEARCH COUNCIL OF CANADA.
1939. EFFECT OF SULPHUR DIOXIDE ON VEGETATION. 447 pp., illus. Ottawa, Canada.
- (13) RHINE, J. B.
1924. CLOGGING OF CONIFERS IN RELATION TO SMOKE INJURY. *Bot. Gaz.* 78 : 226-232.
- (14) SETTERSTROM, CARL, AND ZIMMERMAN, P. W.
1939. FACTORS INFLUENCING THE SUSCEPTIBILITY OF PLANTS TO SULPHUR DIOXIDE INJURY. *Boyce Thompson Inst. Contrib.* 19 (22) : 155-181, illus.
- (15) THOMAS, M. D.
1951. GAS DAMAGE TO PLANTS. *Ann. Rev. Plant Physiol.* 2 : 293-322.
- (16) TOUMEY, J. W.
1921. DAMAGE TO FORESTS AND OTHER VEGETATION BY SMOKE, ASH, AND FUMES FROM MANUFACTURING PLANTS IN NUGATUCK VALLEY, CONN. *Jour. Forestry* 19 : 367-373, illus.

- (17) U. S. DEPT. OF STATE.
1941. TRAIL SMELTER ARBITRATION BETWEEN THE UNITED STATES AND CANADA UNDER CONVENTION OF APRIL 15, 1935. DECISION OF THE TRIBUNAL REPORTED MARCH 11, 1941. U. S. Dept. State Pub. 1649, Arbitration Series 8, 61 pp.
- (18) U. S. TECHNICAL CONFERENCE ON AIR POLLUTION.
1952. AIR POLLUTION, PROCEEDINGS, WASHINGTON, 1950. 847 pp., illus. New York.
- (19) VAN DUYNE, CORNELIUS, AND ASHTON, FRED W.
1915. SOIL SURVEY OF STEVENS COUNTY, WASHINGTON. U. S. Dept. Agr. Advance Sheets, Field Operations of the Bureau of Soils, 1913, 137 pp.
- (20) WELLS, E. A.
1917. RESULTS OF RECENT INVESTIGATIONS OF THE SMELTER SMOKE PROBLEM. Indus. and Engin. Chem. 9 (7) : 640-646.



