

A  
QC  
880  
A4  
no.52

# NOAA Research Laboratories

Air Resources

Atmospheric Turbulence and Diffusion Laboratory

Oak Ridge, Tennessee

THE FEASIBILITY OF REDUCING AMBIENT AIR CONCENTRATION OF  
SULFUR OXIDES BY DISPERSION

F. A. Gifford

U. S. DEPARTMENT OF COMMERCE  
NATIONAL OCEANIC AND ATMOSPHERIC ADMINISTRATION

ARATDL Contribution No. 52

A  
QC  
880  
A4  
no. 52

The Feasibility of Reducing Ambient Air  
Concentration of Sulfur Oxides by Dispersion

F. A. Gifford  
Air Resources Atmospheric Turbulence and Diffusion Laboratory  
National Oceanic and Atmospheric Administration  
Oak Ridge, Tennessee  
April 1971

ATMOSPHERIC SCIENCES  
LIBRARY  
JUL 19 1972  
N.O.A.A.  
U. S. Dept. of Commerce

Presented at the First Annual Industrial Air Pollution Control Conference,  
sponsored by the Department of Civil Engineering, College of Engineering,  
The University of Tennessee, Knoxville, Tennessee, April 22-23, 1971.  
Published in conference proceedings, Joseph R. Duncan, William A. Drewry,  
K. E. Noll, and J. C. Burdick, III, editors.



Let me start out by saying that, when I was asked to present this paper, the title and topic were assigned to me. I say this so that the nearest environmental action group doesn't ride me out of town on a rail. It is of course true that the atmosphere has great capability to diffuse properties, including sulfur pollution, but not an unlimited capacity or one that works all the time, or under adverse conditions. Furthermore there are some significant questions concerning the rate of removal of airborne sulfur by precipitation, as well as regional and global atmospheric contamination by sulfur aerosol, that are at the moment not entirely resolved. Both of these points should be kept in mind in any assessment of the capability of the atmosphere to reduce tall-stack sulfur emissions to acceptable levels of ambient air concentration. On the other hand it is equally true that power companies face continued expansion to meet increasing demands for electricity, that for this purpose they have to engineer power systems many years in advance, that large fractions of this power will be supplied by fossil-fuel combustion for several decades, and that we have right now no satisfactory (i.e. reasonably cheap) full-scale systems to remove sulfur from the combustion gases. Both to achieve and maintain presently required air quality levels and to guide the engineering design of future large-scale systems for removal of sulfur from stack gases, the question of atmospheric dilution

has to be considered. I would like in the following to present the essential meteorological facts of the matter for your consideration. I hope you'll excuse me from giving a lot of formulas. You can find all you need of this in the references. The important thing, I believe, is to understand certain general facts about tall-stack sulfur pollution.

#### I - Sources of Atmospheric Sulfur Pollution

Sulfur is added to the atmosphere by various natural and human activities, and removed by various processes. The current global sulfur balance is illustrated in Figure 1, after Robinson and Robbins.<sup>1</sup> From this you see that the total pollutant SO<sub>2</sub> source is a major part of the sulfur balance, and you can compare it with the other sources, sinks, and exchange processes. It is of interest that natural sulfur emissions, mainly as H<sub>2</sub>S, are about 1/3 greater than industrial emissions of SO<sub>2</sub> and H<sub>2</sub>S; SO<sub>2</sub> is the only significant industrial pollutant. Note that the oceans' net gain of sulfur is 95 x 10<sup>6</sup> tons/yr, as a result of runoff, rainout and deposition, and gaseous absorption. Three quarters of this sulfur comes to the oceans from agricultural applications to the soil and weathering of rocks.

Meteorologists, or at least my colleagues in Oak Ridge and I, find it convenient to classify (stationary) pollutant sources into three groups: 1) tall stack sources; 2) process emissions; and 3) area sources. By tall stacks we have in mind stacks in the height range from 200 ft or so up to giants such as the 800 ft Bull Run Steam Plant stack and even taller ones. The main requirement is



that the stack should be tall enough, and otherwise well-enough designed, so as not to suffer from "downwash," due to the adverse aerodynamic influence of nearby buildings or of inadequate stack draft. By process emissions we mean emissions from lower stacks or vents of industrial processes of all kinds whose height and disposition do not qualify them as tall stacks. And by area sources we have in mind the multitude of individual sources that comprise a city or town, mainly from space heating of all kinds. All these three can be sources of sulfur pollution, and each requires somewhat different consideration from the meteorological point of view. All three types of sources as a rule occur in conjunction because power demand, industry, and urbanization go together. Mine-mouth power plants are, perhaps, an exception.

Tall-Stack Sources. The behavior of plumes from tall-stack sources, as described by Briggs<sup>2,3,4</sup>, is dominated by the initial buoyancy of the hot gases emitted. This causes a strong rise of such plumes, initially, accompanied by a bending over due to the (horizontal) ambient wind. The relative motion between the rising plume and the atmosphere causes small-scale turbulence due to velocity shear at the plume's edge and results in a dilution of the initial plume buoyancy, by entrainment into the plume of ambient air. The upshot is that tall-stack plumes containing sulfur from fuel combustion rise to great heights, usually more than twice the

source height. Stack designers depend on this plume rise to control ground level air concentrations to acceptable levels. Formulas for calculating plume rise under various meteorological conditions can be found in the papers by Briggs.

We understand plume rise fairly well, but our knowledge of processes that bring the sulfur back down to the ground again is rudimentary. Tall stack plume behavior is summarized in Figure 2.<sup>2</sup> Plumes are carried back down to the ground briefly under "looping" conditions. Plume "looping" occurs under conditions of strong thermal convective turbulence in the lower atmosphere, similar to the conditions that are accompanied by cumulus clouds. The result at the ground is very brief periods (a few minutes) of high ground level concentrations, but a fairly low average concentration value, due to the generally good atmospheric dispersion involved. These bursts of high concentration can occur as near to the source as about four times the stack height.

Plumes are also brought to ground level by the so-called "fumigation" condition. This simply refers to a rapid, general mixing downward of the plume under active mechanical turbulence (high wind) conditions, possibly exacerbated by the presence of a capping temperature inversion aloft. Because this condition can persist for longer periods of time, it produces the highest average ground-level concentrations, as a rule.



The normal downward diffusive spreading of plumes can in principle also bring material to the ground. For strongly buoyant plumes this occurs at great distances from the stack, probably no less than 20 stack heights, which means that considerable dilution will have occurred. The great distance is because downward turbulent mixing can be effective only after the stage of actively buoyant plume rise is over.

Sulfur from plumes can also reach the ground by being absorbed by falling raindrops. The extent and time scale of this phenomenon are quite uncertain. Rain is an efficient scavenger of plume  $\text{SO}_2$ , but the droplets evidently give up the sulfur readily as they fall below the plume. Thus the plume sulfur is in effect "smeared out" in the vertical, and the net result of rainfall is to augment vertical  $\text{SO}_2$  dispersion. The result for ground level concentrations as well as the extent of so-called "acid rain" are uncertain. My opinion, based on current, incomplete literature, is that sulfur pollution at the ground attributable to precipitation scavenging of tall stack plumes is not especially intense. That is, the high sulfur concentrations present in the plumes do not result in high ground level concentrations or intense "acid-rain" in precipitation. I would hazard the guess that the plume sulfur reaching the ground from this effect is first fairly well dispersed in the atmosphere by the combined actions of entrainment, turbulent mixing, and the adsorption-desorption effect. Thus

the plume sulfur should be thought of as tending to contribute to the overall, general sulfur content of rainfall rather than as being a source of highly local "hot spots." I believe that the primary source of higher than average rain and soil acidity buildups, such as has been reported in Sweden, will be found to be area and process emission sources and not tall stacks. I should stress however that direct, conclusive evidence for (or against) this opinion does not yet exist.

Process emission sources. The problem of process emissions is illustrated by Figure 3, taken from the ASME Guide<sup>5</sup>. Stacks can emit material either above the aerodynamic building wake, within the wake, or, for very short stacks, within the low-pressure cavity in the lee of the building. In the latter two cases, ground level concentrations can be high at all distances near the source. The problem involved in trying to calculate pollution from process emissions is to balance properly the complex plume buoyancy and momentum effects, aerodynamic building wake effects, and the influence of atmospheric turbulence which, near the ground, is highly variable. Guidance can be found in the ASME booklet.

Area sources. The multitude of small, low-level pollution sources including residential and industrial space-heating chimneys, and other such sources are most conveniently dealt with as total emissions per unit area. The main difficulty here is to establish the area source



strength, and systematic procedures for doing this have been established by HEW. Simplified procedures for calculating area source concentrations can be found in the papers by Gifford and Hanna<sup>12</sup> and Hanna<sup>13</sup>.

## II - The Relative Contribution to Ambient Air Quality of Sulfur Pollution from the Various Source Types

SO<sub>2</sub> from tall stacks emerges high above the ground, then rises still higher because of plume buoyancy, and is carried to the ground only intermittently, by air turbulence or precipitation processes. For this reason tall stacks tend to contribute relatively small amounts of SO<sub>2</sub> to the ambient air concentration at ground level. This was clearly brought out in the recent study by Ross, et al<sup>6</sup>. Figure 4, from their paper shows that while total SO<sub>2</sub> emissions from all sources in UK increased from 1958 to 1968, the annual average concentration decreased. This was due to the controls imposed on the sulfur content of fuels for the low level (lower solid curve) process emissions and area sources, mainly domestic heating. A related result was recently reported by Golden and Mongan<sup>7</sup>, who calculated air pollution for Chicago from emissions data. They found that the chief contribution to ambient air SO<sub>2</sub> concentrations is the area source component.

At a smaller, local scale, the Rockwood-Harriman air pollution study recently completed<sup>8</sup> by my colleagues at ATDL produced an equivalent result, as Table I shows.

Table I

Suspended Particle  
Concentration in the  
Rockwood-Harriman Area

<u>Source</u>	<u>Annual Avg. Amt.</u>
Upwind Cities (Nashville, Knoxville, Chattanooga, etc.)	~ 40 $\mu\text{g}/\text{m}^3$
Kingston Steam Plant	~ 5-10 "
Roane Electric - in Rockwood	~ 50 "
in Harriman	~ 25 "
Space Heating, Both Towns	~ 50 "

The conclusion from all such studies, as argued very eloquently by Ross, et al., is that tall stacks are in fact an excellent, and probably an indispensable way to reduce ground level pollution by the emitted material to tolerable levels.

III - Larger-scale and Global Climate Effects

As I indicated earlier, sulfur pollution from tall stacks is one of the major contributors to the atmospheric sulfur balance. Some atmospheric  $\text{SO}_2$  is quickly removed by precipitation scavenging and other processes, but some becomes oxidized to sulfate aerosol. Such fine particles change the heat balance of the earth<sup>9</sup> by reflection and absorption of both solar and terrestrial radiation. Even such small particles are fairly efficiently removed by precipitation scavenging and other removal processes. Examination of solar radiation



records in North America and Europe has lead Budyko<sup>10</sup> to speculate that the observed 1940 to 1960 solar radiation decrease of about 4% is due to aerosols from anthropogenic pollution. Mitchell<sup>11</sup> on the other hand believes that the man-made aerosol fraction is much less than at from volcanic eruptions. The situation is not particularly clear, and should be studied very carefully. The recent MIT summer study<sup>9</sup> concludes that problems in climate modification due to the increase in aerosol population will tend, because of the somewhat uncertain but relatively short atmospheric residence times of aerosols, to be a regional problem long before they become a global one, and for the same reason the trend can be reversed quickly, within a few months, if pollution control measures are employed.

#### IV - Summary and Conclusions

1. Tall stacks contribute a sizeable fraction of the total sulfur in the air. With other anthropogenic sources they contribute an amount comparable with that from natural sources of sulfur. Most of this is SO<sub>2</sub>.
2. Sulfur pollution sources are of three types: tall stacks, process emissions, and area sources.
3. Tall stack sources of sulfur are buoyant and this produces large additional plume rises, essentially doubling the effective release height.

4. Sulfur in tall stack plumes is brought down to the ground intermittently by atmospheric turbulence, and also by precipitation scavenging.

5. The contribution of tall stack plumes to  $\text{SO}_2$  ambient air quality levels averages appreciably less, by a factor of 5 to 10 or more, than that from lower-level process emissions and area sources (where these are present).

6. The possibility of "acid rain" from tall stack plumes exists. But the (so far rather meager) observational evidence suggests that the contribution will not be especially intense, compared to that from process emission and area sources of sulfur. That is, tall stacks probably should in this respect be thought of as contributing a pro rata share to the general atmospheric sulfur content of a region.

7. Part of tall stack  $\text{SO}_2$  emissions are converted to sulfate aerosols. The precise extent and importance of this effect are not known but, as with the rainfall acidity, any adverse meteorological effects appear to be regional to continental in scale, i.e. neither local nor global.

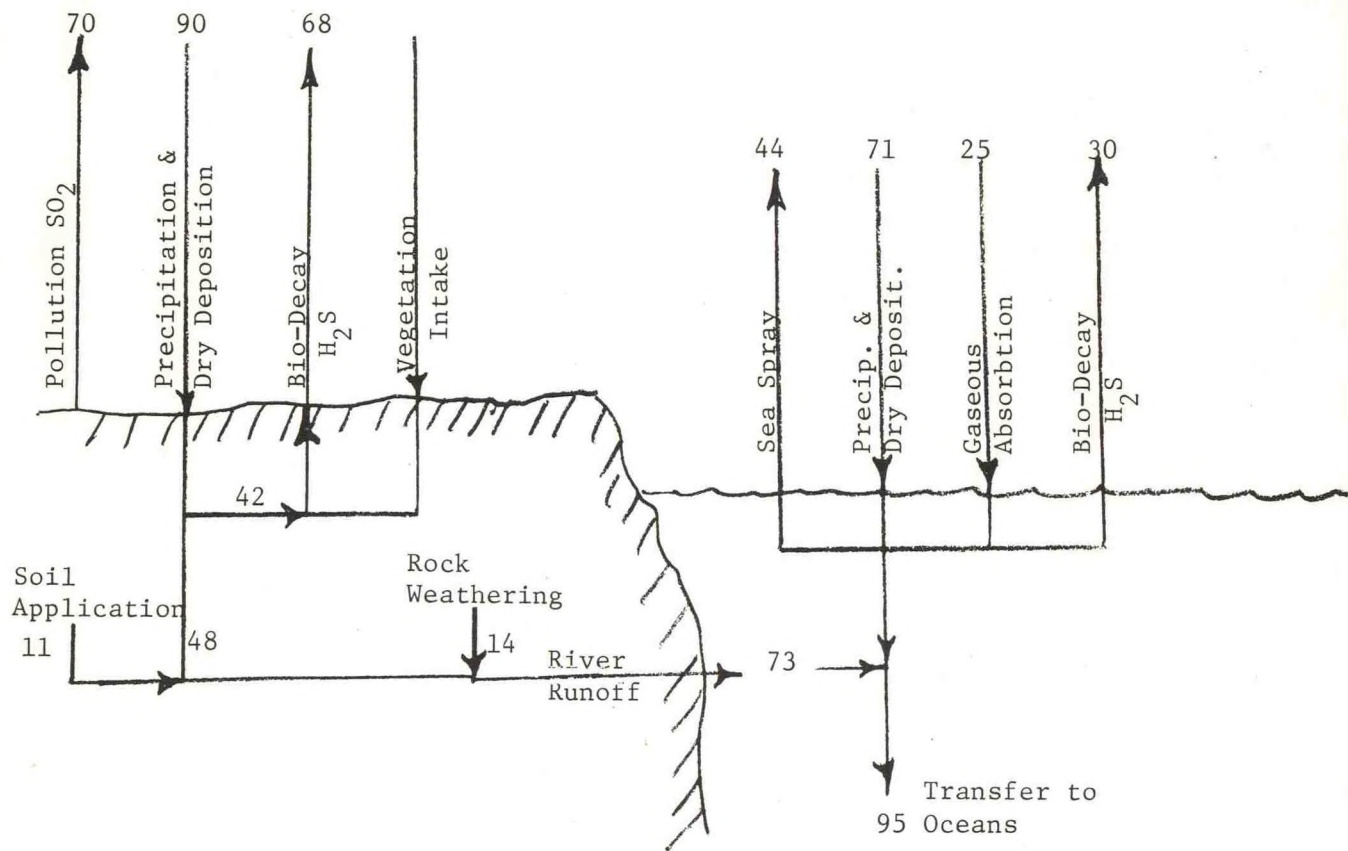


V - References

1. Robinson, E. and R. C. Robbins, 1970: Gaseous atmospheric pollutants from urban and natural sources; pp 50-63 in Global effects of environmental pollution, edited by S. F. Singer, Springer-Verlag.
2. Briggs, G., 1969: Plume Rise, vi and 81 pp, USAEC, DTI.
3. Briggs, G. A., 1969: Optimum formulas for buoyant plume rise; Phil. Trans. Roy. Soc. Lond. A. 265, pp 197-203.
4. Briggs, Gary A., 1970: Some recent analyses of plume rise observations. (Paper presented at the 1970 International Air Pollution Conference of the International Union of Air Pollution Prevention Associations. ATDL contribution number 38.)
5. Smith, Maynard (editor), 1968: Recommended guide for the prediction of the dispersion of airborne effluents, ix and 85 pp.
6. Ross, F. F., A. J. Clarke, and D. H. Lucas, 1970: Tall stacks - how effective are they? Proc. of the Second Int. Clean Air Congress of the Int. Union of Air Poll. Prevention Assoc., Dec 6-11, 1970, Wash., D. C. (to be published).
7. Golden, J. and T. R. Mongan, 1971: Sulfur dioxide emissions from power plants: their effect on air quality. Science, 171, 381-383.
8. Hanna, Steven R. and S. D. Swisher, 1970: Air Pollution Meteorology of the Rockwood-Harriman, Tennessee, Valley, (ATDL contribution no. 40).
9. Mass. Inst. of Tech., 1970: Man's Impact on the Global Environment. MIT Press, xxii and 319 pp.
10. Budyko, M. I., 1969: The effect of solar radiation variations on the climate of the earth. Tellus, 21.
11. Mitchell, J. M., 1970: A preliminary evaluation of atmospheric pollution as a cause of the global temperature fluctuation of the past century; pp 139 - 155 in Global effects of environmental pollution, ed. by S. F. Singer, Springer-Verlag.

12. Gifford, F. A. and Steven R. Hanna, 1970: Urban air pollution modelling, (presented at 1970 International Air Pollution Conference of the International Union of Air Pollution Prevention Associations, ATDL contribution no. 37).
13. Hanna, Steven R., 1970: Simple methods of calculating dispersion from urban area sources, (presented April 6, 1971, at the Conference on Air Pollution Meteorology, sponsored by the American Meteorological Society in cooperation with the Air Pollution Control Association, at Raleigh, North Carolina, ATDL contribution no. 46).





(10<sup>6</sup> tons/yr Sulfur)

Figure 1

BEHAVIOR OF SMOKE PLUMES

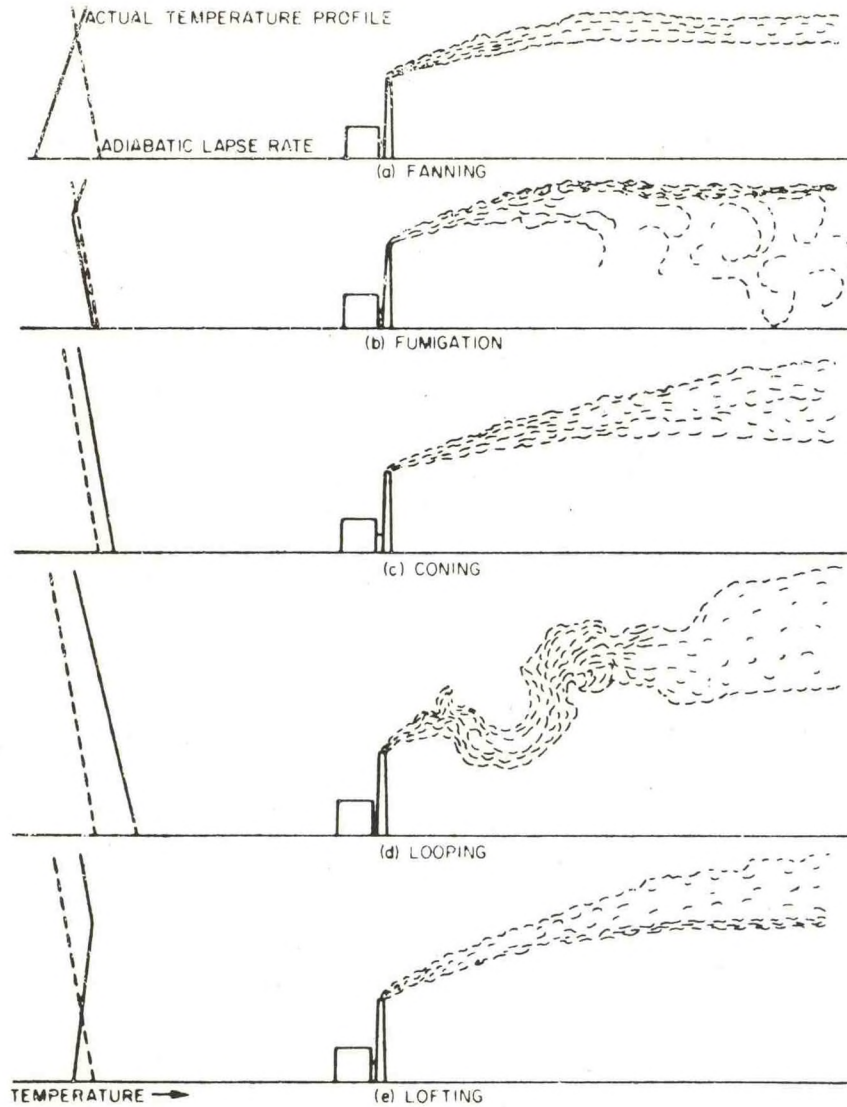


Figure 2 - Effect of temperature profile on plume rise and diffusion



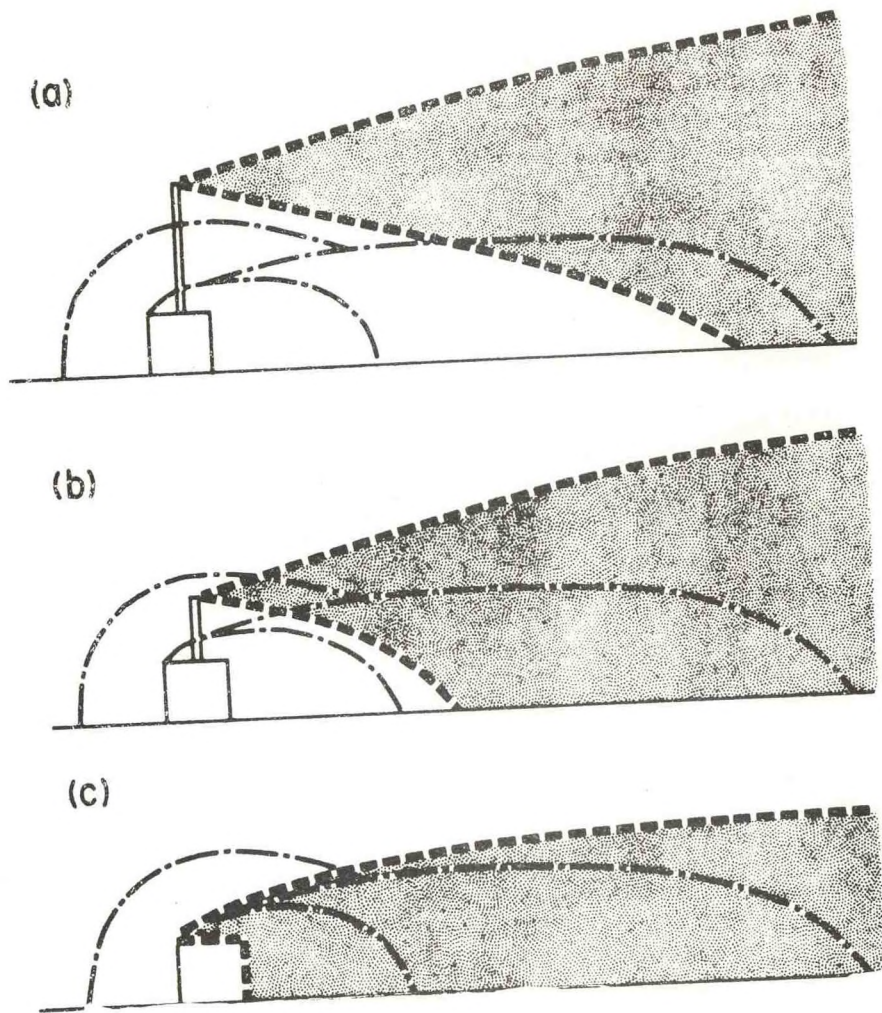


Figure 3 - AERODYNAMIC EFFECT ON PLUME DISPERSION.  
These figures show the behavior of stack  
plumes emitted on various heights and positions  
relative to a cubical building.

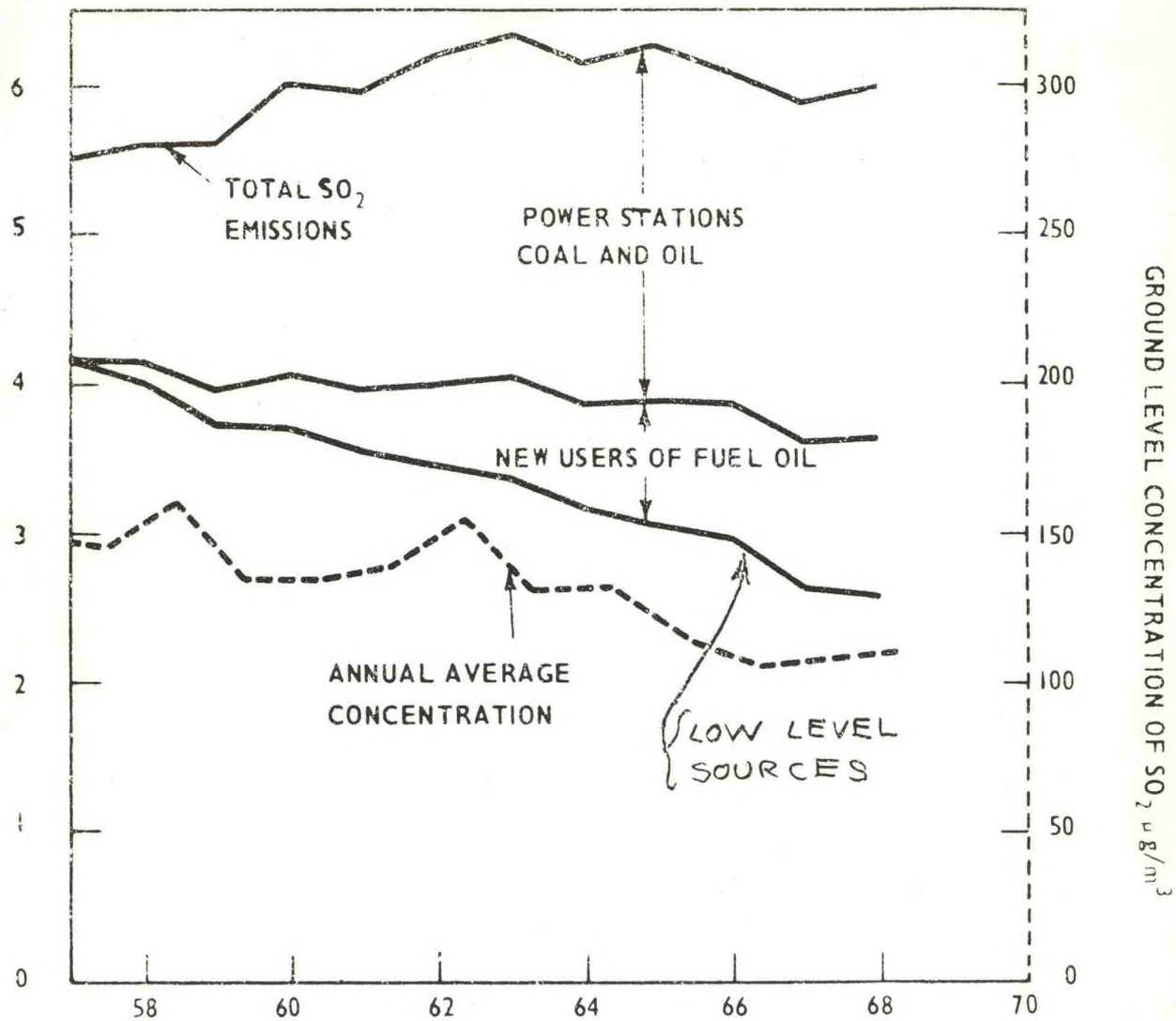


Figure 4 - Annual Emissions and Average Annual Concentrations in Atmosphere of U.K. 1957-1968