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INTERNATIONAL CONFERENCE ON OXIDANTS, 1976 - ANALYSIS OF EVIDENCE AND VIEWPOINTS

Part I. Definition of Key Issues

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ABSTRACT

In recognition of the important and somewhat controversial nature of the oxidant control problem, the U.S. Environmental Protection Agency (EPA) organized and conducted a 5-day International Conference in September 1976. The more than one hundred presentations and discussions at the Conference revealed the existence of several issues and prompted the EPA to sponsor a follow-up review/analysis effort. The follow-up effort was designed to review carefully and impartially, to analyze relevant evidence and viewpoints reported at the International Conference (and elsewhere), and to attempt to resolve some of the oxidant-related scientific issues. The review/analysis was conducted by experts (who did not work for the EPA or for industry) of widely recognized competence and experience in the area of photochemical pollution occurrence and control.

Part I of the overall effort is an explanatory analysis of the problem and definition of key issues, as viewed within the research component of the EPA. Parts II through VIII are written by expert reviewers offering either resolutions of issues or recommendations for additional research needed to achieve such resolution.

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INTRODUCTION

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The photochemical oxidant pollution problem in the U.S. has been recognized for several decades. While some control of oxidant precursor emissions was first enforced in the early 1960s, it was not until 1971 that an official oxidant control strategy was formulated and legislated into use (1). Despite the preceding long years of research and development effort addressed to understanding the problem and conceiving solutions, this first strategy has had all the usual uncertainty problems that accompany any first effort or result.

In the years since the inception of this first strategy, several developments have taken place. First, experiences by enforcement agencies brought to surface several enforcement or implementation problems heretofore unsuspected. Second, considerable new research was conducted and reported with strong implications relevant to the oxidant control problem. Third, the problem, now recognized in other countries, has acquired international dimensions because of the possibility of intercountry pollution transport and because of the impact of emission control regulations upon international trade. These developments raised the question of the need to reexamine the oxidant control policies and their underlying scientific bases, and eventually instigated the International Conference on Photochemical Oxidant Pollution and Its Control, held in Raleigh, N.C., September 12-17, 1976, under the auspices of USEPA and the Organization for Economic Cooperation and Development (OECD) (2).

This paper was originally published as "International Conference on Oxidant Problems: Analysis of The Evidence/Viewpoints Presented. Part I. Definition of Key Issues." APCA Journal, Vol. 27, No. 4, April 1977. Reprinted with the permission of the APCA Journal.

Some one hundred reports on new evidence and/or viewpoints were presented at the International Conference by researchers and air pollution specialists representing the entire spectrum of scientific community, and, as expected, brought into focus some important and controversial issues. Because of the wealth of new information presented on these issues, and because of the often conflicting nature of this information, and finally, because of the great importance of these issues, it was felt that it would be extremely useful to carefully scrutinize and analyze all presented information on each issue, to attempt to reconcile any conflicting evidence and/or viewpoints, and finally to arrive at a judgment that would either pronounce the issue essentially resolved or declare the differences in evidence/viewpoint irreconcilable, and to define needs for additional specific research. Perhaps it should be restated that the subjects of this effort are existing issues, that is, *questions that have been given concrete but conflicting answers*. Needless to say, there are innumerable relevant questions that have not been given any answers or not even studied yet; this effort clearly is not intended to seek answers to these latter questions.

The first step of this analysis effort was completed in October 1976 and is reported here. In this first step, the entire problem of oxidant pollution and its control has been rethought and, drawing upon our experiences and judgment and upon information reported at the International Conference and elsewhere, we have defined certain issues, specifically, those we feel pertained to the key components of an optimum oxidant control strategy.

The second step of this analysis — to be conducted by review panels — consists of a careful review of all Conference papers (and other reported information) directly or even peripherally dealing with a given issue. Each review panel member has been requested to prepare a review, the purpose of which is (a) to examine the reported evidence and viewpoints for conflicts; (b) to make judgments on strengths and weaknesses of opposing viewpoints or evidence and, based on such judgments, attempt to reconcile conflicting viewpoints and evidence; and (c) to derive a factual or judgmental conclusion regarding resolution or the status of the issue and offer recommendations for

additonal research. Such reviews have been solicited from several experts who do not work for EPA.

In the third step of this analysis, the reviews to be obtained for each issue will be combined into a single report that will include (a) the individual reviews intact, and (b) a digest of all reviewers' conclusions with a discussion of their implications regarding the oxidant control problem. It was planned that these reports be published either as journal articles or as EPA reports. If and when review panel meetings are called for, such meetings will be organized and funded by EPA.

THE ISSUES

INTRODUCTION

Questions that must be answered, or, alternatively, distinct components of the problem of oxidant pollution and its control involve the following:

- Nature, extent, and magnitude of the problem.
- Controllable and uncontrollable sources of the problem and relationship between source area and receptor (problem) area.
- Air quality goal that must be achieved, that is, the National Air Quality Standard for Oxidant/Ozone (NAQS-O_x).
- Achievability of the NAQS-O_x.
- Methods for calculating emission control requirements.
- Methods for controlling emissions from sources.
- Methods for reducing sources/emissions in an area.

These questions or problems must be resolved if oxidant control measures are to be developed and applied. Since application of such measures is dictated by law in the U.S. (3), it follows that these questions *must be given answers*, some of which may be based on sound and complete evidence and some

based by necessity on incomplete evidence or merely reasonable speculation. This dictates that the review effort requested here should not be preoccupied with whether there are answers that meet a prescribed set of validity criteria or not. Rather, the objective should be to offer answers, based either on a consensus of viewpoints and evidence or on a "referee" judgment when existing viewpoints and/or evidence are in conflict.

The scope of this review also must be by necessity limited. For reasons mainly of effort manageability and practicality, the scope of this review must be limited to include only some of the questions at issue, obviously those that are relatively more important and were given extensive coverage at the International Conference and elsewhere. Such a piecemeal examination of the oxidant problem often presents difficulties, the most serious of which is the apparent reluctance of the average reviewer to judge the various components of a problem independently of each other. This problem is well recognized but no solution is offered here other than to appeal to the reviewer's better judgment. The success or failure of this effort clearly will depend on the degree to which the reviewer's reasoning adheres to the relevant scientific evidence available and remains unbiased by forseeable consequences or implications of the conclusion.

Based on our own appreciation of the various aspects of the oxidant problem and the coverage these aspects received at the International Conference and elsewhere, we have defined and offer the following issues to be included in this review effort. In the hope of providing some review guidelines of assistance, we have attempted, in each case, to briefly analyze the issue, explain its position and relevance within the oxidant problem picture, and raise the specific questions that constitute the issue, and the answers to which are the very objective of this effort. We have also identified and made available to the reviewers copies of those Conference papers that in the authors' judgment are relevant to each issue. Needless to say, the reviewers may request and will be provided with copies of any additional Conference papers or of any other available reports.

THE ISSUE OF REACTIVITY

In regard to reactivity, the questions most urgent and directly related to the oxidant control problem pertain (a) to the effectiveness of "substitution" (of less reactive for more reactive organic emissions) as an approach to oxidant control and (b) to the identification of those organics that are essentially of no concern insofar as the oxidant problem is concerned. The more specific questions that need to be answered follow.

- Does the scientific evidence alone justify formulation and enforcement of *interim* substitution rules more stringent than Rule 66?
- Considering all relevant factors, e.g., impact upon urban air quality, impact upon rural air quality, cost, technological feasibility, etc., would it be preferable to abandon altogether the idea of developing interim improved substitution rules and devote instead and immediately all attention and resources to development of methods and practices for "nearly indiscriminate" control of organics?
- Are there any organics so little reactive that they would neither cause nor contribute significantly to oxidant buildup at problem levels under any circumstances?

There are also the relatively less important questions regarding definition of reactivity and validity of the data and procedure used to classify organics based on their relative abilities to contribute to the urban oxidant problem.

Of these questions, the one on the merits of substitution has been discussed both internally in EPA and informally at an open meeting (EPA's Forum on Solvent Substitution, Chicago, Ill., Oct 13-14, 1976); there was a consensus that substitution will have a small but possibly significant benefit upon urban air quality — more precisely, the air quality in the vicinity of the source area — but will have less or no benefit upon distant downwind areas. Although not quantitatively answered, the question was nevertheless treated adequately so that further discussion here is not warranted. Also, the question on merits of "nearly indiscriminate" control is outside the scope of this review, since it calls for judgments on cost, technological feasibility, etc.

The question that is offered as the specific subject of this review is the one regarding the possible existence and identities of organics incapable of causing oxidant problems. This question was discussed at the International Conference and drew conflicting answers. The specific issue here is centered around the method used for reactivity-rating the various organics and for defining the borderline separating the reactive ones from those of *virtually no concern with respect to the oxidant problem.* In general, two distinctly different approaches were proposed: The smog chamber approach applicable to all organics (4), and the mathematical modeling approach (5) applicable, at present, to certain organics only, namely, paraffinic and olefinic hydrocarbons and aliphatic aldehydes. To facilitate the process of judging these two and/or any other approaches, it would perhaps be useful to break the issue down to two parts: One pertaining to the reactivity-rating of the organics, especially of those of low reactivity, and one pertaining to the positioning of the borderline separating the almost totally unreactive ones from the reactives. Judgments that must be made are on:

- whether the two proposed approaches agree or disagree in results and to what extent,
- whether one or the other or the two approaches combined in some way or any other approach yields the most reliable results, and
- the specific additional research needed to substantiate or refute these first judgments.

THE ISSUE OF STRATOSPHERIC OZONE INTRUSION

In a broad sense, the question at issue here is whether ozone of stratospheric origin contributes significantly to the ozone problems observed in urban and rural areas. Aside from the possibility that stratospheric-tropospheric exchange contributes directly and significantly to ground-level ozone buildup, stratospheric ozone has also been proposed to have a "reaction-trigger" function that accelerates and enhances photochemical oxidant formation from hydrocarbon-NO_x precursors. The stratospheric ozone intrusion question is part of the broader question regarding the magnitude and extent of

the ozone problem caused by natural causes, which in turn is a part of the issue of achievability of the National Air Quality Standard for Oxidants (NAQS-O_x). To further explain the interest in the stratospheric ozone question, it should be clarified and stressed here that this question, in fact the entire issue of achievability of the NAQS-O_x, has no bearing whatever upon the justification of the NAQS-O_x; such justification is based strictly on health effects considerations. The stratospheric ozone question needs to be answered only for the purpose of more accurately estimating the benefits to be derived from anthropogenic emission reduction.

Evidence interpreted to show accumulation of stratospheric ozone within the troposphere varies widely in type and degree of directness. Thus, high levels of ozone were measured in the upper troposphere near tropopause discontinuity points (6), evidence that attests to stratospheric origin most directly. On the other extreme, ground-level oxidant buildup in some instances was attributed by investigators to stratospheric intrusion only because these investigators did not have or would not accept any other explanations (7). Overall, direct, unequivocal evidence on the impact of stratospheric ozone intrusion upon tropospheric air quality is lacking, and for this reason it may be expected that the viewpoints and interpretations of evidence expressed to date reflect to some — perhaps substantial — degree a subjective judgment.

At present, a realistic assessment would suggest that the extent (by area), intensity (by concentration), and frequency of occurrence of stratospheric ozone buildup at ground level all vary widely so that single answers and answers to all of the questions that constitute the issue cannot be given. It would, therefore, be more productive to define and offer as the subject of this review only those that are most relevant to the oxidant control strategy issue and receive substantial research attention. These questions are proposed here to be as follows:

1. Accepting that intensive stratosphere-troposphere exchanges do occur at tropopause discontinuity points, what is the extent, frequency, duration, and spatial/temporal predictability of such occurrences?

The terms "extent" and "spatial" here refer to areas at high altitudes, that is, near the tropopause, not at ground level. While quantitative answers are not expected, at least, a judgment should be made whether such exchanges are sporadic, unpredictable incidents causing local ozone accumulations or are significantly extensive and predictable. Main interest, of course, is in occurrences within the U.S.

2. Accepting that localized high concentrations of stratospheric ozone can occur in the *upper* troposphere, what fraction of such ozone is expected to reach ground level

   (a) under meteorological conditions most conducive to downward transport, and

   (b) under meteorological conditions most conducive to photochemical oxidant formation?

The questions asked here, in essence, are again whether or not stratospheric ozone excursions *to ground level* are sporadic, unpredictable incidents causing only local, short-lived (e.g., a few hours) ozone accumulations, and whether or not such excursions are likely to occur during smog episode periods.

The question concerning the possible "reaction-trigger" function of stratospheric ozone is not raised here because, thus far at least, it has been a subject of speculation only; no relevant evidence apparently exists, except for a few as yet unreported smog chamber experiments. Nevertheless, comments from the reviewers on this question are welcome.

THE ISSUE OF NATURAL ORGANIC EMISSIONS

This issue was originally raised as a result of an early report that, on a global basis, rates of organic emissions from vegetation are considerably higher than those from man-made sources (8). Although such natural emissions and the anthropogenic emissions are for the most part geographically segregated, it is nevertheless reasonable to suspect that natural organics, either

emitted within the urban or nonurban area or brought in through transport, could contribute to the ambient oxidant problem to an important degree. These suspicions became considerably stronger — and for obvious reasons — as a result of two recent findings: The occurrence of a pervasive rural oxidant problem (9), and the high reactivity of terpenes (10,11). In either case, the finding could be interpreted to mean that natural organics may constitute a significant source of oxidant. As in the case of stratospheric ozone, the question regarding the importance of natural emissions as an oxidant source needs to be answered only for the purpose of more accurately estimating the benefits from man-made emission control.

From a first glance examination of the evidence relevant to this issue, it becomes immediately apparent that certain components of the issue have obvious answers or have been resolved based on scientific evidence, whereas other components remain uncertain or unresolved. For example, it is unquestionable that vegetation does emit organic vapors and that some of these vapors (terpenes) play the dual role of oxidant precursor and oxidant destruction agent. What is in question is (a) the nature (other than terpenic) and emission rates of such vapors, and (b) the net effect upon oxidant of the atmospheric reactions of natural organics. Some evidence relevant to these questions does exist but does not necessarily provide answers. Researchers are, in general, less familiar with the natural organic emissions, and this lack of familiarity naturally casts doubts over all types of evidence available, from data on chemical identity and ambient concentrations to information on atmospheric photochemistry and on emission and sink processes. It is because of this general lack of confidence that the specific questions offered here as the questions at issue, include some of relatively basic nature. These questions are:

1. Given the fact that terpenes are emitted by vegetation, does it automatically follow, or is there evidence to show, that terpenes are present in ambient air at levels commensurate with their emission rates? Are such levels significant?

While it is almost certain that these questions can be answered one way or

another, based on evidence available, the judgments needed here are not only on the interpretation of such evidence but also on the reliability or overall quality and conclusiveness of the evidence available.

2. Accepting the possibility that terpenes are present in ambient air at significant levels, does the available evidence — from either direct or indirect data or theoretical inferences — support a predominantly ozone-producing or ozone-destruction role or both roles for such terpenes?

In deliberating such a question, the distinction should be made and recognized between urban and rural atmospheres. It should be clarified and stressed here that the fact that the current concern is mainly about the urban problem does not justify overlooking the rural situation. One reason for this is that oxidant formed in rural areas could contribute importantly to the urban problem. It is precisely this possibility that should be explored in deliberating this question. To further explain, it is conceivable that in rural areas, that is, in forested and thinly, but nevertheless significantly, populated areas, the terpene and anthropogenic emissions could yield mixtures with organic composition and organic-to-NO_x ratios conducive to oxidant formation. This is one theory that could explain the "oxidant/ozone blanket" phenonmenon — of uniform variation of oxidant/ozone levels over large areas — observed in parts of the country (12). Since this "blanket oxidant/ozone phenomenon" could be explained by several theories, namely,

(a) photochemistry of terpenes mixed with anthropogenic emissions in rural areas,

(b) photochemistry of anthropogenic emissions (alone) in rural areas,

(c) pollutant transport from large urban centers, and

(d) stratospheric ozone intrusion,

the judgment called for here is for the relative credibilities of the four theories, and specifically, for the credibility of the terpene theory.

3. The third question is essentially a solicitation of evidence and/or viewpoint — reported or unreported — on the existence and photochemical pollution role of natural organic emissions other than terpenes.

THE ISSUE OF OXIDANT TRANSPORT

It is to be remembered that this review-analysis effort is concerned only with existing issues, that is with questions that have been answered but conflictingly. If such a definition of issues is to be observed strictly, then while there are numerous unanswered questions there may be no issue related to oxidant transport, at least, none other than those already defined and discussed in connection with the stratospheric ozone and the natural emissions. To explain, one major question relevant to the oxidant problem is on the relative strengths of the natural and the anthropogenic sources in a given region or area. This question has not been answered unequivocally and quantitatively because anthropogenic pollutant transport makes it difficult to assess the strength of the natural sources. The question has been answered qualitatively, a consensus being that pollutant transport does occur and contributes to oxidant buildup in areas far from the sources (13-15). Quantitative answers, however, have not been agreed upon, and this disagreement constitutes the issues already presented in the preceding two sections of this analysis.

Aside from its connection to the natural vs anthropogenic sources question, the phenomenon of oxidant transport is of interest for yet another extremely important reason. This reason is the strong possibility — a fact, to some investigators — that oxidant and/or oxidant precursors transported from upwind sources obscure the role and impact of local emissions to a degree that local control requirements cannot be estimated with confidence. In fact, this obscuring effect is a problem of much broader nature, affecting both main components of the oxidant control strategy, namely:

(a) the source-receptor relationship, that is, the geographical definition of the area within which emission control must be applied in order to reduce the oxidant levels observed in a (given) locality, and

(b) the quantitative relationship between oxidant-related air quality and precursor emission rates.

This connection between oxidant transport and oxidant control strategy is a conclusion arrived at as a result of numerous recent field studies (13-15). Specifically, these studies established that the emission-dispersion and photochemical reaction processes do not have a simple and "local" nature as was assumed in the designing of the first — and current — oxidant control strategy. The phenomena of urban oxidant plume formation and movement, rural oxidant occurrence (at problem levels), "Sunday-weekday effect," and nighttime oxidant occurrence, previously either unnoticed or thought to be "odd," are now believed to be manifestations of an extremely complex emission/pollutant dispersion process. Such complexities, for example, are introduced by horizontal and/or vertical transport of oxidants and/or of precursor mixtures to long distances without excessive dilution.

It is this connection between oxidant control strategy and oxidant transport, and, within this context, the specific areas of nature, extent of, and consequences from pollutant transport, in which several questions exist and need to be answered. Some of these questions have been given conflicting answers, but the supporting evidence was in almost every case either scant or none. For this latter reason, these questions should perhaps be considered as "unanswered" questions rather than as questions at issue. Nevertheless, for important and urgent reasons these questions will be included in this review/analysis with the understanding that the need here is either for answers or for specific recommendations for research that would provide answers. These questions and related explanations/discussion are as follows:

1. What is the maximum range of ozone transport?

The question pertains specifically to ozone, and is concerned with the maximum distance downwind that ozone can travel without excessive destruction or dilution (e.g., no destruction or dilution more than 80 percent). Answers have been offered, but are nonspecific and vary by several tens of kilometers. Thus, from direct observations upon an urban oxidant plume it cannot always be

ascertained whether the transported oxidant constitutes a fraction of the concentration at the point of origin or is fresh oxidant formed during transport. Most likely, both types of oxidant exist but in unknown, and not easy to determine, proportions. The question involves considerations of chemistry and meteorology (dispersion) and could perhaps be answered in parts. For example, it would be relevant and useful to answer the following questions:

(a) What is the photochemistry-related lifetime of ozone?

Answers have been calculated for an ideal atmospheric system containing no HC and NO_x pollutants, except for methane (and CO) at their global back-ground levels (16). It is conceivable, however, that in the presence of trace-levels of HC and NO_x — levels such that their potential for O_3 formation is either negligible or predictable — that the lifetime of O_3 may be quite different.

(b) What is the range of ozone lifetimes related to atmospheric (at ground level) on surface destruction processes?

(c) For an inert pollutant, what is the longest lifetime related to the atmospheric dilution process?

2. What is the maximum range of oxidant-precursor transport?

The question pertains to HC and NO_x pollutants as a mixture — not to the individual precursors — and is concerned with the maximum distance downwind that a HC-NO_x mixture can travel without excessive loss of its potential for oxidant formation, (e.g., no loss of oxidant potential more than 80 percent). The question is far more complex than the preceding one on ozone; nevertheless, answers have been offered, although again nonspecific and diverse. For example, analysis of aerometric data provides evidence suggestive of "long-range" transport but does not identify corresponding source and receptor areas. Also, based on meteorological modeling techniques, it has been calculated that the residence time of air parcels — and their pollutants — in high pressure systems can be as long as several days (17); but this does not necessarily mean that the resident pollutants preserved a significant potential for oxidant

2. *Smog Chamber Approach.* The approach entails deriving cause-effect relationships between oxidant and precursors through laboratory testing. This approach could be characterized as semi-empirical because the relationships are derived from laboratory observations alone; they are not product of theoretical derivations. Further, as in the preceding case, this approach is intended to predict only changes in air quality resulting from changes in emission rates.

3. *Mathematical Modeling (or AQSM) Approach.* The approach entails deriving the requisite air quality-emission relationships entirely from theory. Its intended use is to predict both absolute levels of and changes in air quality from given emission rate and meteorological data.

To be usable, all methods of relating air quality to emissions, regardless of approach, must be validated and evaluated for accuracy. The distinction between "validation" and "accuracy-evaluation" follows. Validation refers to the agreement between model-predictions and observations when the input information fed into the model is perfectly accurate; thus, validation is the process of checking the validity of the principle underlying the method. Accuracy-evaluation refers to the agreement between model-predictions and observations for a model based on a perfectly sound principle; thus, accuracy-evaluation is an assessment of the error introduced by inaccuracies of the input information. Another term often used in connection with model evaluation is "verification," referring to the agreement between predictions and observations for the specific case in which the observations used for verification were taken from the same pool of data used to develop the model. This is the case, for example, of development and verification of AQSMs from the St. Louis RAPS data. In the discussion here, verification will be considered to be a limited form of validation.

Between validation and accuracy-evaluation, the latter appears to be relatively simpler, especially for the empirical and semi-empirical methods and for the relatively simple AQSM methods. Thus, useful accuracy evaluations can be made from estimates of the errors associated with the input information

and from numerical sensitivity tests to determine the impacts of such errors upon model-predictions. Unlike accuracy-evaluation, complete and direct validation of a model is extremely difficult — if at all possible — to accomplish for the main reason that the requisite "real world" data — on quality and emissions — are either not available or not easy to obtain. Thus, for the empirical and semi-empirical models relating emission changes to air quality changes, data on such changes either do not exist, or, if they do exist, as for the Los Angeles basin, they are useful only for verification of a "local-use" model. For the AQSM methods, intended to relate absolute levels of air quality to emission rates, validation hinges upon solution of several problems, one of which is the definition of absolute air quality in terms of commonly obtained air quality monitoring data. *At the present time*, these problems in validating models are considered to be prohibitive by some investigators, but not insurmountable by others. *It is this latter disagreement among investigators that constitutes the issue to be examined here.*

More specifically, the question at issue here is:

- At the present time, are there any air quality simulation models sufficiently validated/evaluated and appropriate for use in designing urban oxidant control strategies?

The Environmental Protection Agency has not issued nor does it have plans for immediate issuance of strategy design guidelines (for oxidant control) involving use of AQSMs. Furthermore, EPA is conducting an extensive study (RAPS project) to first verify and subsequently further validate and evaluate several of the presently available AQSMs. Obviously, therefore, EPA does not feel that *at this time* there are AQSMs ready for use. Contrary to this EPA viewpoint, some investigators have suggested that there is one sufficiently validated air quality simulation model (Bell Lab). The Bell Lab model was discussed at the International Conference; however, the more detailed description of the model and justification of the Bell Lab viewpoint are to be found in a subsequently published journal article (18).

There have been numerous reports, presented at the International Conference and elsewhere, on the principle, status of validation, and intended utility of several AQSMs. It is important, however, to make the distinction between true model applications and model exercises that merely demonstrate the *intended* utility of a model. The judgment called for here is whether there can be true model applications, that is, *whether there are models presently available that can and should be used immediately in designing urban oxidant control strategies.*

THE ISSUE REGARDING THE EVALUATION OF A SMOG CHAMBER METHOD AS A REPLACEMENT FOR THE APPENDIX-J METHOD.

The subject smog chamber method has been described and discussed in detail at the International Conference and elsewhere. Questions at issue here pertain to the merits claimed for the smog chamber method both relative to the Appendix-J method and in an absolute sense. For urgent reasons, examination of this issue has already begun in the form of Task Group activity conducted by EPA and non-EPA experts. Therefore, this issue will not be included in the analysis effort contemplated here.

THE ISSUE OF OXIDANT/OZONE MEASUREMENT

Recent studies have resulted in some disconcerting evidence regarding the performances of the various oxidant/ozone measurement methods in existence (1922). All KI procedures for either measuring ambient oxidant/ozone or for calibrating oxidant/ozone measurements methods were found to disagree with each other; the disagreement varied in degree depending on study or analyst. There was also disagreement between certain KI procedures and the more ozone-specific chemiluminescence and UV photometry methods. It is, generally, agreed that part of the disagreement is caused by the usual precision and accuracy errors associated with the various procedural steps, and part with the difference in response specificity among the various methods. Thus, all KI methods show response to ozone as well as to all vapors capable of oxidizing iodide ions or reducing iodine. Since these vapors and O_3 do not all cause equivalent responses, it follows that the KI measurement reflects not only the

concentrations but also the composition — to the extent that such composition varies — of the responding vapor mixture. Further, the expected differences in response specificity are larger between the KI methods and the chemiluminescence and UV methods.

In the face of these differences in precision, accuracy, and response-specificity among the various oxidant/ozone measurement methods, the obvious question relevant to this analysis is whether these imperfections in the analytical method invalidate any component of the oxidant control strategy. To explore this question, the functional relationship between the analytical method for oxidant/ozone and the oxidant control strategy, first, needs to be clarified.

Of the various components of the current oxidant control strategy the only one linked to the analytical method for oxidant/ozone is the one related to the calculation of emission control requirements. Such calculation requires that the following three entities be defined:

- Present air quality (PAQ), i.e., second highest 1-hour oxidant/ozone in the reference year,
- desired air quality (DAQ), i.e., the NAAQS for oxidant/ozone (0.08 ppm O_3), and
- a quantitative relationship between air quality and emission rates.

Of these, PAQ and DAQ are obviously the entities specifically linked to the oxidant/ozone measurement method. The preceding question, therefore, is now reduced to whether the analytical method imperfections invalidate (a) the PAQ data, and (b) the NAAQS for oxidant/ozone. Each of these two cases is examined separately.

To explore the impact of the analytical method imperfections upon the PAQ data, it might be useful to break down the impact of such imperfections into two parts: the impact arising from the usual precision and accuracy errors of the methods, and the impact arising from the nonspecificity of response. The

precision/accuracy errors vary depending on (a) the method (e.g., KI methods, chemiluminescence, or UV photometry) and (b) the entity to be measured. To explain the latter, the magnitude of the precision/accuracy error is greater when the entity to be measured (i.e., PAQ) is expressed in terms of a single datum (e.g., second highest value) out of a population of data, than when it is the population average. Note, however, that the criterion for selecting the "second highest concentration" or the "average concentration" (or any other concentration) as the entity to be measured, is not the magnitude of the analytical error; rather, it is the health effects of oxidant/ozone. EPA has interpreted the health effects evidence available to mean that oxidant-related air quality should be defined in terms of a "highest" or "second highest" rather than "average" oxidant/ozone concentration. *Whether this interpretation of the health effects of oxidants/ozone is correct is outside the scope of this analysis.* Thus, in the light of this discussion, the first specific question that needs to be answered is:

1. Do the precision/accuracy errors invalidate the Federal reference method for oxidant/ozone? If yes, what method should be chosen instead?

The impact upon PAQ data of errors related to the specificity of response is far more complex than that of the precision/accuracy errors. The main complication arises from the rational requirement that the method measure that or those chemical species that have been found to have adverse health effects. However, since an important part of the health effects evidence is of an epidemiological nature, *those species could not have been specified unequivocally.* This problem was circumvented by devising and using the concept of "surrogate" species, that is, species believed to represent those with the adverse effects. EPA initially proposed to promulgate that "oxidants," as measured by a specified KI method, be used as the surrogate species; in the final promulgation, "oxidants" measured by a (more ozone-specific) chemiluminescence method was pronounced the surrogate species. Rationally, measurement of PAQ by the chemiluminescence method should give lower results than by any KI method. However, there have been reports to the contrary. In the light of this latter dispute, and of the fact that the intended use of all methods is

to measure surrogate species, the relevant question that must be answered next is:

2. Do the response-specificity errors invalidate the Federal Reference Method for oxidant/ozone? If yes, what method should be chosen instead?

The preceding two paragraphs dealt with the impact of the analytical method imperfections upon the PAQ data. The remaining discussion will deal with the impact upon the NAAQS for oxidant/ozone. The first obvious, and direct question to be asked here is:

3. Do the imperfections of the analytical methods for oxidant/ozone invalidate the air quality standard for oxidant/ozone?

Following is a proposed answer to this question, and the reviewer judgment called for is on the correctness or incorrectness of this proposed answer.

The answer to this question, to a large degree, depends on the evidence and reasoning underlying the development of the air quality standard for oxidant/ozone. The underlying evidence is known to consist of associations between adverse effects and concentrations of "oxidants" measured by a variety of analytical methods. It should be noted that a major part of this associative evidence is not of a cause-effect nature. This means that the oxidant species responsible for the adverse effects could not have been unequivocally specified, which in turn means that the air quality standard did not have to be defined in terms of one or more specified oxidant species. Thus, the standard could be defined in terms of surrogate species, that is, in terms of a "response" given by any "oxidant" measurement method. In conclusion then, the validity of the qualitative definition of the NAAQS for oxidant/ozone should not be questioned.

The quantitative part of the (air quality) standard (i.e., the "0.08 ppm") can only be a result of analysis and interpretation of the health evidence available. Apparently the judgments made by the EPA experts with

respect to the severity of the health effects and to the safety margin required were such that they justified use of the *0.08-ppm limit as measured by a specified analytical method (Federal Reference Method)*. These judgments may or may not be sound. However, this clearly pertains to another issue, namely the issue of health justification of the NAAQS-oxidant/ozone, and not to the issue of oxidant/ozone measurement. In conclusion, again, the imperfections of the analytical methods do not invalidate the 0.08-ppm part of the oxidant/ozone standard, or, to put it differently, *cannot have much different impact on the validity of a higher or lower standard*.

THE ISSUE OF THE OPTIMUM OXIDANT CONTROL STRATEGY

As discussed in the introduction paragraphs preceding the first six issue sections, there are several questions or problems that must be resolved if oxidant control measures are to be developed and applied. Some of these questions, by virtue of their importance and basic nature, and the considerable contradictory attention paid to them, have attained an issue status and have been treated individually in the preceding sections. Resolution of those issues will definitely and considerably advance the understanding of the oxidant problem, but will still leave the primary issue of optimum oxidant control strategy somewhat open. There are still several questions more directly and specifically addressed to the subject of oxidant control strategy that need to be given definitive answers. These questions are to be dealt with in this section.

Departure points in this discussion/analysis will be two facts: The existence of an oxidant control strategy since 1971, and the generation, since 1971, of considerable new scientific evidence pertaining to the oxidant control problem. The immediate and obvious question that arises from these facts is whether the new evidence supports or invalidates, partly or wholly, the first, existing oxidant control strategy. If the strategy is shown to be invalidated in some respects, then the next obvious question is what strategy revisions are dictated or can be justified by the new evidence. To explore these questions or possibilities, it is necessary that the new scientific

evidence, or, more directly, the changes in understanding brought about by the new evidence be specified.

The most important change in understanding that has been brought about by the new evidence concerns the source-receptor relationship, that is, the geographical or spatial relationship between areas in which emissions are discharged and the areas where the air quality is impacted by these emissions. Much of the discussion needed here about this relationship has already been presented as part of the discussion on the Oxidant Transport issue. The control strategy implications, however, of the source-receptor relationships, as now understood, need to be further expanded.

The first implication is that long-range pollutant transport introduces a link between the urban oxidant problem and the rural oxidant problem. This means that in many areas, urban emissions and oxidant significantly affect rural air quality and, conversely, oxidant-carrying rural air upwind from a city significantly affects the city's air quality. Thus, from a control standpoint, the urban and rural problems are not entirely disassociated, and therefore, respective optimum control strategies should not be pursued entirely independent of each other.

A second implication is the one arising from the vertical mixing patterns observed and associated with radiation and subsidence inversion phenomena. Such mixing patterns suggest that local emissions may have significant carry-over effects upon next day's local air quality. This and the previous implication depict a new picture of the photochemical processes responsibile for a city's oxidant problem. According to this picture, the local oxidant problem is the composite of contributions originating from:

(a) local, fresh emissions,

(b) local but previous day's emissions,

(c) extraneous emissions (probably from previous days), and

(d) natural sources.

The third implication relates to the relative roles of the HC and NO_x emissions in the oxidant problem. The existing oxidant control strategy formally recognizes only a HC role; no oxidant-related controls are imposed upon NO_x emissions. Such roles of the precursors, however, are now thought to be incorrect quantitatively and, perhaps, qualitatively also. Thus, the oxidant-to-HC dependence is not independent of the NO_x factor. Also, and more importantly, the effects of local HC and NO_x emission controls on oxidant are expected to vary depending on whether the oxidant results from local fresh emissions or from previous day's local emissions or from extraneous emissions.

In the light of these implications of the recent scientific findings, the specific questions that need to be answered here are as follows.

1. Is the qualitative basis of the existing oxidant control strategy still sound? That is, is hydrocarbon emission control an optimum approach to urban oxidant reduction?

Because of the link between the urban and the rural oxidant problem, the answer must be based on considerations of both types of problem. Consideration must also be given to the situation in which local emissions have multi-day carryover effects upon local oxidant. Finally, consideration should be given to the relative importances of the anthropogenic and the natural sources. In essence then, the first question asked here can be reworded as follows:

> Considering the four origins of <u>urban</u> oxidant [cases (a) - (d) described in preceding paragraphs], is control of local (urban) HC emissions expected to be an effective means to local oxidant reduction?

Relevant evidence consisting of both laboratory and field data has been reported at the International Conference and elsewhere, and is conflicting. Thus some field studies showed HC control to have a strong beneficial effect, others did not show a detectable effect, and others showed effects varying with attendant NO_x emission change. Smog chamber studies showed that for the portion of oxidant formed from the day's local emissions, control of HC is beneficial except for atmospheres with extremely high — ordinarily not observed

— hydrocarbon-to-NO_x ratios; for such atmospheres HC control, unless drastic, will have very little effect. For the portion of oxidant formed through multiday irradiation of emissions, the evidence is scant and inconclusive by some investigators but conclusive by others. Those who feel that the evidence is conclusive claim that HC control will have small effects upon multiday, irradiated air masses, relative to the effect upon single-day, irradiated air. Control of HC emissions upwind, again, has little effect by some investigators, undetermined effect by others. While it is certain that the effectiveness of the HC control approach is different for different localities, the judgment called for here is whether this approach should be retained or be replaced by another approach.

2. Insofar as the urban oxidant problem is concerned, is NO_x emission control imperative? desirable? tolerable? undesirable? intolerable?

Again, for the questions to be answered properly, consideration must be given to the various sources of urban oxidant, namely, pollutant transport, local/fresh emissions, local/aged emissions, and natural sources. Evidence from field studies is conflicting in that it shows higher NO_x emission rates to be associated with lower oxidant concentrations in some cases, and no such association in others. Smog chamber data exist only for the situation in which the urban oxidant forms from the day's local emissions. For this situation, NO_x control has varying effects depending on the hydrocarbon-to-NO_x ratio of the reacting emissions. The effect of control of the upwind emissions of NO_x also may be in dispute. Again, as with the preceding question, the judgment called for here is whether the NO_x emission factor should continue to be nearly ignored — as is the case with the existing strategy — or should be considered, and how.

The preceding paragraphs dealt with the qualitative bases of an optimum oxidant control strategy, that is, with the questions pertaining to the directional impacts of the HC and NO_x emission controls. The questions that need to be defined next deal with the quantitative bases of an optimum strategy, that is, with the quantitative impacts of controls. Such quantitative bases consist of the following two components:

- The quantitative relationship between ambient oxidant concentrations and emission rates, and
- The definitive relationship between source area and receptor area, meaning the definition of the geographical area where the required control — *as calculated from the oxidant-emission relationship* — must be applied to solve the oxidant problem observed in a given locality.

The quantitative relationship between oxidant and emissions is the subject of the issue on the replacement of the Appendix-J method, and will not be treated here. Nevertheless, it might be helpful to mention here that the smog chamber method, proposed as a replacement of the Appendix-J method, provides a cause-effect relationship between oxidant and precursors and, unlike the Appendix-J method, does not take any specific source-receptor relationship for granted. Thus the smog chamber method does not prescribe, as the Appendix-J method does, that control be confined within the urban area where the oxidant problem was observed.

Assuming that the replacement of the Appendix-J method will be a method based on a cause-effect relationship between oxidant and emissions, it will be necessary that "cause" and "effect" be identified, respectively, with a "source area" and a corresponding "receptor area," the latter being the area where the air quality is impacted by the source area.

Defining the source-receptor relationship consistent with the cause-effect nature can be approached, in theory at least, in several different ways. By one, first approach, "effect" is identified with the maximum oxidant observed in the "receptor" area, and "cause" is identified with the emissions from *all* sources — *local and upwind* — that impact the receptor area. This approach requires that all source areas impacting the receptor area be identified, a requirement which is extremely difficult — if at all possible — to fulfill.

By the second approach, "effect" is identified with the fraction of the

observed maximum oxidant concentration attributable to the local emissions, and "cause" is identified with the local emissions. This approach has at least two problems:

- The determination of the oxidant fraction associated with the local emissions, and
- The requirement that another source-receptor relationship be defined for the fraction of oxidant associated with the extraneous, upwind emissions.

The first problem is not an insolvable one: e.g., a rough estimate of the oxidant fractions associated with the local and the extraneous emissions could be obtained from oxidant measurements upwind and downwind from the receptor area. The second problem, however, is extremely difficult — if at all possible — to solve.

A third approach could be conceived as a simplified compromise between the two preceding ones. By this third approach, "effect" is identified with the maximum oxidant concentration observed in the receptor area, and "cause" is identified with the local emissions. The assumption is made here that the local emissions are the sole and whole cause of the oxidant problem. This assumption, it is well recognized, is not valid in oxidant transport situations in which the local emissions cause only part of the problem. Nevertheless, the assumption is justified on grounds that in the very same (oxidant transport) situations, the local emissions almost surely contribute to or cause additional problems to downwind area. It might appear at first glance that application of controls calculated by this approach upon the receptor area as well as upon the upwind areas will result in over-control. In actuality, however, this will not necessarily be the case because the sum total of the "local" and "transported-in" contributions to oxidant may exceed the 0.08-ppm standard even though the individual contributions are each less than 0.08 ppm.

Finally, a fourth approach could be conceived as a more-stringent-control version of the second approach. By the fourth approach, "effect" is identified

with the *fraction* of the maximum oxidant concentrations attributable to the local emissions. However, calculating control requirements so as to reduce a *fraction* of the oxidant down to 0.08 ppm, obviously, will not solve the oxidant problem. To solve the problem, control requirements should be calculated so as to reduce the "local" oxidant fraction below 0.08 ppm, that is, to a level such that the *total* oxidant will be, if possible, at or below 0.08 ppm. Thus, by the fourth approach, the oxidant contribution from the extraneous (upwind) sources is partly or wholly offset by imposing increased control of local emissions.

The preceding analysis identifies four conceivable approaches to formulating an optimum strategy for oxidant control. Obviously, there may be others. The question to be asked here is:

Which of these four, or any other, approaches is the one to be recommended?

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16. ABSTRACT

In recognition of the important and somewhat controversial nature of the oxidant control problem, the U.S. Environmental Protection Agency (EPA) organized and conducted a 5-day International Conference in September 1976. The more than one hundred presentations and discussions at the Conference revealed the existence of several issues and prompted the EPA to sponsor a follow-up review/analysis effort. The follow-up effort was designed to review carefully and impartially, to analyze relevant evidence and viewpoints reported at the International Conference (and elsewhere), and to attempt to resolve some of the oxidant-related scientific issues. The review/analysis was conducted by experts (who did not work for the EPA or for industry) of widely recognized competence and experience in the area of photochemical pollution occurrence and control.

Part I of the overall effort is an explanatory analysis of the problem and definition of key issues, as viewed within the research component of the EPA. Parts II through VIII are written by expert reviewers offering either resolutions of issues or recommendations for additional research needed to achieve such resolution.

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