CONF-8806460--1

BNL--42102

DE90 008518

Overview of the Methodologies Used to Compile Large Scale Emissions Inventories

Carmen M. Benkovitz
Atmospheric Sciences Division
Department of Applied Science
Brookhaven National Laboratory
Upton, N.Y. 11973

INTRODUCTION

Development of emissions inventories for large geographic areas is currently based on the use of "mass balance" equations to estimate the emissions from each source. The general equation used states that the emissions (E) from a particular source for a specific pollutant in a specified time frame are equal to the product of the activity rate of the source (A) in the selected time frame, an emission factor (EF) which estimates the amount of pollutant emitted per unit activity, and additional parameters which vary according to the type of source whose emissions are being estimated. Examples of these parameters are the characteristics of the fuel burned, such as sulfur or ash content, the efficiency of any control device used, etc. Mathematically stated:

$$E = A \times EF \times P_1 \times P_2 \times \ldots\ldots$$

where P_1, P_2, etc. are the additional parameters. The time period most frequently used in the application of this equation is one year.

Emissions for large stationary sources (point sources) are calculated on an individual basis; for smaller sources and for mobile sources methodologies currently in use estimate emissions as area aggregates. In general, the classification of sources is done according to the criteria outlined in the Federal Register, November 25, 1971, 51.1(k). When finer time, space or species resolution are desired, current practice is to develop "disaggregation factors" to be applied to the basic emissions data. Thus the above equation becomes:

$$E = A \times EF \times P_1 \times P_2 \times \ldots\ldots \times DF_1 \times DF_2 \times \ldots\ldots$$

where DF_1, DF_2, etc are the disaggregation factors.

Values for the parameters and disaggregation factors used in emissions calculations can be estimated in two different ways. "Individual" estimates can be made for the parameters by estimating a value for each source. "Averaged" estimates can be made for some parameters; in this case, the set of all sources is divided into categories that are thought to be internally more homogeneous than the general population. For each of these categories, a value is estimated for the parameter and used to calculate the emissions from all the sources in the category.

The objective of this paper is to present an overview of the methodologies currently used to estimate the values of the parameters needed to compile large scale inventories of anthropogenic emissions, to discuss the major approximations used and to offer recommendations for future refinement of the inventory data.

EMISSIONS ESTIMATION PARAMETERS

Emission Factors

An emission factor has been defined [1] as "a statistical average or a quantitative estimate of the rate at which a pollutant is released to the atmosphere as a result of an activity such as combustion or industrial production, divided by the level of that activity". Emission factor values may be estimated for an individual source and used to calculate the emissions of that source, or one emission factor may be estimated from data for several sources and used to calculate the emissions from all sources in the same category. Several techniques have been used to develop values for emission factors; these techniques vary from detailed testing of one or more sources with many measurements that include all known process variables and process parameters, to engineering estimates of a given process. Published results may be simple averages of all available data of acceptable quality, without regard to the influence of source parameters, or empirical formulae developed to relate the emissions to the actual values of the source parameters. The U.S. Environmental Protection Agency (EPA) has developed source classification categories and assigned Source Classification Codes (SCC) to each category. For each SCC, emission factor values have been compiled for primary pollutants that have been classified as criteria pollutants (pollutants for which National Ambient Air Quality Standards have been established); these values are presented in EPA's publication AP-42 [2]. Programs such as the National Acid Precipitation Assessment Program (NAPAP) [3] have developed SCC-based emission factor values for a few other primary pollutant species of interest.

Errors in emission factors vary according to the techniques used to estimate these values. The most commonly used methods are to estimate values from test data and to estimate values based on engineering analysis or judgement; the following discussion will be limited to these two methods. Errors associated with the use of values estimated from test data are of three main types: (1) source category characterization errors, (2) measurement errors, and (3) data processing errors.

Source category characterization errors can be the most obvious or the most difficult to define and correct. The most obvious cases occur when the emission factor for one category has been used for another, "similar", category; error correction includes a definition of the important source variables and parameters and a well documented, quality controlled measurement program designed to generate an emission factor calculation technique that includes all of these. The most difficult cases are those where an emission factor is based on values derived from test data of sources in the category, but where: (1) the important source variables and parameters were not correctly identified and/or properly accounted for, (2) source characteristics may have changed over time, (3) there has been a significant improvement in the measurement technology, (4) the measurement program was based on a small number of non-representative sources in the category or (5) the averaging period of the test data is very different from the period used in the emission calculation. Error correction here should include a restudy of the category definitions, with possible reclassification of sources and creation of new categories, and the design and implementation of new measurement programs that properly address the representativeness and time averaging concerns.

Application of strict quality control and adherence to proven analytical techniques should reduce measurement errors to the statistical values that can be calculated based on known accuracy and precision of the measurements, making these errors both controllable and quantifiable. Data processing errors are associated with the transcribing of data and the use of calculations to derive the desired quantities from the experimental data. Use of advanced computer-based techniques for data capture and verification should minimize human error and machine malfunction; wrong application of mathematical formulations and errors in calculations are more subtle and need well established and documented quality control procedures implemented at every step of the process.

For point sources, emission factor values based on engineering analysis or judgement can be obtained only for some processes that emit particulates, sulfur oxides (SO_x) and hydrocarbons and are generally used for single sources. Errors associated with these estimation methodologies are linked to how accurately the processes are known; quantification of these errors is not directly feasible and is usually based on test data results for similar processes. Estimation of a better emission value for these sources usually implies implementation of measurement programs.

Emission factor values for an important sector of area sources are based on a combination of source testing and engineering judgement. Values of the nitrogen oxides (NO_x) and hydrocarbon emission factors for highway vehicles are estimated using EPA's MOBIL3 [7] model (to be replaced by MOBIL4); county-specific emission factors are computed for nine vehicle classes. Algorithms used in the model attempt to include all parameters that could affect the emissions rates of these mobile sources; some parameters are included without great detail, some factors that may be important are not currently included. A recent study by Chun [8] presents analyses of how more detailed treatments of some of the parameters affect the calculation of these emission factors. Refinement of the estimation methodologies used for this very important sector should be given high priority; further studies are needed to incorporate results into the model and to address additional parameters of importance.

Activity Rate

Activity rate values for stationary sources are reported on an individual basis; for area and mobile sources these are estimated at a county level.

Activity rate values reported for large stationary sources are based on company records of fuel use and/or production figures. In general, larger installations know and report these figures to a higher degree of accuracy than do smaller ones. Largest source of errors here are usually data processing errors, which include rounding errors due to changes in the conventions used to report these data to different agencies. Annual figures have been found to be fairly accurate; when seasonal or finer breakdowns have been requested, errors have increased. Use of advanced computer-based techniques for data capture, data conversion and data verification and a strong quality assurance program should help to minimize errors.

Activity rate values for area sources are quantified using a combination of top-down and bottom-up approaches. Bottom-up calculations start at the county level and use algorithms based on surrogate indicators such as population or employment figures. There are numerous sources of errors implicit in this approach, which include: (1) how well the chosen surrogate represents the true activity, (2) how well do the algorithms represent the processes being modeled and (3) how accurate are the values for the surrogates chosen. The bottom-up values are calculated and summed to obtain totals for each state. The top-down approach starts with state level figures, obtained either directly or by adjusting national figures, and corrected for point source levels. "Adjustment factors" are used when bottom-up results do not agree with top-down values. Additional errors include (1) accuracy of top level values, (2) how well do the breakdown algorithms represent the true breakdown and (3) accuracy of point source corrections applied.

Mobile source activity rates are expressed as vehicle miles traveled (VMT) and are based on figures published by the Department of Transportation [4]. When available, county level VMT data are used (approximately 30 to 40% of all counties in the nation compile these data). VMT data are based on actual highway traffic monitoring; the Federal Highway Administration has set guidelines to be used in this type of monitoring [5]. Not all states follow these guidelines reliably; thus quality of VMT data may not be uniform for all states. If county level VMT data are not available, state level VMT data or annual gasoline consumption figures [6] are used; the gasoline consumption figures are based on reports from state tax agencies and are considered accurate. County level fuel consumption values are obtained using algorithms that are based on vehicle registration data; these data are then combined with an averaged fuel efficiency for each vehicle type to obtain VMT estimates. Thus the resulting data are a mix of top-down disaggregation and county level data.

Improvements to the area source activity rate methodologies include periodic revision of the algorithms and surrogate data used and development of new/refined algorithms and surrogates to be included in the analyses. These revisions are particularly critical for mobile sources; not only because this is a major source of NO_x and hydrocarbons, but also because the surrogate data used are quite variable from year to year and location to location. The use of national averages should be discouraged; improvements in the current methodologies can be obtained if more regional figures are calculated and used.

Ash and Sulfur Content of Fuel

The ash and sulfur content of a fuel burned are needed to estimate the particulate and SO_x emissions from the combustion of fossil fuels; since these source categories are major emitters, errors in the values of these parameters can have major impacts on emissions totals. The American Society of Mechanical

Engineers (ASME) has developed test codes [9,10] for determining fuel ash and sulfur content with known reproducibility and/or precision. Users of large quantities of fuels periodically test for these quantities; economic factors, such as fuel prices and/or allowed emissions, are usually the reasons for performing these tests. The inherent variability of both ash and sulfur contents, combined with the large size of lots tested, can introduce considerable errors in emissions calculations. To improve emission values, frequent measurements taken over long periods of time are needed to obtain representative monthly or annual averages. Short average data are critical when emissions are to be estimated for shorter time periods.

Efficiency of Control Devices

Values for the efficiency of control devices are needed to estimate particulate and SO_x emissions from stationary sources equipped with these devices; in general, these are large fuel combustion or industrial sources. Methods for determining the efficiency of control devices include: (1) stack measurements (before and after device), (2) use of manufacturer-supplied design efficiencies (3) estimation or guess. A well designed and implemented stack measurement program should reduce the error in the resulting values to the statistical errors of the measurements; however, additional errors are introduced when data taken for limited time periods are extrapolated to larger periods. Performance degradation over time, equipment down time and possible equipment malfunctions can introduce errors in these longer term averages. Use of manufacturer-supplied design efficiencies introduces additional errors if these values are based on operating conditions that are not representative of the plant in question. The methodologies used for estimation or guess must be individually examined before errors can be identified.

Actual measurements of control device efficiency over extended periods of time and close monitoring for unusual occurrence are necessary to obtain reliable estimates for this parameter.

Control devices are also used on certain types of mobile sources; the efficiencies of these devices are included in the algorithms used by MOBIL3. Values used in the model are generally group averages; availability of representative values for different conditions is an important consideration. Improved measurement programs to provide representative values which account for vehicle type, age, and operating conditions are needed to refine current values of control device efficiency.

DISAGGREGATION FACTORS

The time and space scales of emissions data are provided by the scales at which values of emissions estimation parameters can be obtained. Disaggregation factors are applied to the resulting emissions data when finer time and/or space resolution are desired. In general, data for most emissions estimation parameters are available on a monthly, or at least a yearly basis for both point and area sources; individually for point sources or on a county, state or national level for area sources. For legislative and assessment work, county level area sources and individual point sources on a yearly timeframe provide adequate resolution; thus the methodologies that have evolved to estimate emissions already incorporate "disaggregation" algorithms to obtain this level of resolution. As new sources of monthly data on a more regional basis are developed, the algorithms should be modified to incorporate these new data, calculate emissions at the finer resolution and aggregate to higher levels if desired.

A relatively new use of emissions inventories is to provide input for numerical models of atmospheric long-range transport and transformation processes; it is this use that has required finer spatial and temporal resolution. Because of the complex chemical reactions that take place in the atmosphere, more detailed speciation is needed for hydrocarbon, SO_x, NO_x and particulate emissions. Emissions of some of the additional pollutants, such as primary SO_4^{2-}, have been separated from the "generic" pollutant and are calculated using newly developed emission factors and the methodologies described above.

The approach taken to provide finer temporal, spatial and species resolution has been to develop disaggregation factors, i.e., averaged parameters that are applied to the emissions values to provide the resolution needed. These parameters are treated in a similar fashion to emission factors; averaged values are developed and applied all sources in a (hopefully!) homogeneous category. To date, very few disaggregation factor values have been developed based on measurement programs; most are based on paper surveys, surrogate data and estimation algorithms. Methodologies to develop and apply disaggregation factors are too numerous and complex to detail here; suffice it to say that their application not only greatly increases the uncertainty in the emissions estimates, but also adds complexity to the statistical formulations needed to correctly quantify this uncertainty. Quantification of uncertainty in emissions values is addressed in a following section.

COMPLETENESS OF THE INVENTORY

There are two major questions to be answered in determining the completeness of an emissions inventory: (1) have all categories of sources emitting the pollutants of interest been identified? and (2) have all existing sources within those categories in the region of interest been included in the inventory? The latter question is somewhat easier to address; once the categories have been defined, surveys that include many sources of data, such as industry specific publications, economic reports, scientific literature, etc. will help present a fairly complete picture of the existing major sources. The ultimate check, although an expensive one for large geographic regions, involves surveying areas covered by the inventory.

The question of identifying all possible categories of sources emitting the pollutants of interest is a more difficult one. Literature surveys as described above can provide a starting point; the data thus collected must be combined with engineering knowledge of each process to obtain an assessment of the importance of the process emissions. The continuing change within modern industry, where new processes are constantly being developed, makes this a difficult question and one that must be constantly addressed.

In general, determining inventory completeness is more difficult when addressing area sources. Most of the major stationary sources are well known and under legislative control; even the smaller stationary sources are slowly being included in regulatory activities. Area sources, because of their small and disperse nature, are more difficult to identify and address. Until recently, emissions from processes such as publicly owned treatment plants, bulk terminals and plants, crude oil and natural gas production fields, etc. were not accounted for in the major existing inventories. Areas that are not generally addressed yet include light-duty diesel vehicles, frost control, rocket testing and launching, etc. The importance of these new areas must be assessed in light of the targeted use of an emissions inventory.

QUANTIFICATION OF UNCERTAINTY

Quantification of the uncertainty associated with estimated emissions is necessary for any realistic use of these values. Easier said than done! While standard statistical formulas have been developed to calculate quantities such as variance, standard deviation, bias and mean square error (MSE), and to use these values in error propagation calculations, these formulas are based on the availability of measurement data and on assumptions (normality, independence of parameters, etc.) that are usually not available and/or applicable in the emissions estimation methodologies described above.

The first projects that addressed the problem of assigning, in statistical terms, quantitative values to the uncertainty of emissions data were undertaken in the early 1970's [11,12]. These projects, as well as others which followed in the early 1980's [13-15], based their calculations on the statistical approximations for error propagation derived for "small" error values. Subsequent work [8,16-19] has addressed the problem more rigorously in two areas: (1) theoretical studies in error propagation without restricting assumptions and (2) assignment of realistic quantitative values to the uncertainty in the values of the emissions estimation parameters. This new work has demonstrated that:

1. In order to obtain a correct quantification of the error in emissions values we must accurately define which parameters are used to estimate what quantities, i.e., the error that results from the use of an averaged emission factor to estimate the true mean emission factor for the entire source population is not quantified by the same formulation as the error that results from the use of that averaged emission factor to estimate the emissions from one source or the error that results from the use of that averaged emission factor to estimate the emissions from a group of sources. For emissions inventory work, the last two are the quantities of interest.

2. The uncertainty in emissions values that results from the use of averaged values as parameter estimates is quite different from the

uncertainty that results when the parameter values are estimated for each individual source. Statistical formulations needed to calculate the MSE when averaged parameters are used have been developed for the two-parameter emissions calculations [18].

3. Theoretical implications of non-independent parameters and time-varying parameters on emissions uncertainty calculations have been examined; derivations based on autocorrelation models and covariance values currently in use for emissions inventory work indicate that no appreciable bias is introduced in the error estimation [18]. These studies should be repeated if autocorrelation and/or covariance change.

4. Estimation of the uncertainty associated with the values of the parameters used in emissions calculations is complicated by several factors: (a) non-normality of the base data, (b) possible non-representativeness of the sources sampled (introduction of bias), (c) lack of the original data and/or documentation used in deriving the parameter values [8,17].

Improvements in this very important area of inventory work should proceed on several fronts. Theoretical work should be continued to develop uncertainty calculation methodology that includes realistic representations of all calculations used in inventory work, including those needed to quantify the uncertainty associated with the values of the emissions estimation parameters. Work done to quantify the uncertainty associated with current values of the emissions estimation parameters should be continued to refine the data available and should be coordinated with work in the theoretical area to insure use of appropriate methodologies in all segments of this work. Standards should be set for the quantification of uncertainty associated with new values for parameters used in emissions calculations; uncertainty calculations should be incorporated as an integral part of all projects that develop these new values.

CONCLUSIONS

Since the estimation of the emissions from every source is an impossible task when compiling emissions inventories for large geographic areas, we cannot abandon methodologies currently in use that estimate the desired emissions. However, we must keep in mind the limitations imposed and the errors introduced by both the methodologies and data used to obtain the emissions values. The mass balance model approach used relies on engineering judgement of the process in question and on the use of either limited measurement data or on surrogate data to estimate the values of the model parameters. Thus, while results may represent fairly well yearly emissions from some emitting sectors in large geographic areas, results for other sectors may not represent more than a "guessestimate". Errors can increase exponentially when more spatial, sectoral, species and/or time resolution are desired. Unless supported by a direct measurement program, emissions estimates obtained by the methodologies described above should not be used to represent conditions during specific timeframes.

RECOMMENDATIONS

Upgrading the quality of the emissions estimates for large geographic areas is a resource intensive project; priorities must be set to select the most critical tasks for each application. Recommendations outlined in the body of this paper can be summarized as follows:

1. Emission factors.

   a) Review of the types of sources included in each SCC category should be undertaken to insure that each category is still as homogeneous as possible. If needed, reclassification of sources and/or development of new SCC categories should be undertaken.

   b) If the current value of an emission factor has been transferred from another category, or is based on a scant and/or old set of measurement data, a new measurement program should be implemented using proven analytical methods and state-of-the-art quality assurance/quality control techniques to derive upgraded values.

   c) If the current value of an emission factor has been derived using models and/or surrogate data, the algorithms and surrogate data in use should be reviewed in light of current knowledge of the processes and all known data sources. If now feasible, new measurement programs should be undertaken. This recommendation is specially applicable to the calculation of emission factors for the transportation sector, where studies have demonstrated the importance of further refinement in the algorithms used, in the treatment of current parameters and in the surrogate data values used.

2. Activity rate.

   a) For stationary sources that currently report activity rates, it would be highly desirable if all agencies that require such data would standardize on the methodologies and units used by the sources to report these values. An alternative would be to develop standardized, automated ways of calculating these conversions.

   b) Some of the methodologies and data currently used to estimate non-mobile area source activity rates were developed in the 1970's; each area source sector should be examined to determine if the methodologies and surrogate data used can be improved and which data sources provide the best and most current data.

   c) Activity rates for mobile sources are expressed as vehicle miles traveled (VMT) and are estimated using a mix of direct measurements and surrogate data and algorithms. National average values are used for some of the parameters included in these calculations (ex, vehicle type fuel efficiency, age distribution of the vehicle fleet); since these parameters can be highly variable depending on location and/or year, values used should be estimated on a more regional and current basis.

3. Ash and sulfur content of fuels. Due to the inherent variability of these fuel parameters, frequent measurements over long enough time periods represent the most feasible way to improve the values for these parameters.

4. Efficiency of control devices.

   a) Stationary sources. To improve the values used for this parameter in the calculation of emissions from stationary sources, either actual measurements of control efficiency over extended periods of time or good values for a range of operating conditions and constant monitoring of these conditions are needed.

   b) Mobile sources. Improved measurement programs by vehicle types, including ranges in operating conditions, age, etc., together with better data for surrogates such as the age distribution of the vehicle fleet by geographic location, average efficiency of control devices by vehicle types, tampering rates, etc. are needed to improve the resulting emissions values.

5. Completeness of the inventory. The ultimate completeness of an inventory is difficult to prove; only detailed surveys of the geographic area in question can provide definite answers. Such surveys are resource intensive activities, well beyond the scope of most inventories. Paper surveys can go a long way to help insure completeness; however, even with good bibliographic referral systems, researchers can be buried among a plethora of disperse information. Nonetheless, the issue of completeness should be explicitly addressed by all inventory compilation projects; documentation should clearly state the tasks performed in this area, the sources consulted, etc.

6. Quantification of the uncertainty of emissions values. Refinements in this area are need on several topics.

   a) The basic theoretical work must be extended to include realistic formulations to quantify the uncertainty associated with all calculations used in inventory work, including the uncertainty associated with the values of the emissions estimation parameters.

   b) Work done to quantify the uncertainty associated with current values of the emissions estimation parameters should be continued and coordinated with the theoretical work.

   c) Standards should be set for the quantification of the uncertainty associated with any new values developed for the emissions estimation parameters; these calculations should be incorporated as an integral part of all projects that develop these new values.

ACKNOWLEDGEMENT

This paper has been authored under contract DE-AC02-76CH00016 with the U.S. Department of Energy as part of the National Acid Precipitation Assessment Program (NAPAP). Accordingly, the U.S. Government retains a non exclusive, royalty-free license to publish or reproduce the published form of this contribution or allow others to do so, for U.S. Government purposes.

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