

EMISSIONS DATA

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OVERVIEW AND SUMMARY OF RECOMMENDATIONS

P.A. Makar and M.D. Moran

The emissions from the multitude of sources in the environment are critically important inputs in the airshed models that must be used to assess impacts or reactivities of VOCs. The amount of emissions determines the nature of the chemical environment in which the VOC reacts, and thus affects not only the absolute amount of ozone and other secondary pollutants formed, but also the incremental effects of any additional VOC on the formation of those pollutants (i.e., the VOC reactivities). Emissions data also impact directly on government policy. Changes in emissions levels over time reflect the effectiveness of government regulations, as well as changes in activity level. The accuracy and completeness of emissions databases are therefore of great importance.

The creation of an emissions database takes place in several stages, with each stage having associated errors and uncertainties. From sources such as Moody et al (1995), the SPECIATE website (Ryan, 1998), Moran et al (1997), and the AP-42 database (EPA, 1995), typical stages for determining anthropogenic emissions are as follows:

- Source profiles (which provide a breakdown of the total VOC emitted from a given source type into a detailed VOC fractionation) are constructed from measurements. A limited number of profiles are currently available (e.g. about 300 used in SPECIATE).
- Source types are categorized using Source Classification Codes (SCC), Area Mobile Source codes (AMS) and Standard Industrial Classification codes (SIC). Typical emissions processing systems have thousands of source classifications (Ryan, 1998).
- Each classified source type is assigned a source profile from the set of available profiles.
- The total annual VOC emission for each source type is determined from the product of the appropriate emission factor, activity rate, and control factors (AP-42; EPA, 1995).
- Individual sources from the database are assigned total annual VOC emissions, source classification codes, and source profiles.
- The total annual VOC emission rates may be multiplied by meteorological factors (e.g. to include dependence of emissions on ambient temperatures).
- The source profiles, linked to the individual sources in (5), are used to convert the total annual VOC emission into a speciated annual VOC emission.
- The detailed speciation is lumped into a smaller number of species for modelling purposes (e.g. Middleton et al., 1990).
- Temporal allocation occurs; temporal factors are applied to the model species annual emissions to create seasonal, daily, then hourly emissions estimates.

- Spatial allocation factors are used to create the spatial distribution of area sources. Smaller point sources are sometimes included with the area sources.

For biogenic emissions, the processing differs somewhat:

- Spatially varying biomass factors (mass of emitting foliage / unit horizontal area) are linked with monthly frost/seasonal variation factors and a database of vegetation categories.
- Net vegetation standard emission rates (30°C, 1000 $\mu\text{mol}/\text{m}^2/\text{sec}$ photo-synthetically active radiation (PAR)) are calculated for each part of the vegetation grid.
- Temperature and / or PAR corrections are applied for the meteorology at the site.

For both forms of emissions, regridding from the database spatial grid to the model grid is often required.

Each of the stages listed above has uncertainties, many of which have not been characterized in a quantitative way. The limited number of source profiles compared to SCC, SIC, or AMS codes is an example. The extent to which the available source profiles adequately represent the much larger set of source classifications is unknown. The source profiles themselves are the result of a relatively small number of measurement studies; in some cases a single study is used to represent the emissions from an entire industry. Errors in the total annual VOC emission rates are usually not estimated. Similarly, errors in temporal emission factor estimates are usually not estimated. For the biogenic emissions, the existence of detailed vegetation databases is required; these are not always available with sufficiently fine spatial detail for adequate resolution of emitting regions.

Some of the sources of uncertainty have been characterized. The effect of lumping detailed speciation emissions into model speciation has been examined by Middleton et al (1990) - comparisons there and in later work (Kuhn et al., 1998; see also “Lumping” in the Atmospheric Chemistry chapter of this assessment) indicate that the effects of speciation in model ozone production may be a smaller source of error than other model approximations. Similarly, the procedures for determining standard emission rates for specific vegetation types are well established and the uncertainties associated with these measurements are known (c.f. Guenther et al., 1996).

From the standpoint of VOC reactivity and ozone formation, it is essential to determine which emissions-related uncertainties have the biggest impact on the predicted reactivity of the atmosphere. This in turn could be used to design measurement initiatives directed at reducing the most significant uncertainties. Sensitivity studies of model results towards errors in the temporal factors, source profile assignment and total VOC emission rates should be directed towards determining which aspects of the emissions system have the biggest impact on reactivity. A few preliminary studies of this nature are discussed in the “Emissions Uncertainty” section, below. Emissions uncertainties can contribute 30 to 50% of the overall uncertainty in O₃ predictions, while the contribution to uncertainty in VOC reactivity may be smaller (8%).

Certain aspects of anthropogenic emission factor calculation are described in the “Status of Anthropogenic Emissions” section, below. There it is noted that all available data for point sources should be taken into account, for both minor and major emitters. In addition, time factors and other parameters have typically been estimated based on episode days; non-episode, non-attainment days may therefore be incorrectly represented in emissions databases. Biogenic hydrocarbon emissions are described in a separate section, below, with the need for improved vegetation databases noted as the most significant need for further research.

Emission reduction effects such as volatility suggest that some AP-42 emissions estimates may not adequately describe the amounts of VOCs that ultimately enter the atmosphere. These arguments are presented separately in the “Emissions and Volatility” chapter of this assessment, since they are relevant to assessing reactivities of individual VOCs as well as the quality of the overall emissions inventory.

Overall, the following recommendations can be made based on the work outlined above and in the subsequent sections:

- There is a clear need to determine the impact of the approximations used in the emissions assignment process on the results of models used for reactivity calculations. Sensitivity analyses should be conducted to identify the effect of approximations in temporal factors, source profile assignment and total annual VOC emission rates on model reactivity estimates.
- Very limited research to date suggests that, at least in the Southern California area, emissions uncertainties have a more limited impact on our ability to quantify VOC relative reactivities. In part, this makes sense since the addition of more VOC tends to reduce the reactivities of all VOCs, such that the relative reactivity of one species to another is not so impacted. However, the robustness of this conclusion is not yet demonstrated. For example, the uncertainties in the biogenic emissions are large, and may have a more marked effect in domains like the eastern United States. Given the continued, and seeming perpetual existence, of large uncertainties in emissions, particularly biogenics, this issue should be explored further.
- The relative impact of emissions uncertainties on both absolute O₃ mixing ratios and on the estimated VOC reactivity should be examined in sensitivity analyses. The impact on O₃ is expected to be larger than on relative reactivities, but this should be confirmed in simulations for different airshed conditions.
- There is a need for the improvement of the vegetation land-use database and area average factors used in biogenic emissions estimates to reduce their uncertainty. Other significant data gaps include lack of factors genetic variability within species and for wounding of vegetation, inadequate characterization of emission underestimates due to chemical losses, and the need for refinement of emission activity and escape efficiency parameters.
- EIIP procedures, operational information, and stack sampling should be used to ensure adequate speciation of anthropogenic VOCs, with the encouragement of industry participation.

- Time factors for apportionment of VOCs on non-episode days need to be designed, as well as appropriate changes to engineering and VOC composition parameters to reflect non-episode emissions days.
- Updates to the limited set of speciation profiles are essential for improvement to the emissions databases.
- Volatility factors need to be designed to account for the effects of volatility on net emissions. This is discussed further in the “Volatility and Fate” chapter of this assessment.

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EMISSIONS UNCERTAINTIES AND IMPACT ON REACTIVITY ASSESSMENTS

A. Russell

One of the great uncertainties in photochemical modeling of ozone is in the estimation of emissions. As reviewed in the recent NARSTO assessment, the uncertainties in the emissions, primarily of VOCs, is likely the greatest limitation in our ability to improve ozone simulations. This is true for both anthropogenic and biogenic emissions. This impacts our ability to quantify reactivity as well, but likely not to the same degree. Limited studies suggest that the impact of emissions inventory errors on reactivity quantification is significantly smaller than on the absolute ozone estimates.

Examining, first, the issue of uncertainties in our ability to simulate ozone, Hanna et al., (1998), Gao et al., (1996), Russell and Dennis (1999) and Bergin et al., (1999) find that the uncertainties in absolute ozone are on the order of 25% (depending on study, all of which used different domains and parameters for uncertainty quantification). This can be compared to typical model performance which finds normalized errors on the order of 35% (Russell and Dennis, 1999). The reason for the increased error when looking at model performance is that the latter also includes errors in the meteorological fields (which were not completely treated in the other studies), grid resolution errors (which is linked to the commensurability problem), limitations in the model formulation and errors in the spatio-temporal distribution of emissions. We can not, yet, fully account for the myriad of contributors to uncertainty, so the above studies identified the major contributors for study. As an example of the importance of the emissions contributions to overall uncertainty, the Bergin et al., (1999) study found that emissions related uncertainties contributed between about 30 and 50% of the overall uncertainty in the ozone predictions.

While errors in the emissions estimates may be a major contributor to the errors in the absolute ozone, it is not necessarily the case that they play the same role in quantifying VOC reactivity. The reason lies in expanding the reactivity estimation procedure as a Taylor series. Most of the terms between the base simulation and the perturbed case are the same, so they drop out when taking the difference. It is the second order terms that become important. A study by Yang et al., (1999) explicitly identified and the major contributors to overall uncertainty in relative reactivities of compounds in the Los Angeles area. While they found that uncertainties in the VOC emissions inventory (which were assumed to be log-normally distributed with a standard deviation of 2) accounted for about 8% of the total uncertainty ranking about 6th or 17 parameters tested), chemical parameters (e.g., the HCHO photolysis rate and the NO₂ +HO reaction rate) had a greater influence. Their overall reactivity estimates and uncertainties were similar to those found by Bergin et al., (1996), which did not include emissions uncertainties. Uncertainties in the NO_x emissions had a significantly smaller impact. This study suggests that, at present, errors in the emissions estimates are not limiting our ability to quantify reactivity. However, this results of this study may not hold for other regions, and this finding should be tested.

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STATUS OF USE OF ANTHROPOGENIC EMISSIONS IN AIR QUALITY ASSESSMENTS

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This report attempts to provide a brief summary on the estimation and use of anthropogenic emissions in the assessment of ozone air quality. The report draws heavily from the critical review papers developed under NARSTO¹⁻², EPA³⁻⁸ reports, and from Bergin et al (1998). While estimation and use of emissions in determining air quality has been a common practice, it is increasingly becoming important to characterize issues such as uncertainties and errors in the estimated emissions or in speciation characteristics, etc., and to assess their effect on air quality.

Traditionally emissions are often estimated and reported on an annual basis in terms of their broad source categories – stationary (point and area), mobile, and biogenic. However, in recent times there has been a need for information on emissions at a much more detailed level resulting in the development of emissions modeling systems¹⁰⁻¹³. The references 1 through 8 provide a wealth of information on the development of the inventories and some of the problems associated with them and their use in assessment of air quality. This report attempts to look at some of those issues from an end-user point of view and the need for improvement.

Stationary Sources

This broad categorization includes all sources that do not get classified to be associated under the mobile category. Thus for all practical purposes all stationary sources can be characterized as point sources, implying information on the location of the source, and pertinent stack information are available. However, it is often impossible to acquire relevant information for all but only those individual sources that are characterized by high emissions and associated with stacks, while the remaining sources are too numerous to be treated individually and therefore they fall under the area source category. Typical examples of the point sources are electric utility generators, while gasoline stations fall under the area source category. In addition, the distinction between point and area sources could be based upon a set emission level cut-off by pollutant. For example in the case of a severe ozone non-attainment area, the emission reporting requirements could seek information from sources that are at or above 5 tpy of VOCs or NOx, while in other areas of the state emissions reporting requirement are set at 100 tpy level. It is important to take into account the definition of point source and the attendant information that is available to characterize the source emissions since such information and quality of the data could vary significantly across geo-political sub-divisions of a state or a region. Further, it should be noted, that the quality of information sought to estimate emissions from point sources should not be limited only to the large emitters but allowance be made to acquire data for the low emitters as well, because such information provides critical backup in the development of controls as the cut-off limits are reduced.

As noticed in the draft EPA Guidance⁴, the emissions data are to be speciated into chemical classes as required by the air quality models. Also, the emissions data are to be allocated on a diurnal basis or temporal allocation to provide hourly information for the air quality model. The speciation and temporal allocation processes are independent operations and in many instances currently utilize default profiles depending upon the nature and type of a source.

Point and Area sources

In many instances the emissions and other pertinent information that are gathered by the regulatory agencies is based upon actual operations or from the permit to operate the facility. In recent years the emissions have also been obtained from continuous emissions monitoring (CEMs) systems. These data require appropriate adjustment for use in air quality models. Such a manipulation is accomplished by emissions modeling tools such as EPS2¹⁰, EMS95¹¹, SMOKES¹², FREDs¹³, and CEPS¹⁴. These tools have built in checks and balances to ensure that the data provided are within standard engineering practices in terms of information such as stack parameters, and emission estimates based upon the SCC. However, experience from the OTAG¹⁵ has shown that there may be a need to go-back, as quite often the engineering data may be missing or faulty and even fundamental datum such as the location of the point source may be in error due to such things as data entry, conversion from English to Metric system of units or vice-versa, etc. Although procedures used by states to generate emissions is undergoing changes through the EIIP³⁻⁸ process, questions remain as to the allocation of resources and the cooperation of the facilities in providing these data requests.

As noted above the temporal profile is often assumed in the apportionment of the emissions, and allocated to each hour. Along with the emissions, other engineering parameters are also needed in the air quality model. However, such information is often not available, resulting in the use of default data for the entire period of air quality simulation. Such a procedure may be reasonable when simulating episodic events that last 2 to 3 days and are generally associated with high 1-hr ozone levels that tend to occur on high temperature days and nearly stagnant conditions. However to assume or use default engineering data reflecting episodic conditions may not be meaningful and may result in over predictions in situations that cover extended periods that contain non-episodic days. Examination of ambient air quality data suggests that exceedances of the 8-hr ozone NAAQS could occur under non-episodic conditions as well. Therefore there is a need to address on how to apportion the emissions between episodic and non-episodic days as well as how to estimate similarly the changes in the engineering parameters such as stack temperature and stack flow rate, etc. Also, it is important to address if there are changes or differences that may accompany in the level of VOC emissions and its composition between episodic and non-episodic conditions in the development of emissions. Another important parameter that may not be readily available is the information on the operational hours of a facility. Depending upon the time-of-the day and operational hours of the facility, emissions are to be apportioned on hour by hour basis and may have an effect on the air quality predictions if the emissions are wrongly characterized.

Often a majority of the area source emission categories are estimated based upon population as a surrogate parameter. The emission factors for area source categories are often adapted from EPA, unless data are available from local surveys or other such data bases. EIIP has recommended preferred and alternative methods for estimating for about 15 source categories that are predominantly VOC emitters. Often these methods require local surveys which will not be undertaken unless significant resources are directed by the state agencies for the development of emission inventories.

Mobile sources

Emissions from these sources are estimated in a top-down approach in which vehicle miles traveled (vmts) at the county level are utilized to estimate the emissions based on EPA's mobile emissions model. The emissions reported are in terms of NO_x, CO, and VOCs, while the air quality model requires speciated data. Currently there are some indications that the states may wish to develop information from a bottom-up approach. Such an effort has been undertaken in the Lake Michigan Air Directors Consortium modeling work for the Greater Chicago area ozone air quality assessment. This requires collection of VMT and vehicle class information on a link basis to estimate emissions with the emissions model.

Currently the mobile source model is undergoing updates and revisions. The model is not designed to provide speciated hydrocarbons but total hydrocarbons. The selection of an appropriate speciation profile that reflects the fuel characteristics of an area is a very important step in the emissions modeling system. For example, introduction of the reform-fuel (RFG) in ozone non-attainment areas in 1995 required the use of appropriate speciation profile and not the speciation profile of conventional gasoline. Often, such information may not be readily available, requiring the use of default profiles in air quality assessment.

Speciation

The emissions data are compiled and reported generally on an annual basis for the criteria pollutants NO_x, CO, and VOCs at the county level. However the air quality models require the emissions data in terms of individual chemical species or class of species. This is often accomplished by using EPA's models such as SPECIATE or FIRE, even though the majority of these data are based on testing of sources done in 1970's and early 1980's (see Ref. 1, 2). Use of updated speciation profiles should be considered **a must** for improving the emissions data base for use with air quality models. It should be noted that the VOCs estimated for some sources may not reflect the composition of the speciation profile due to elimination of some of those species through emission controls. Another important aspect of the speciated data is the need to ensure to what specific class of VOCs it is being referred to, such as - total organic gases (TOG), total hydrocarbons (THC), non-methane organic compounds (NMOC), non-methane organic gases (NMOG), reactive hydrocarbons (RHC), etc. Also, state level controls may result in revisions to manufacturing processes and chemical formulation that may be reflective of that state only and should be accounted for in the preparation of inventories for air quality applications.

Future work

It is recommended that the EIIP procedures be followed to the maximum extent where feasible in the development of emissions inventory. The data collected should include besides operational information, stack sampling or other information to facilitate speciation of the VOCs. Use of default information to estimate the emissions should be discouraged as there may be no incentive to improve the data. Industry participation in the development of speciation profiles is a critical first step towards addressing issues and controls based upon HC reactivity.

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BIOGENIC EMISSIONS AND CHEMICAL INTERACTIONS

P. A. Makar

The current state of the science for measurements and modelling of biogenic emissions has been extensively reviewed in Guenther et al. (1998). Some of the main conclusions of this work with regards to reactivity will be repeated here, as well as chemical effects from recent work not considered in the above reference.

Several sources referenced in Guenther et al. (1998) have shown that model predictions of ozone mixing ratios are strongly influenced by the effect of biogenic VOC emissions, and can determine the choice of a NO_x or VOC control strategy. Accurate estimates of biogenic emissions are therefore of great importance for estimating atmospheric reactivity.

The factors for determining biogenic emissions include area-average factors (emission per unit area of ground for standard meteorological conditions), emission activity factors (canopy-microclimate; leaf temperature and photosynthetically active radiation being dominant effects) and escape efficiency factors (the extent to which deposition decreases the actual emission to the above canopy air) (Guenther et al., 1998). In addition, recent work by Makar et al. (1998) and Forkel et al (1998) suggests that chemical losses may result in underestimates of biogenic emissions. The uncertainties associated with these factors and effects are a useful means of setting priorities for improving knowledge of biogenic emissions.

Uncertainties for the area average factors for individual tree species (which dominate biogenic emissions) were estimated at 20 to 50%, while crop emissions and non-terpene emissions from all plant species were estimated to have uncertainties of a factor of 3 or more. Genetic variability within and between plant species have shown large variations in emissions (factor of 4 to 6 within same species; Guenther et al. (1998)). In addition, the construction of vegetation databases was discussed in the context of the BEIS3 model. Uncertainties associated with composition and foliar density of landcover were given as ranging from 25% to a factor of 3, depending on the landscape. Some of the basic data for constructing the database was unavailable in Canada, necessitating the use of approximations for forest density parameters. The reduction of these uncertainties through the continuation of ongoing measurement programs was identified as a research priority.

The uncertainties associated with light and temperature emission activity factors were estimated at 15 to 25%, with seasonal factors being greater than 25%. Activity factors associated with wounding (damage of leaves by environmental conditions) had uncertainties “much greater” than 25%. Stomatal conductance factors, which affects the emission rate of oxygenated compounds, were assumed to have uncertainties of 25 to 50%. Leaf temperature parameterizations and those for soil temperature were assumed to have maximum uncertainties of 30 and 25%, respectively.

Escape efficiency was assumed to have an uncertainty of 5 to 30%.

One effect not considered for emissions estimates which has appeared in two recent studies (Makar et al., 1998; Forkel et al, 1998), is the effect of photochemical reactivity on estimates of emissions fluxes. Both of these modelling studies utilized one-dimensional canopy models to simulate biogenic emissions. In each case, detailed photochemical models were coupled with emission factor formulae and vertical mixing parameterizations. The effect of chemical removal on emissions estimates was examined by running “chemistry” and “no-chemistry” simulations. In Makar et al. (1998), neglecting effects of chemical losses was found to result in an underestimate of isoprene emission fluxes by as much as 40%. The study of Forkel et al. (1998) found a 20% isoprene emission underestimate if chemistry was neglected. Makar et al. (1998) noted that chemical loss effects would only be significant when the emitted compound was the most reactive in the ambient atmosphere; this has been confirmed by more recent work by Forkel (personal communication, 1998) where the absence of chemical losses was found to have a greater impact on the estimates of emissions of more reactive terpenes than for isoprene.

From the above uncertainties, the improvement of area average factors and land-use categories have the greatest need for further research, with activity factors, escape factors and chemical losses having a secondary and approximately equal effect on the overall uncertainty in the emissions. The effects of plant wounding could be considerable; research is required to determine the best means of estimating these effects on emissions.

Other reactivity related conclusions from Guenther et al (1998) included noting that pre-BEIS3 emissions models greatly overestimate alkane and aromatic compounds, while noting that alkene emissions do occur, model validation is required for conditions outside of midday, summer conditions, emissions of C₁ - C₃ species and oxygenated VOCs need to be better characterized, and expected changes in species composition and biomass density require better characterization.

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