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Uncertainty Calculations and Their Relationship to the Problem of Early Detection of Ozone Change

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Laboratory measurements of rate coefficients for chemical reactions are usually input to stratospheric photochemistry models as single "best" values for each reaction. In general, a laboratory measurement is done numerous times and our best knowledge is actually a central value and a distribution about that value. We have done a Monte-Carlo calculation of uncertainty propagation through a stratospheric photochemistry model in an attempt to include the full range of information concerning chemical reaction rate coefficients. The result of such a calculation is the prediction of probability distributions for the concentrations of stratospheric species and for the changes in ozone and other species due to specified perturbations.

These probability distributions can be used in several ways to attack the problem of model-measurement intercomparison and the search for the best way to detect future changes. We have examined several possibilities for using measurements to reduce the uncertainty range of model predictions. A study of the calculated present-atmosphere ozone concentration versus altitude indicates that it has little predictive capability for changes in the ozone column induced by chlorine perturbations. This probability distribution for ozone concentration at 40 and 50 km did overlap the data with the model being generally below the data. At 30 km the model was consistently higher than the data. In none of the more than 300 cases run did the model fall within the one-sigma variability range of the data at all three altitudes. The 50 or so cases which showed the best fit with the data showed no preference for either high or low ozone depletion predictions.

The best correlations for ozone depletion predictions was with lower stratospheric parameters. It was found that sets of reaction rate coefficients which predict high ozone depletions for small chlorine injections tend to yield larger disagreements with data than do other cases. Specifically, the model predicts lower stratospheric ClO concentrations which are on the high side of measurements. Since the ambient ClO/ClX is one of the best predictors of chlorine perturbation efficiency, models with low initial ClO are favored by the data. This eliminates virtually all cases which give more than 10% ozone change for continued release of fluorocarbons at their present rate. Examination of NO and NO_2 data indicates that the model tends to yield concentrations pretty much in the center of the measurement range. The measurements thus indicate that models on both extremes of the range of odd nitrogen compounds should be discarded. This procedure eliminates a

number of low ozone depletion cases (when odd nitrogen is high) and also a number of high ozone depletion cases (many of the same cases as discarded on the basis of disagreement with ClO). When all three of these are taken together, 125 of 329 cases remain with an ozone depletion of 3±2% for steady-state at the present fluorocarbon flux. This is contrasted with 6±5% before screening with the measurements.

An interesting poor correlation is that of the change in ozone due to a chlorine perturbation with the lower stratospheric hydroxyl radical concentration. We have begun examining the correlations of column ozone change with a number of measured parameters and are attempting to determine a strategy for the best method of early detection of change as well as for an identification of the cause of change.