Atmos. Chem. Phys. Discuss., 3, S411–S413, 2003
www.atmos-chem-phys.org/acpd/3/S411/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the relationship between acetone and carbon monoxide in air masses of different origin" *by* M. de Reus et al.

Anonymous Referee #2

Received and published: 21 April 2003

The paper presents a compilation of concurrent acetone and CO measurements from a number of airborne campaigns. As such the paper presents a useful contribution towards a better understanding of the atmospheric composition. Acetone is acknowledged as a potentially important source of peroxy and consequently hydroxyl radicals, in particular for the upper troposphere, where the production of OH from ozone photolysis is reduced due to, e.g., low water vapour concentrations. Therefore, a better understanding of the distribution of acetone and its different sources will help to better describe the distribution of OH and hence the fate of many longer lived trace gases in global models. Linking acetone concentrations to those of carbon monoxide, which is much better treated in these models is indeed provides a practical solution, since the simple chemical mechanisms used in global models cannot deal with the complex hydrocarbon chemistry that leads to secondary production of acetone.

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My comment concerns the way the experimental data are treated solely in terms of linear correlations. Points that need clarification concern the scientific basis for the linear model and the representativity of the measurements in the different compartments of the atmosphere:

An atmospheric measurement reflects the state of the atmosphere at a given point in space and time. Relating this state to the sources of the different species concerned, their ageing between emission and measurement as well as mixing of so-called air masses of different origin and history remains an as yet unsolved puzzle, that requires a full atmospheric chemistry and transport model which is run backwards in time. In their discussion, the authors state that the slope of the linear relation between acetone and CO should decrease with air mass age. This statement is somewhat misleading as chemical ageing within an air parcel will not produce a linear relation between two chemicals having very different reactivity, as is easily derived from a simple kinetic model. Pseudo-linear relations are always produced by mixing of air masses with distinct chemical composition. The tightness of the correlation gives us some hint on the homogeneity of the air masses and possibly on the time scales of the mixing processes involved. This part of the discussion needs revisions. I do not understand how can a correlation with a correlation coefficient of 0.19 be called significant. Figure 5 gives no hint on the existence of a linear correlation for the plume data either. The appearance of the data points in the scatter plots does not always back up the calculated slopes. This cannot be solely do to the way the measurement errors are included but must be do to a very uneven distribution of the data. Also, the upper end often shows some curvature that requires more attention.

In order for the data compilation to be useful for modelling purposes, I suggest to show box and whisker plots of acetone (or the acetone/CO ratio) as a function of CO. The number of measurements entering each box should be made fairly equal in order to bring out the density and of the data and the tightness of the correlation. The original data could be displayed as thin pixels and it would still be possible to include the

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calculated linear regression. Such a presentation would be of help for models and in addition would give a clearer view of the data. It would also be more in line with my initial statement of measurements representing primarily the state of the atmosphere.

In the discussion the different campaigns and air masses, I strongly missed an analysis with respect to air mass origin and time expired since potential emission on the basis of air mass trajectories. Such an analysis would indeed provide useful information on the potential for secondary acetone production from hydrocarbon oxidation relative to that of CO and would help to put the data into better perspective. These changes would significantly enhance the impact of the paper.

The accuracy and precision of the measurements should be included in the summary of ratios given in the conclusions. The strong correlation between acetone and CO in biomass burning plumes is not really backed up by the data presented (see above).

Minor comments: I suggest to exclude the model study from Table 3.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 1017, 2003.