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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 #1

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General Comments:

This paper presents acetone/CO ratios from 5 airborne field experiments STREAM97, STREAM98, LBA-CLAIRE, INDOEX and MINOS and demonstrates that the observed relationships differ between the marine boundary layer, free troposphere and stratosphere. This sort of information is very useful for modelling studies where no acetone concentrations are available and they are assigned on the basis of observed CO concentrations and a pre-defined relationship. The paper begins to provide some possible explanations for the different relationships, but overall, I was a little disappointed that it did not go further in providing more substantive reasoning. This could be aided by some calculations illustrating the sensitivity of the relationship to various factors or processes.

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Specific Comments:

As mentioned in the paper, the relationship between acetone and CO can be affected by many factors: the different lifetimes of the 2 species and thus the age of the air; the variation in lifetime in different locations of the atmosphere (e.g., increased photolysis of acetone at higher altitudes); the mixing of different air masses; fresh emissions (e.g., biogenic acetone emissions); secondary production (e.g., of acetone from oxidation of hydrocarbons). It would therefore appear that this relationship could be the function of many processes. Given that, it is somewhat surprising that the observed relationships are so strong and that they are so similar for the similar altitude ranges at very different geographical locations and that the difference between the relationships between the free troposphere and boundary layer are consistent between MINOS and INDOEX.

The authors conclude that this difference between the free troposphere and boundary layer indicate that the acetone is emitted by the ocean in relatively clean air masses and taken up by the ocean in polluted air masses. This may well be the explanation, but it is really only speculative with no real supporting evidence presented. For example, there is no discussion as to the oceanic concentration of acetone or air-sea exchange rate that would be necessary to produce both the positive and negative fluxes required to sustain the observed atmospheric concentrations. Can the difference in slope really be explained by air-sea exchange of acetone (page 1032 line 27 to page 1034 line 2) ? Comparing conditions for INDOEX and MINOS might also provide further insight given that the boundary layer acetone concentrations were generally higher for MINOS.

The marine boundary layer is a region of rapid OH production through the photolysis of ozone in the presence of the high water vapour concentrations. This might lead to the preferential loss of CO and possibly increased production of acetone via the oxidation hydrocarbons if the air is polluted. Have the authors considered how this might affect the acetone/CO ratio and their conclusions ?

The title of this paper suggests that the origin of air mass might affect the observed

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relationships. For the most part it was only the location of the sampling that was considered and not the origin of the air, which could be indicated by trajectory calculations or other tracers. The authors state that both the INDOEX and MINOS campaigns were performed downwind of major anthropogenic pollution sources (page 1025 lines 1-2). It would be useful to get an idea from trajectories and possibly other tracer ratios how aged the air was. Some of the free tropospheric air observed during INDOEX is likely to have been of recent marine boundary layer origin, it having been transported by convection. In contrast the free tropospheric air during MINOS was influenced by long-range transport from the US (page 1025, lines 11). How do these different origins affect the observed relationships ? Do the different emission sources (i.e., biomass and biofuel (India) and fossil fuel (Mediterranean)) affect the atmospheric acetone/CO ratios ? Can some of the variability in the each of the ratios plots be explained by varying origins and ages of the air ?

The difference in intercepts between flight campaigns is interpreted as indicating varying degrees of biogenic emissions of acetone. This source would lead to a positive intercept. Can the authors offer an explanation for the negative intercept ? A larger intercept is also found for the marine boundary layer compared to the free troposphere. This could be discussed in terms of the proposed oceanic source.

Technical Corrections:

I found the abstract rather confusing (lines 6-11) particularly when read as standalone. The ratio for measurements in the ANTHROPOGENICALLY-INFLUENCED marine boundary layer are given and then, by comparison with those in the free troposphere, it is concluded that acetone is emitted by the ocean in RELATIVELY CLEAN AIR MASSES. This appears to be illogical and confusing.

The authors continually switch between the terms acetone-CO and CO-acetone relation. It would help if a consistent term were used and given that the ratio is presented as acetone/CO, I would suggest using acetone-CO.

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Page 1024, line 19: than in the troposphere

Page 1029, line 18-19: than aged plumes

Page 1030, line 12: relation from decreasing with altitude

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