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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hydroxyl radicals maintain the self-cleansing capacity of the troposphere" *by* J. Lelieveld et al.

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The authors report on factors controlling the self-cleansing capacity of the troposphere through the abundance of hydroxyl radicals. They hypothesize that concomitant changes of reactive carbon and NOx caused by changes in anthropogenic emissions largely cancel out source and loss processes influencing the HO concentration. It is argued that 70-75% of methane, CO and NMHC is released to the atmosphere due to anthropogenic emissions. While for NOx this might be true, emission inventories for CO, methane and NMHC suggest that on a global scale only ˜23% (based on carbon) can be attributed to anthropogenic emissions. 15 % is due to biomass burning and at least 62% is primarily released and controlled by the biosphere (Guenther et al., JGR 1995). While agricultural practices and deforestation are usually seen as the main factors increasing emissions from biomass burning, it is also noted that human influence

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can lead to a decrease of emissions from burning vegetation (especially in the Northern Hemisphere). In some places like the US, for example, fire suppression over the last 100 years has probably led to a decline of emissions from biomass burning. In recent years catastrophic wildfires in the western US were primarily intensified due to the accumulation of biomass over the last decades. It has been shown that woody encroachment is becoming an increasing problem in many other places in the US; this seems to indicate that ecosystems were subjected to fires more frequently before intensive settlement started. While in-situ measurements during TOPSE showed that atmospheric chemistry in the remote atmosphere can be reasonably well represented based on in-situ measurements, the picture above densely forested areas is much less complete. For example: Faloona et al. (JGR, 2001) reported a significant nighttime production of HO. Di Carlo et al. (Science, 2004) claim that the HOx budget above a deciduous forest can not be closed based on our present understanding. These observations seem to indicate that our knowledge on biogenic VOC emissions is still scarce. Kurpius and Goldstein (GRL, 2003) speculated that current estimates of terpene emissions could be underestimated by as much as a factor of 10, while Kesselmeier (GBC, 2002) suggested that natural NMHC emissions could potentially even offset the terrestrial carbon budget. From our own measurements we have learned that current emission estimates of many oxygenated compounds might be a lower limit. Can the authors really claim that counteracting anthropogenic emissions of reactive carbon are the primary cause that has led to an increased loss of HO given these fundamental uncertainties and the magnitude of global natural reactive carbon emissions? Can the biosphere be assumed in steady state? It appears that effects influencing the abundance of natural VOCs, methane and CO through climate and landuse change could have also had a profound impact on a changing HO sink.

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