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Combining ambient measurements of OH radicals and OH reactivity in a tropical rainforest during the OP-3 project: Resolving the magnitude of the missing OH source

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The self cleansing capacity of the troposphere and the lifetime of key greenhouse gases are controlled to a large extent by the OH radical. In environments where biogenic VOC emissions are high and NO_x concentrations are low, for example in tropical rainforests, the current understanding of tropospheric chemistry suggests that OH radical concentrations should be suppressed. OH measurements made in such regions, however, have highlighted higher than expected OH radical concentrations that cannot be replicated by chemical models constrained with the currently known OH formation pathways (Lelieveld et al, 2008).

Here we report OH and OH reactivity measurements made during the OP-3 project that took place in the Borneo rainforest in 2008. Concentrations of OH displayed a clear diurnal cycle, peaking at solar noon, with significant concentrations observed: up to 8.7×10^6 molecule cm^{-3} (60 min average). Although $j(O^1D)$ levels and humidity were high, low ozone concentrations limited the rate of primary OH production from ozone photolysis. The measured OH reactivity was very high, up to 87 s^{-1} (corresponding to a lifetime for OH approaching 10 ms) and indicates that significant OH sources must be present – in addition to primary production – in order to maintain the elevated OH levels recorded. OH concentrations up to 6×10^5 molecule cm^{-3} (60 min average) were also recorded on several nights after $j(O^1D)$ had dropped to zero, suggesting a non-photolytic source of OH, most likely from the reaction of nitric oxide with HO_2 and higher peroxy radicals.

Using OH and the OH reactivity, and applying the steady-state hypothesis, we are able to fully resolve the magnitude of the missing OH source. We propose that the photolysis of HONO may represent an important OH source during the morning whilst direct formation of OH from photo-labile hydroperoxy-aldehydes produced via OH initiated oxidation of isoprene (Peeters et al, 2009) could act as an important source of OH throughout the afternoon when the missing OH source is at its maximum.

J. Lelieveld, T. M. Butler, J. N. Crowley et al.: Nature, 2008, 452, p.737 – 740

J. Peeters, T. L. Nguyen, L. Vereecken.: PCCP, 2009, 11, p.5935 - 5939