

Supporting Information

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1 Particle size distributions

Particle number size distributions measured during the observation period are shown in Figure S1 for particles with electrical mobility diameters 10 - 496 nm (5 - 16 February) and 11 - 460 nm (rest of campaign). Estimations of condensation sink were made from these measurements for the particle sizes measured during the entire campaign - (11 - 460 nm) using the method
5 described in Kulmala et al. (2001) and (2012). Consistent with previous observations of increased accumulation mode particles (0.1 - 2.5 μm) during the dry season (IOP 2), higher concentrations in this size range are evident in Figure S1b.

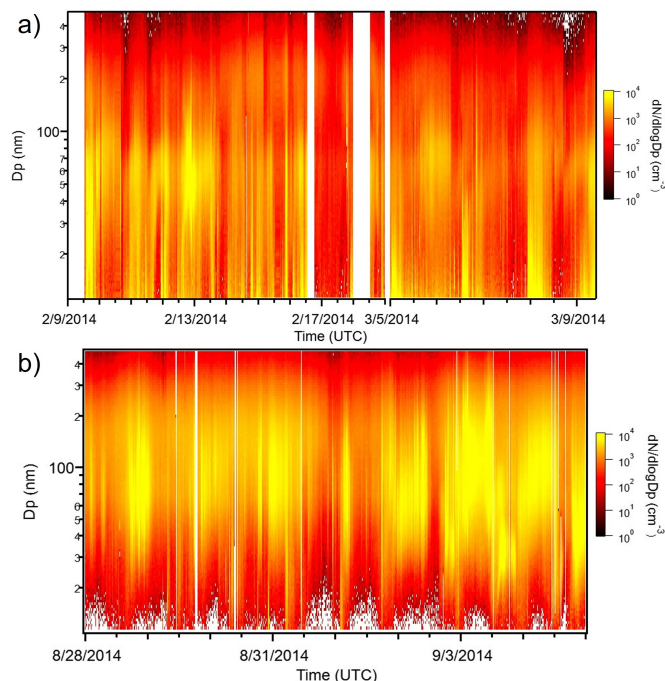


Figure S1. Particle number size distributions measured during IOP 1 (a) and IOP 2 (b). Note the time jump (2/19 to 3/5) on the IOP 1 time axis. Areas with white boxes represent periods where the instrument was not functioning properly.

2 Comparison of Radiation with H₂SO₄, OH

To determine the pseudo rate constant k' for Proxy 2, we determined the linear fit between measured H₂SO₄ and the product of SO₂, global radiation, and CS⁻¹ for the entire observation period (Fig. S2). The value of k' was found to be 2.43×10^{-10} m² s⁻¹ W⁻¹. A poor correlation ($R^2 = 0.59$) was observed between the measured and estimated values, largely from the IOP 2 data.

During both IOPs, but particularly IOP 2, there are many measurements of H₂SO₄ and OH when there is no radiation (GlobRad = 0) (Fig. S3), which can explain the poor correlation seen in Figure S2. These illustrate why Proxy 2 (Fig. 2b) does such a poor job estimating H₂SO₄, and why substituting radiation for OH can lead to order of magnitude underestimations.

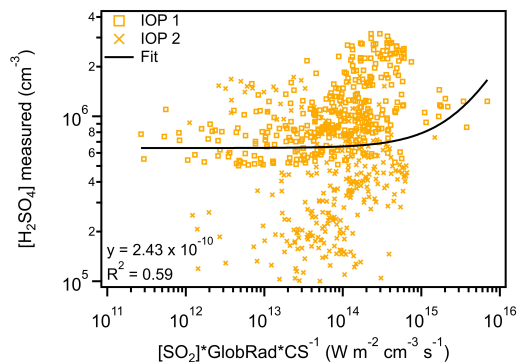


Figure S2. Measurements of H_2SO_4 for the entire observation period plotted against the product of SO_2 , global radiation, and CS^{-1} to determine the pseudo rate constant k' for use in Proxy 2 (Fig. 2b). The line represents the linear fit to determine k' .

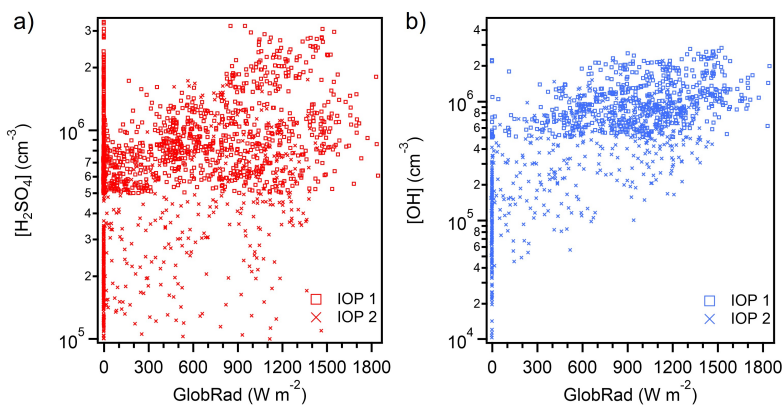


Figure S3. Comparison between measurements of (a) H_2SO_4 and (b) OH with global radiation. Note that there are numerous measurements of both species when global radiation was 0 W m^{-2} .

- 15 We also tested the best predictive proxy reported in Petäjä et al. (2009) (Fig. S4), which is very similar to Proxy 2 of this work. They only differ in the value of k' used in the estimation, which reflects the dependency of the proxy on radiation. While this proxy has improved estimations compared to Proxy 2, it still tends to underestimate measured H_2SO_4 , particularly for IOP 2.

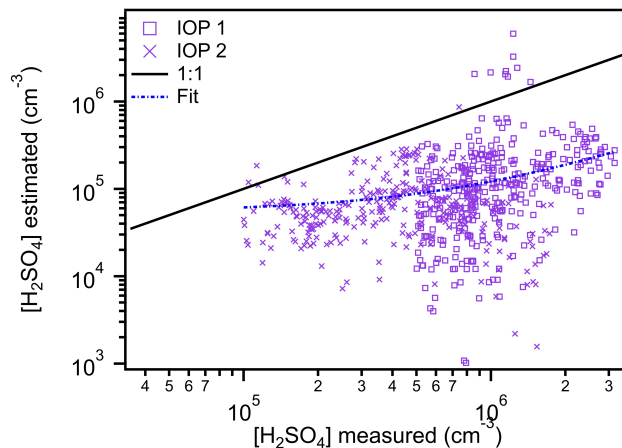


Figure S4. Estimated concentrations of sulfuric acid from the best predictive proxy reported in Petäjä et al. (2009) versus measured concentrations. Data from IOP 1 is plotted as boxes and data from IOP 2 is plotted as crosses. The 1:1 line is plotted to guide the eye. The fit line represents the fit between the measured and proxy-estimated values of sulfuric acid.

3 Analysis of alkene concentration for Proxy 4

- 20 To determine if Proxy 4 would give a better nighttime estimate of H_2SO_4 if more alkenes were included in this proxy, we determined estimations using the combined concentrations of isoprene and monoterpene as the Alkene term. These results show there is minimal difference between the two estimations (Figs. S5 and S6), supporting the hypothesis that OH oxidation of SO_2 is also contributing to nighttime H_2SO_4 in this region.

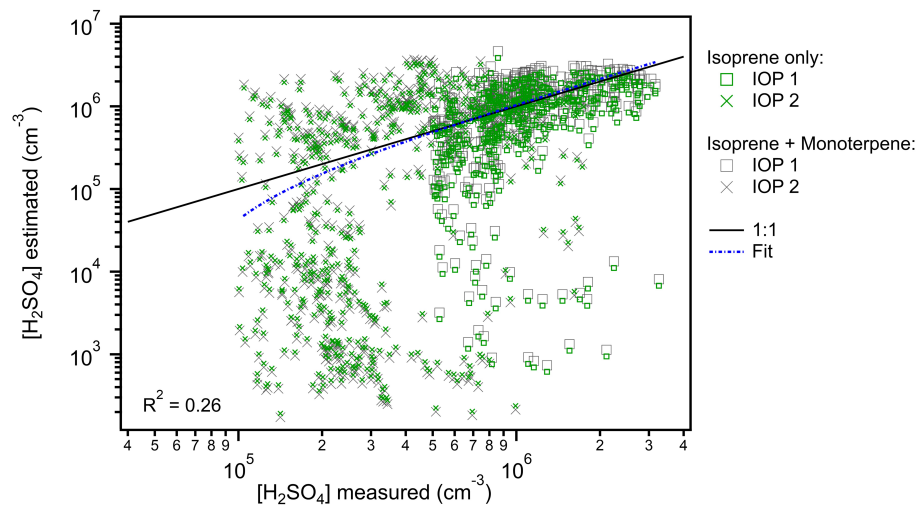


Figure S5. Estimations of sulfuric acid from Proxy 4 versus measured concentrations. Green points are estimates made only using the concentration of isoprene as the alkene term, and gray points used the combined concentration of isoprene and monoterpenes. Data from IOP 1 is plotted as boxes and data from IOP 2 is plotted as crosses. The 1:1 line is plotted to guide the eye. The fit line represents the fit between the measured and proxy-estimated values of sulfuric acid.

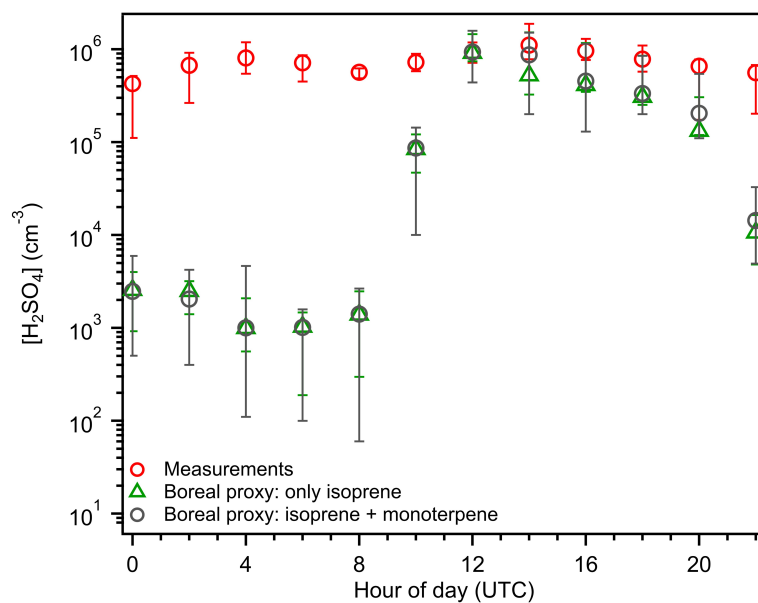


Figure S6. Two-hour averaged diurnal variation of the median sulfuric acid measurements (red), and estimations from Proxy 4 made using only isoprene concentrations (green), and using the combined isoprene and monoterpene concentrations (gray). Daylight hours: 08:00 - 22:00 UTC.