Supporting Information

# Surface Water Structure and Hygroscopic Properties of Light Absorbing Secondary Organic Polymers of Atmospheric Relevance

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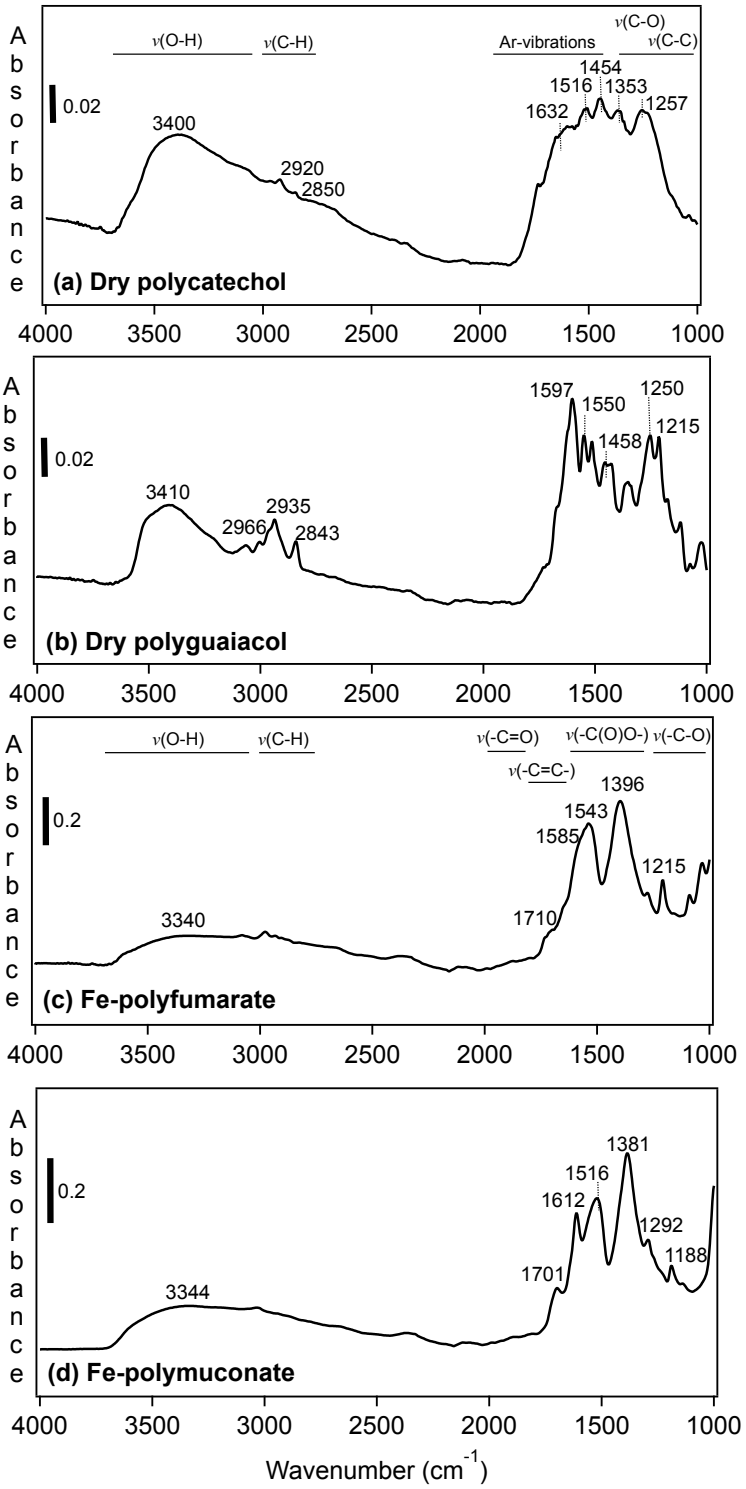


**Figure S1:** Representative DRIFTS absorbance spectra of solid organic polymers mixed with diamond powder after overnight drying at a flow of dry air: (a) 2% wt/wt polycatechol, (b) 2% wt/wt polyguaiacol, (c) 6% wt/wt Fe-polyfumarate, and (d) 7% wt/wt Fe-polymuconate. A spectrum of dry diamond powder only was used as the reference to generate these absorbance spectra. See reference 46 in main manuscript for assignment.

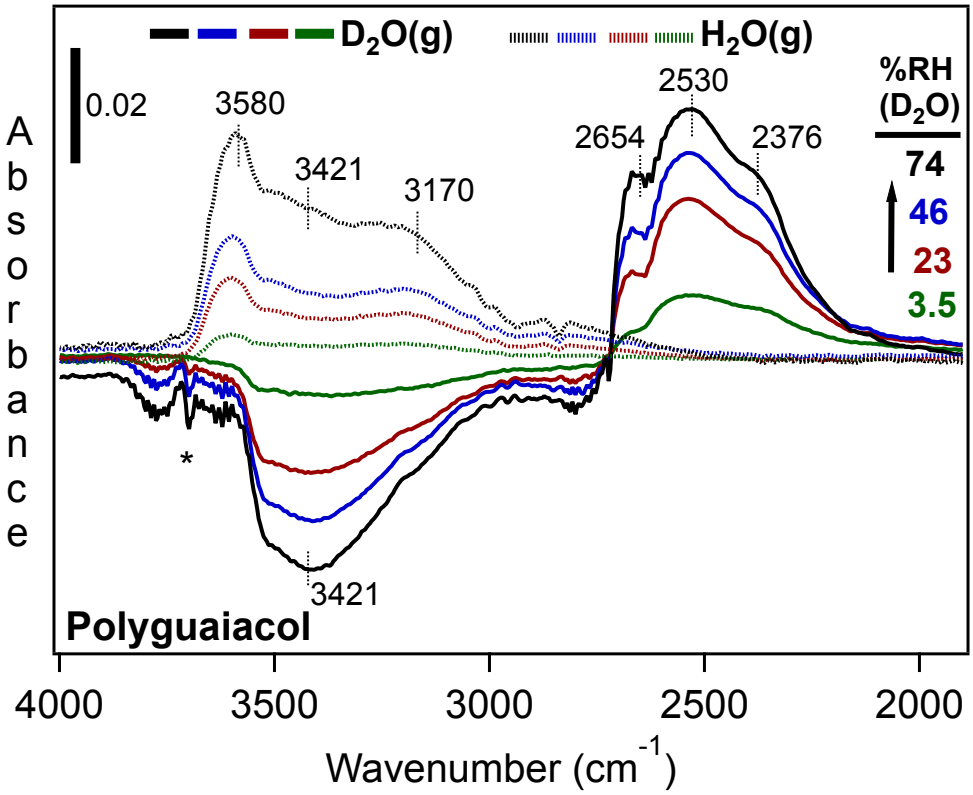


**Figure S2:** Selected DRIFTS absorbance spectra of surface water collected as a function of increasing %RH on a 2% wt/wt polyguaiacol/diamond sample. The solid lines are spectra collected using gas phase $D_2O$. The dashed lines are the same spectra shown in Figure 1b collected using gas phase $H_2O$ at 5.5, 25, 41 and 86% RH. The '*' donates incomplete subtraction of gas phase water lines.

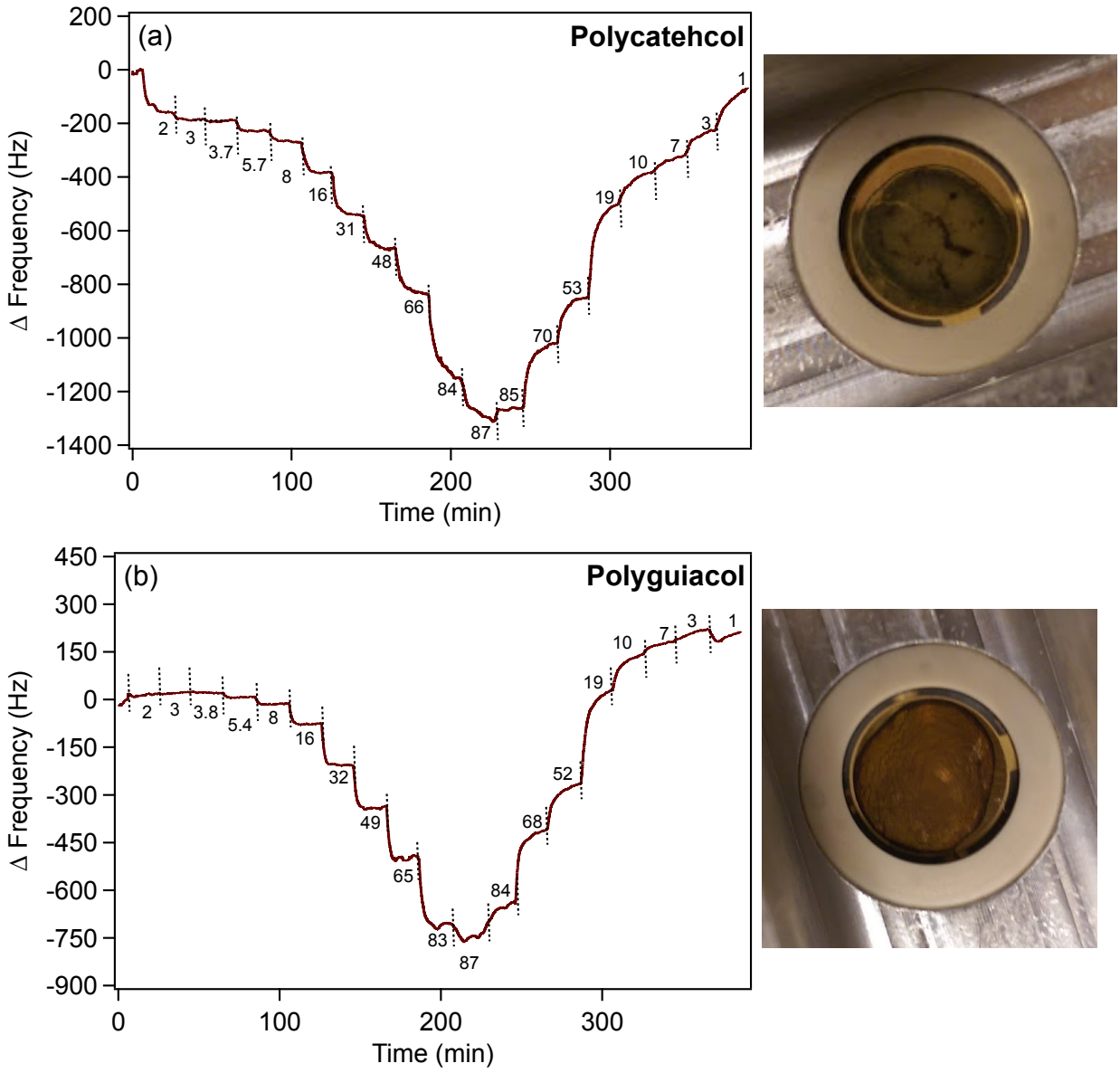


**Figure S3:** Raw QCM data showing change in frequency, $\Delta F$, as a function of increasing relative humidity (%RH, numbers between vertical dashed lines) relative to signal recorded while flowing dry air on (a) polycatechol (0.3 mg), and (b) polyguaiacol (0.1 mg) thin films. Photos to the right show representative samples prior to starting the water uptake experiments.

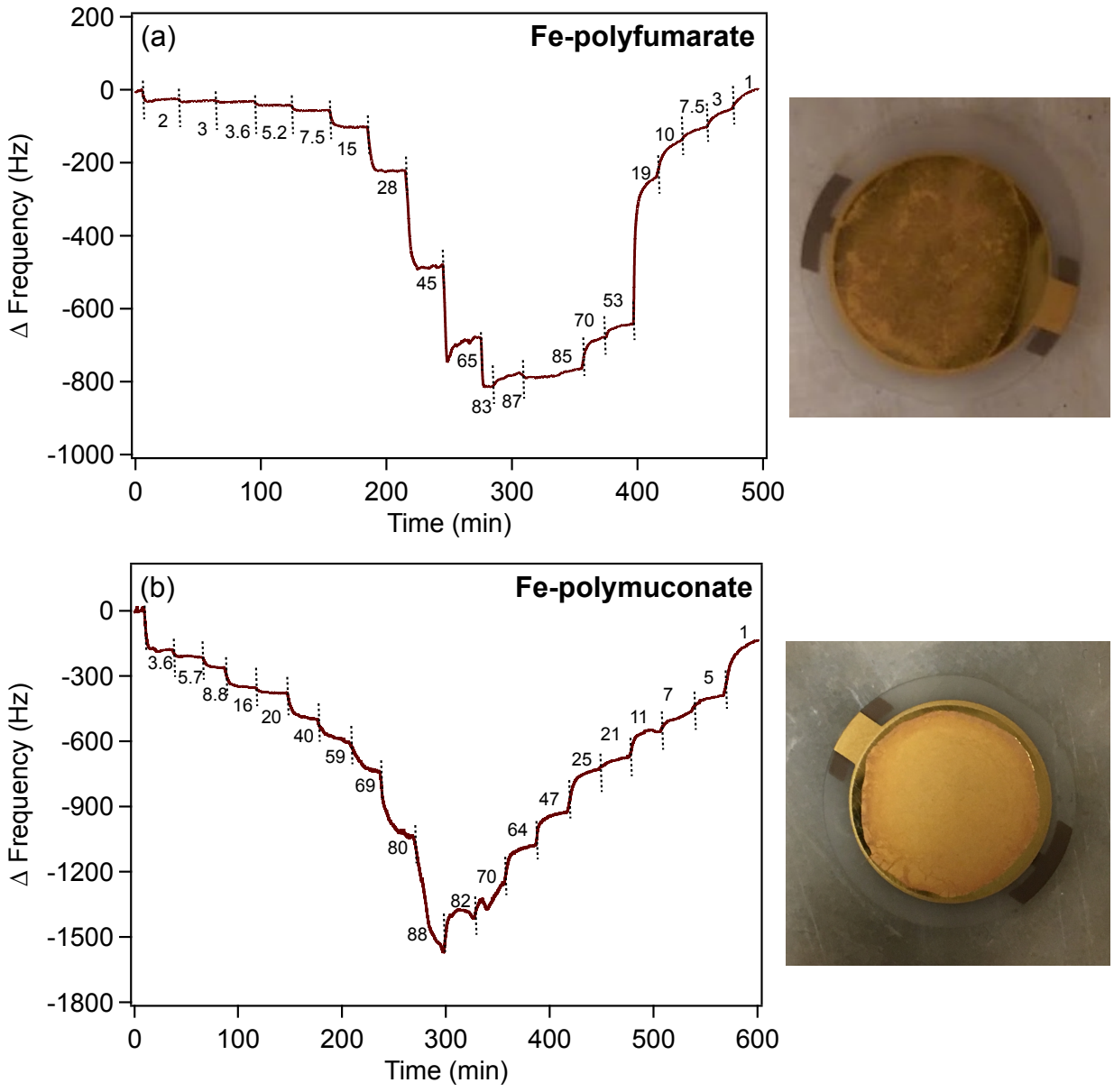


**Figure S4:** Raw QCM data showing change in frequency, $\Delta F$, as a function of increasing relative humidity (%RH, numbers between vertical dashed lines) relative to signal recorded while flowing dry air on (a) Fe-polyfumarate (0.037 mg), and (b) Fe-polymuconate (0.11 mg) thin films. Photos to the right show representative samples prior to starting the water uptake experiments.

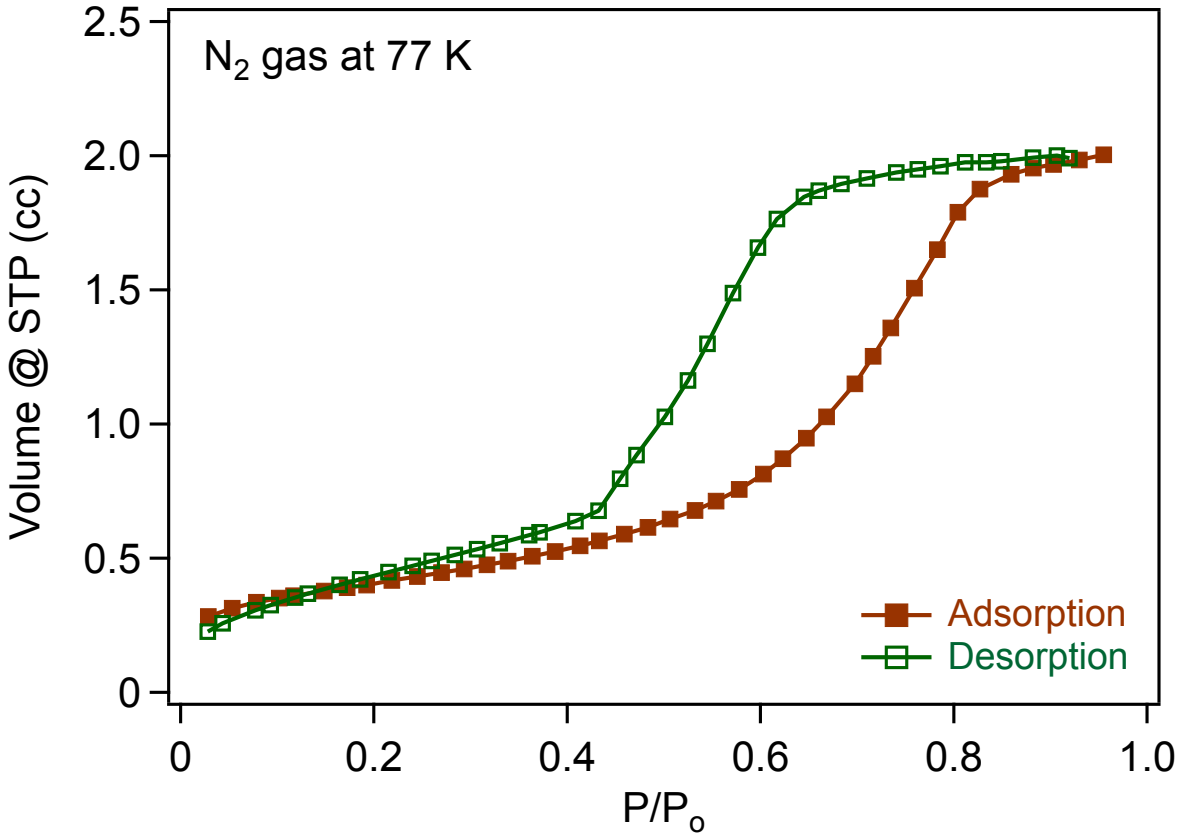


**Figure S5:** Adsorption isotherm of $N_2$ gas on Fe-polyfumarate at 77 K confirming the porous structure of this organometallic material.

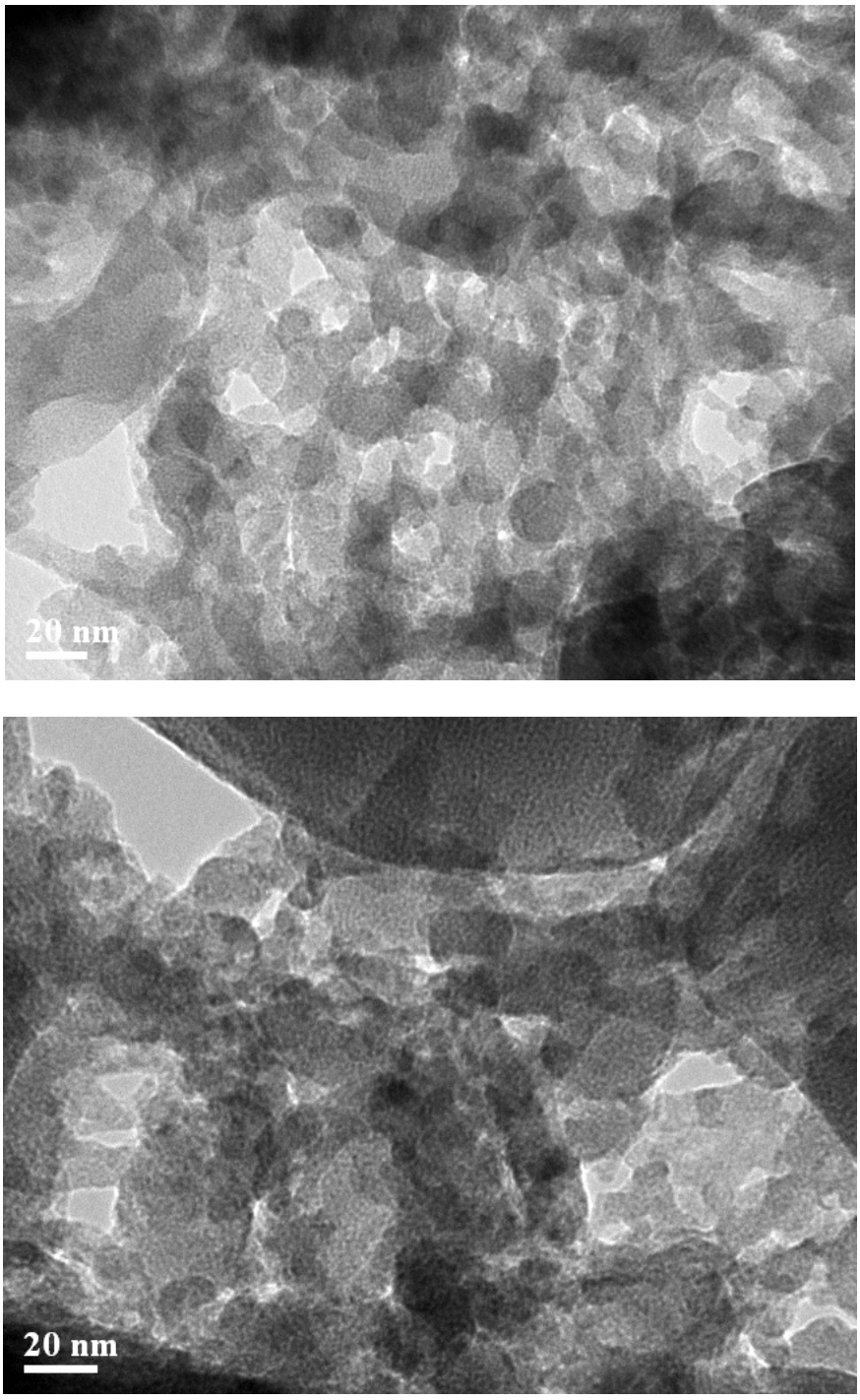


**Figure S6:** High resolution transmission electron microscopy (HR-TEM) images for Fe-polyfumarate showing the pore structure of this material.