## **Supporting Information**

## On the correlation between gate dielectric, film growth, and charge transport in organic thin film transistors: the case of vacuum-sublimed tetracene thin films

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**Figure S1.** Surface inclination for 3  $\mu$ m × 3  $\mu$ m regions of 35 nm thick tetracene films on polystyrene (PS), parylene C (PARY C), polymethylmethacrylate (PMMA), hexamethyldisilanzane (HMDS) and untreated SiO<sub>2</sub>. Please note the different color scales. The plots were obtained by taking the first derivative of the corresponding Atomic Force Microscopy height images. Tetracene films on HMDS have the flattest island surface followed by PS, while the surface of tetracene islands is the most curved on PARY C.



**Figure S2.** Fluorescence Microscopy images of 17 nm thick tetracene films deposited on the different dielectric layers. The images were obtained on a Zeiss Axio Imager.M1 fluorescence microscope. The insets show the corresponding Atomic Force Microscopy (AFM) images. While tetracene films on PS, PARY C, and PMMA have reached full substrate coverage at 17 nm nominal thickness, the black regions on the fluorescence images for tetracene on HMDS and SiO<sub>2</sub> reveal the existence of voids down to the substrate. We associate the bright spots on the fluorescence images to the higher islands, visible on the AFM images.



**Figure S3.** Reconstructed pole figures for the (001) crystallographic plane of 50 nm thick tetracene films deposited on different dielectric layers. The reconstruction is based on the Orientation Distribution Function calculated in the extended Rietveld refinement. The center of the pole figure corresponds to the sample normal. Tetracene grains on PS, HMDS, and SiO<sub>2</sub> are aligned along the sample normal. In contrast, most grains on PMMA and PARY C are aligned under a certain angle to the sample normal. Tetracene films on PMMA and PARY C thus have an inclined texture.



**Figure S4**. Transfer characteristics at saturation for tetracene OTFTs with PMMA and HMDS ( $V_{DS} = -60$  V). The linear fit of  $\sqrt{|I_{DS}|}$  vs  $V_{GS}$  is used to extract the hole mobility and the threshold voltage. OTFT geometry: W = 4 mm and  $L = 100 \mu$ m.



**Figure S5.** Output characteristics (drain-source current,  $I_{DS}$ , versus drain-source voltage,  $V_{DS}$ ) at different gate-source voltages,  $V_{GS}$ , for tetracene OTFT with PMMA and HMDS. Channel length L=100 µm and channel width W = 4 mm.