

# The adsorption of benzo[*k*]fluoranthene on self assembled films of octadecyltrichlorosilane

Gillian L. G. Goring, R. Stephen Brown and J. Hugh Horton\*

Department of Chemistry, Queen's University, Kingston, Ontario, Canada K7L 3N6.

Tel: +01-(613)-533-2379; Fax: +01-(613)-533-6669; E-mail: hortonj@chem.queensu.ca

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Organosilane compounds, such as octadecyltrichlorosilane (OTS), are commonly used in the formation of thin films for microelectronic, biosensor or chromatographic applications. In this paper, self-assembled OTS monolayer films were characterized by atomic force microscope (AFM) imaging and Fourier transform infrared-attenuated total reflectance (FTIR-ATR) spectroscopy. Conditions to provide a monolayer free of OTS aggregates were established. These films were then exposed to aqueous solutions of benzo[*k*]fluoranthene (B[*k*]F), a carcinogenic polycyclic aromatic hydrocarbon (PAH) that is often present in both aqueous and terrestrial environments. Through the B[*k*]F associations with these OTS monolayer films, it was possible to obtain a film-solution partition coefficient ( $\log K_{fs}$ ) value of  $7.0 \pm 0.1$  using fluorescence detection; this value is similar to partition coefficients previously observed in octanol-water systems. Using AFM images and fluorescence spectroscopy measurements, a maximum surface coverage of  $9 (\pm 1) \times 10^{-4}$  molecules of B[*k*]F per  $\text{\AA}^2$  was measured within these films. An upper limit to B[*k*]F concentration in solution was found, above which aggregates of B[*k*]F molecules were formed on the surface.

## Introduction

Thin organic films are commonly used in many research applications including chemical sensors,<sup>1,2</sup> microelectronics,<sup>3-6</sup> and chromatography.<sup>7</sup> These films often contain silane compounds or their functionalized derivatives,<sup>8</sup> with the most predominantly studied silane in the literature being octadecyltrichlorosilane (OTS).<sup>9,10</sup> Although there are many methods by which organic thin films can be deposited including thermal deposition,<sup>11,12</sup> sputtering techniques,<sup>13</sup> chemical vapour deposition,<sup>3,14</sup> and Langmuir-Blodgett techniques,<sup>15,16</sup> the self-assembly (SA) deposition process<sup>17,18</sup> is most often used for OTS. In the SA method used here, one to several layers of OTS self-assemble from an organic solution onto a substrate base.<sup>18</sup> The formation of these self-assembled monolayers (SAM) occurs through a combination of both physisorption and chemisorption processes.<sup>19</sup> OTS films can also be used as a base to which other films<sup>20</sup> or molecules<sup>21,22</sup> can be successfully attached or adsorbed. They may thus be used in various chemical sensor or chromatographic applications. Applications of this inclusion property of OTS films can be extended to the association of these films with environmentally hazardous compounds.

One of the main difficulties in preparing SAM is determining the quality of the monolayer formed. Many reports assume that a monolayer is formed based on the method used and previous reports, without sufficient characterization. This may be even after significant changes from previous methods such as the use of different substrate materials or substrate preparation methods. These changes can lead to differences in deposited film morphology from sub-monolayer to multilayer coverage. Important film properties including thickness, fluidity and homogeneity are all critically dependent on film morphology, thus affecting the intended function of the film as well. To properly assess SAM film formation, it is necessary first to demonstrate that a true, reproducible monolayer is present.

One contemporary use of SAM is to provide interactions with insoluble organic molecules in water. Films such as the

octadecyl (C-18) hydrocarbon chains have been used for everything from solid-phase extraction of compounds from water to partition controlled delivery of materials to a solution. An important class of insoluble compounds is the polycyclic aromatic hydrocarbons (PAH), which are present widely in soil, sediment and aqueous environments and are considered to be toxic to plants and animals in high concentrations. Certain PAH species, such as benzo[*k*]fluoranthene (B[*k*]F), are known to be carcinogenic to animals and humans. The degradation reactions and fates of PAH molecules, which depend on the presence of other chemicals, the surrounding environment and temperature, require significant analysis.<sup>23</sup>

Most of the more toxic PAH compounds have been studied in octanol-water mixtures to determine the partition coefficient,  $K_{OW}$ , of a given species between the water and the organic phase.<sup>24</sup> The numerical value of  $K_{OW}$  is influenced by the hydrophobic effect, with larger values indicating that the PAH molecules preferentially absorb or partition into the organic phase from the aqueous phase.<sup>24</sup> C-18 films are similar to the organic phase, in that the same hydrophobic effect can drive a PAH molecule to partition out of an aqueous solution and into the film. These associations can be labeled as either absorption processes, where the PAH molecule is physically included into the film, or as adsorption processes, where the PAH molecule will adsorb to either the surface of the film or to other surfaces within the exposure system.<sup>19,25</sup>

In order to create a surface with optimal interactions with PAH, such as B[*k*]F, SA deposition techniques may first be used to generate a C-18 monolayer film on a suitable substrate. The deposition in this work used octadecyltrichlorosilane (OTS) as a source of C-18 film, as has been extensively reviewed in the literature.<sup>3,7-10,26-28</sup> Here, we have used mica, pre-exposed to HCl, as the substrate. The exposure to HCl exchanges  $K^+$  for  $H^+$  on the mica surface, forming nucleation sites for the hydrolysis of the OTS molecules, and their subsequent cross-linking on the surface. Mica is an ideal candidate because it can be easily cleaved to reveal an atomically smooth surface to which monolayer films can be deposited.<sup>9</sup> This smoother surface permits a much more

straightforward interpretation of surface images using atomic force microscopy (AFM).

The object of this study is to examine the adsorption of a PAH, B[k]F, on OTS films in order to better understand the processes which would take place in such a film during chromatographic or sensor applications. The OTS–mica system does not perhaps first appear to be the ideal one as OTS is a notoriously difficult molecule to use for monolayer coatings. However, our purpose here is to examine the behaviour of PAH's in films that are used in chemical sensor applications, and it is the OTS films that most closely mimic this application. One important objective is determining the partition coefficient value for a given PAH compound between the OTS film and the surrounding aqueous solution. In order to study the inclusion processes of B[k]F, the system is examined by a combination of FTIR-ATR spectroscopy, AFM imaging to determine the morphology of the OTS films, and fluorescence spectroscopy in order to quantitatively determine the amount of B[k]F present.

## Experimental

### Sample preparation

Grade 5 research quality muscovite mica squares (SPI supplies, West Chester, Pennsylvania) were cut into rectangles (1.4 cm × 1.8 cm). Both sides of the samples were cleaved in order to provide a clean atomically smooth surface. Prior to use, these samples were then mounted vertically in a glass holder, to allow for uniform exposure of the 0.50 M HCl solution (reagent grade: Fisher Scientific, Mississauga, ON) to both surfaces of the mica sample slides. After a three-hour exposure to the HCl solution, the slides were removed, dried at 0.1 Torr N<sub>2</sub> (high purity grade: Air Products, Kingston, ON) in a desiccator for 30 minutes and stored in a N<sub>2</sub> atmosphere.<sup>27</sup> An alkylating solution of 3 mM OTS (95%: Aldrich Chemical Co., Milwaukee, WI) in cyclohexane (certified ACS spectranalyzed: Fisher Scientific) was prepared and placed in an ultrasonic bath for two minutes, where it was thoroughly mixed. The activated mica slides were then immersed in the silane solution, again in a vertical position in a glass holder for five minutes. The holder had previously been exposed to a concentrated solution of OTS in order to passivate the surface to further alkylation. The slides were then removed from the alkylating solution, liquid droplets located on the corner of the slides were drained with a careful touch of a Kimwipe, and the slides were cured in a 120 °C oven for two hours. When the samples were removed from the oven, they were allowed to cool to room temperature prior to being stored in an airtight container to prevent the adsorption of water molecules on their surfaces. FTIR-ATR spectroscopy confirmed the presence of the silane molecules on the mica slides and AFM imaging characterized the film thickness and distribution across the surface.

Some of these OTS-coated mica slides were then submerged in aqueous solutions of B[k]F of varying concentrations for different lengths of time. The slides were mounted vertically, this time in a stainless steel holder, which permitted the solution to have uniform access to both sides of the mica slides, while a stirring bar ensured an even distribution of the B[k]F molecules throughout the solution. The amount of B[k]F associated with these films was determined by either solid phase fluorescence spectrometry of the solid sample when removed from the aqueous B[k]F solution, or by extraction of the slide in a glass vial with ethanol (HPLC grade: Fisher Scientific) followed by a solution fluorescence measurement.

### Instrumental analysis

The OTS monolayer films that were deposited on the mica slides were analyzed by FTIR-ATR spectroscopy and by AFM

imaging. The FTIR-ATR consisted of a TlBr ATR crystal (Spectra Tech: Shelton, CT) and Bomen IR Spectrometer (MB-Series, Hartman & Baun: QC). The AFM instrument was a scanning probe microscope (Molecular Imaging: Phoenix, AZ) with Nanoscope E software (version 4.22; Digital Instruments: Santa Barbara, CA). The majority of the images were obtained in contact mode using standard Si<sub>3</sub>N<sub>4</sub> tips with the tip immersed in a droplet of distilled water. The fluorescence results were obtained from a QuantaMaster QM1 fluorescence spectrometer (Photon Technology International: London, ON). The excitation light source was a 75 W Xenon arc lamp. The spectrometer was equipped with Czerney–Turner excitation and emission monochromators with slits set to a 2 nm band pass for all of the measurements.

## Results and discussion

### Characterization of the mica substrate and the OTS films

After the mica substrate had been cleaved, AFM images were obtained to confirm that the surface was atomically flat. These images were featureless; the cross-sectional height measurements did not vary by more than 3 Å. Identical images were obtained from mica substrates that had been exposed for three hours to dilute 0.5 M HCl solution.

FTIR-ATR spectra were obtained from the bare mica and the OTS-coated mica slides over the spectral range of 400 cm<sup>-1</sup> to 4000 cm<sup>-1</sup>. The bare mica substrate did not exhibit any absorption peaks above 1400 cm<sup>-1</sup>. However, when the OTS-coated mica samples were analyzed, two peaks appeared at 2850 cm<sup>-1</sup> and 2918 cm<sup>-1</sup> and they are assigned respectively as the CH<sub>2</sub> symmetric and asymmetric stretches of the OTS alkyl chain.<sup>26,29,30</sup> A sample FTIR-ATR spectrum of a five-minute OTS deposition sample is shown in Fig. 1. Occasionally, due to poor sample preparation technique, polymerised OTS layers were formed containing very large (up to 1 µm diameter) roughly spherical amorphous particles, as indicated by AFM.

To ensure that AFM could confirm formation of monolayers, including measuring film thicknesses, it was necessary to prepare OTS films which did not completely cover the mica substrate. Systematically varying both the concentration of the OTS solution in cyclohexane and exposure time indicated that a five-minute exposure to a 3 mM OTS solution resulted in the best partial monolayer films in terms of images acquired by the AFM. A typical AFM image of a five-minute sample is seen in Fig. 2. The images reveal that the islands were dendritic in shape, similar to previous observations on this system.<sup>7</sup> From a cross section plot of the islands, the thickness was found to vary in height between 20 Å and 56 Å, indicating a film between one and two monolayers thick. Bearing analysis indicated that the OTS film covered approximately 50% of the mica substrate. At longer exposure times or higher concentrations than 3 mM, complete coverage of the mica substrate occurred along with

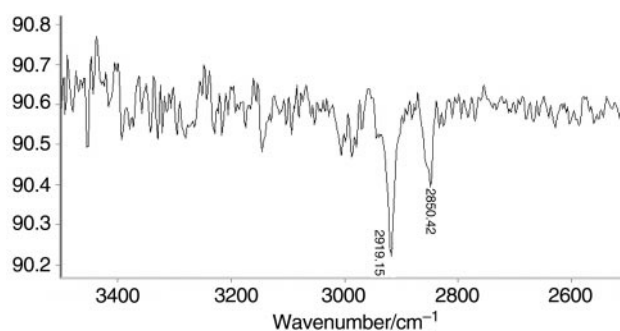
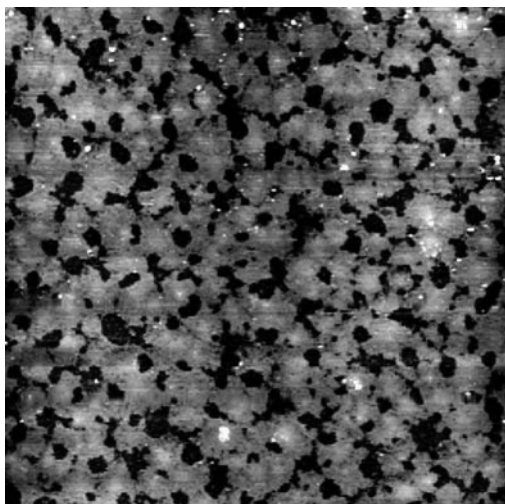
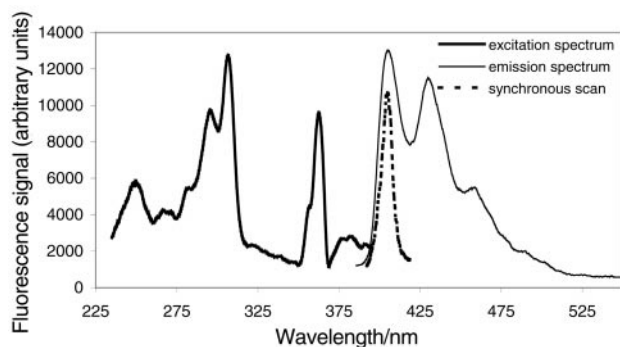


Fig. 1 FTIR-ATR spectrum of OTS films obtained from five minutes exposure of mica to a 3 mM solution of OTS in cyclohexane.



**Fig. 2** Typical contact-mode AFM image in water of an OTS film obtained from five minutes exposure of mica to a 3 mM solution of OTS in cyclohexane. Scan area 10  $\mu\text{m}$  square,  $z$  range 5.0 nm.



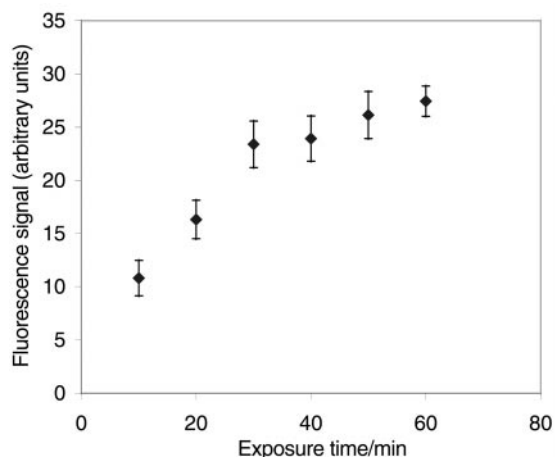
**Fig. 3** Fluorescence spectra of B[k]F obtained in aqueous solution.

the formation of secondary islands on top of the initial layer; these secondary islands were of uneven thickness.

#### Determination of $K_{FS}$

The OTS-coated slide was immersed in aqueous solutions of B[k]F of concentration between  $1 \times 10^{-9}$  M and  $5 \times 10^{-8}$  M. An example of B[k]F excitation, emission and synchronous fluorescence scanning (SFS) spectra is shown in Fig. 3. Initially, the B[k]F SFS spectra were acquired directly from the film. An equilibration time for the exposure of a partially OTS-coated slide to a  $2.5 \times 10^{-9}$  M B[k]F solution was found to be between 30 and 40 minutes (see Fig. 4). A 40 minute exposure time was thus used for subsequent samples to ensure equilibrium had been reached.

The film-solution partition coefficient,  $K_{FS}$ , is defined as the ratio of the concentration of B[k]F in the film to the concentration of B[k]F in solution at equilibrium ( $K_{FS} = [\text{B}[k]\text{F}]_{\text{film}}/[\text{B}[k]\text{F}]_{\text{aq}}$ ). For sparingly soluble compounds,  $K_{FS}$  must be determined by directly measuring concentrations in solution and in the film after equilibrium is established. This is because a significant portion of the total compound added may be lost by other processes such as adsorption to vessel walls or other solid surfaces, so initial concentrations and concentration differences cannot be used reliably. In this work, the amount of B[k]F in water was determined directly using fluorescence spectrometry. The amount associated with the film, film substrate, glass vessel, holder apparatus and other objects or surfaces was determined by rinsing each object with ethanol and diluting to a known ethanol volume. Multiple rinses were



**Fig. 4** B[k]F fluorescence signal as a function of exposure time for an OTS-coated mica slide exposed to  $2.5 \times 10^{-9}$  M aqueous B[k]F solution.

**Table 1** Percentage of B[k]F molecules desorbed from individual components

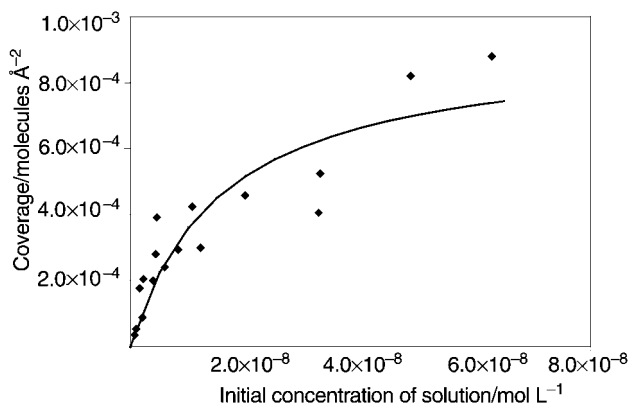
Initial concentration ( $\times 10^{-9}$ M)	0.87	5.4	33
Percentage remaining in the solution after 40 minutes	$17 \pm 3$	$42 \pm 7$	$69 \pm 11$
Percentage in slide extract	$33 \pm 3$	$16 \pm 3$	$9.7 \pm 0.9$
Percentage in sample holder extract	$35 \pm 4$	$23 \pm 2$	$11 \pm 1$
Percentage in stirring bar extract	$12.6 \pm 0.4$	$7.5 \pm 0.3$	$4.7 \pm 0.2$
Percentage in vial rinse extract	$1.58 \pm 0.06$	$3.4 \pm 0.1$	$4.6 \pm 0.2$
Recovery	99	92	99

done to ensure complete removal of the B[k]F. The proportion of B[k]F detected in the various compartments is shown in Table 1. The mass balance indicated that all B[k]F was accounted for, providing greater confidence in the water and film concentration values.

A further complication arises from the fact that it is possible that B[k]F might adsorb onto bare regions of the mica substrate (which as seen in Fig. 2, comprise about 50% of a typical sample surface). In order to calculate the concentration of B[k]F in the OTS film itself, it is necessary to know the dimensions of the film. For each experiment, two OTS films were produced simultaneously to ensure a similar morphology. One underwent exposure to B[k]F. The second sample was subjected to analysis by AFM in order to determine the film thickness and extent of surface coverage, as described above. Average values were determined for three points on the film. Bare HCl-treated mica slides exposed to B[k]F solution were found to adsorb B[k]F at 23% efficiency compared to the adsorption on a slide completely coated with OTS. The final  $K_{FS}$  values reported take this correction into account. Over a B[k]F concentration range between  $1 \times 10^{-9}$  M and  $5 \times 10^{-8}$  M in the aqueous solution, it was found that the average value of  $K_{FS} = 9.1 (\pm 1.5) \times 10^6$ . The log of the partition coefficient for B[k]F was thus 7.0. The  $K_{FS}$  values obtained are presented in Table 2, along with a comparison of literature B[k]F  $K_{OW}$  values.<sup>24,31</sup> The value obtained is slightly higher, suggesting that the OTS films have a better affinity for B[k]F compared with octanol solvent. It should be pointed out that while this

**Table 2** Comparison of experimental and literature  $K_{FS}$  values

Source of $K_{FS}$ value	Log $K_{FS}$ or Log $K_{OW}$ values
Experimental value	Log $K_{FS} = 7.0$ , range: 6.9–7.0
de Maagd <i>et al.</i> <sup>24</sup>	Log $K_{OW} = 6.11$ , range: 5.86–6.28
ADC Labs, Toronto <sup>31</sup>	Log $K_{OW} = 7.0$ , range: 6.11–6.86



**Fig. 5** Plot of B[k]F surface coverage on the OTS-coated mica as a function of B[k]F concentration in solution. While the data are scattered at high concentrations (see text) the data suggest a limiting coverage of  $9 \times 10^{-4}$  molecules/Å<sup>2</sup>.

method gave fairly precise results, an error in estimated parameters such as film thickness or surface coverage would introduce a systematic error in the  $K_{FS}$  value determined, which could also contribute to the difference from  $K_{OW}$ .

#### Mechanistic aspects of B[k]F deposition on OTS

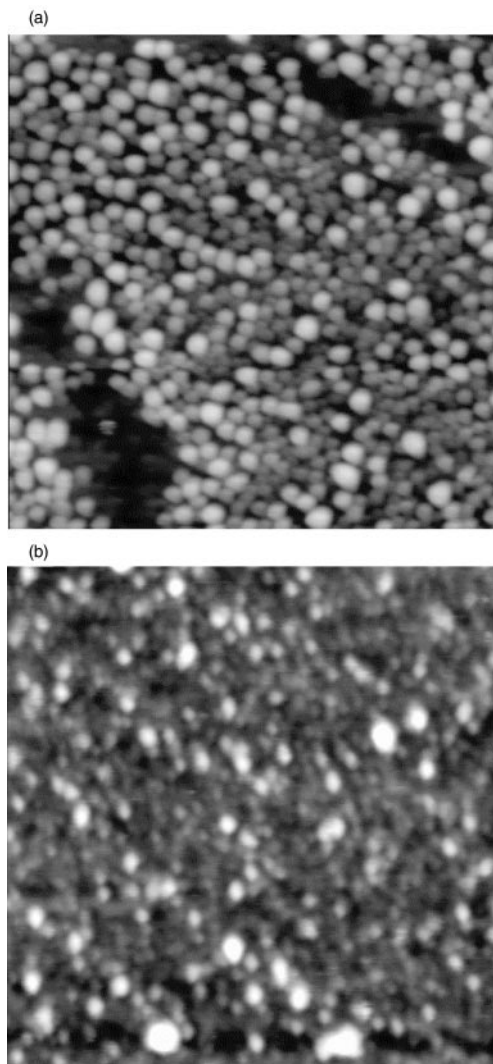
Again by analyzing identical pairs of samples exposed to B[k]F by both AFM and fluorescence spectroscopy, B[k]F adsorption onto the OTS films may be examined as a function of the initial B[k]F concentration in the aqueous solution (see Fig. 5). While the data are fairly scattered at high concentrations, it is evident that the curve tends towards a maximum B[k]F coverage of approximately  $9 \times 10^{-4}$  molecules Å<sup>-2</sup> of film. The scattering at high concentrations is probably related to the fact that the concentration of B[k]F in the aqueous solution was near its saturation value. As demonstrated below, this can lead to the nucleation of B[k]F crystallites on the sample.

Given that the OTS density within a monolayer<sup>3</sup> is approximately  $8 \times 10^{-2}$  molecules Å<sup>-2</sup> this limiting value corresponds to a concentration of approximately 1% B[k]F molecules within the OTS film at saturation.

While in general no change was seen in images of OTS films exposed to B[k]F, under certain conditions considerable changes in film morphology could be observed. This always occurred on slides exposed for longer periods of time (> 60 minutes) to aqueous B[k]F solutions whose concentrations were near the aqueous solubility limit of this PAH. Published values for the aqueous solubility of B[k]F range between  $5.5 \times 10^{-8}$  M and  $4.3 \times 10^{-9}$  M.<sup>32</sup> Fig. 6 shows two examples of films exposed for 3 and 6 hours to a solution of concentration  $5 \times 10^{-8}$  M. In these experiments, the OTS coated mica slides were allowed to lie on the bottom of the container without the presence of the stainless steel holder or the stirring bar. Fig. 6 shows the nucleation of small spherical crystallites on the surface. These grew in number as the exposure time was increased. These features were confirmed as B[k]F deposits by solid sample fluorescence spectroscopy. Quantitative analysis of concentrations could not be performed, as the fluorescence signal was not highly reproducible from run to run, even on the same sample, probably due to light scattering by the crystallites. Thus, direct comparison to the solution phase fluorescence data is not possible.

#### Conclusions

Exposure of HCl-treated mica slides to 3 mM solutions of OTS in cyclohexane for five minutes resulted in suitable deposition conditions for an OTS monolayer film covering approximately half the mica substrate surface area. Cross-sectional analysis of



**Fig. 6** Contact mode AFM images of OTS films exposed to an aqueous solution of B[k]F of  $5.0 \times 10^{-8}$  M (a) after 6 hours exposure, scan area  $2 \mu\text{m}$  square,  $z$  range 20 nm (b) after 3 hours exposure, scan area  $2 \mu\text{m}$  square,  $z$  range 10 nm.

these same AFM images revealed that these films were indeed one or two OTS molecules in height. Forty minute exposure times of these films to aqueous B[k]F solutions allowed the PAH molecules to equilibrate between the solution and the film. A film solution partition coefficient value for B[k]F,  $\log K_{FS}$ , of 7.0 was obtained for these partial monolayer films. Within experimental error, this is consistent with previously published values for the octanol–water partition coefficient,  $K_{OW}$ . AFM images of slides that were exposed for longer periods of time to B[k]F concentrations approaching the aqueous solubility limit of B[k]F indicated the presence of microcrystals. The maximum number of B[k]F molecules that could be absorbed on the partial OTS film was  $9 \times 10^{-4}$  molecules Å<sup>-2</sup>. Since this value corresponds to a 1% concentration of B[k]F within the OTS film, it is difficult to observe any changes in morphology using AFM. Further research with other silane films or different PAH molecules may provide more information for a database for use in the development of solid phase extraction experiments and biosensors to detect these PAH compounds in aquatic or terrestrial environments.

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