Supplementary Information

‘Simple Rules and the Emergence of Complexity in surface Chirality’

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Figure S1 Surface chirality generators for the (i) homochiral ($C_2 \times E$) and (ii) heterochiral ($S_2 \times E$) footprint templates within a $(3 \times 2)$ packing. The chiral outputs arising from the enantiopure and racemic adsorption of both single-footed and two-footed molecules are derived for each template and are labelled in terms of their overall surface chirality, $C_H$.
Figure S2 Surface chirality generators for the (i) homochiral (E × E) and (ii) heterochiral (S₂ × E) footprint templates within a (4 × 2) packing. The chiral outputs arising from the enantiopure and racemic adsorption of both single-footed and two-footed molecules are derived for each template and are labelled in terms of their overall surface chirality, $C^H_F$. 
Figure S3 Surface chirality generators for the (i) heterochiral ($\sigma_v \times S_2$) and (ii) heterochiral ($C_2 \times S_2$) footprint templates within a (4 × 2) packing. The chiral outputs arising from the enantiopure and racemic adsorption of both single-footed and two-footed molecules are derived for each template and are labelled in terms of their overall surface chirality, $C_P$. 
Figure S4 Surface chirality generators for the (i) heterochiral \((E \times S_2)\) and (ii) heterochiral \((S_2 \times S_2)\) footprint templates within a \((4 \times 2)\) packing. The chiral outputs arising from the enantiopure and racemic adsorption of both single-footed and two-footed molecules are derived for each template and are labelled in terms of their overall surface chirality, \(C_H^\gamma\).
**Figure S5** Surface chirality generators for (i) homochiral and (ii) heterochiral randomized footprint templates within a (4 × 2) packing. The chiral outputs arising from the enantiopure and racemic adsorption of both single-footed and two-footed molecules are derived for each template and are labelled in terms of their overall surface chirality, $C_F^H$. 
Figure S6. Examples of chiral surfaces that may arise from either a statistically rare event or tailored enantiospecific interactions within (i) a homochiral footprint template and (ii) a heterochiral footprint template. (i) For racemic adsorption of a two-footed molecule in a homochiral template, we would expect a random arrangement of enantiomers if (S) and (R) can occupy λ and δ footprints equally, leading to a surface described by \( C_{Rn}^{Cn} \) (see surface chirality generators above). However, consider an event in which, by chance, enantiomers order as a racemic compound. The resulting surface would be a conglomerate in terms of the footprints but a racemic compound at the handedness level, \( C_{Rc}^{Cn} \). This outcome is not disallowed by the rules but could only arise from a statistically rare event. Alternatively, enantiospecific interactions could be tailored such that heterochiral enantiomer interactions (R)-(S) are favoured over the homochiral interactions. In this instance, the chiral surface described by \( C_{Rc}^{Cn} \) could arise. (ii) Similarly, for racemic adsorption of a two-footed molecule in a heterochiral footprint template we would expect a random arrangement of enantiomers, \( C_{Rn}^{Rc} \) (see surface chirality generators above). However, a statistically rare event could also lead to a conglomerate at the handed level, \( C_{Rc}^{Cn} \) should enantiomers organize in separate domains. Again, this is a statistically unlikely event, but it is not forbidden by the rules. Alternatively, a conglomerate within the heterochiral footprint template, \( C_{Rc}^{Cn} \) could arise if interactions between homochiral enantiomer pairs (S)-(S) and (R)-(R) are preferred over heterochiral ones (R)-(S).