Thermoelectric detection and imaging of propagating graphene plasmons

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propagating plasmons in graphene has not yet 62 mid-infrared plasmon detector, where a single graphene sheet serves simultaneously as the plasmonic medium and detector. Rather than achieving detection via added optoelectronic materials, as is typically done in other plasmonic systems, 8-15 our device converts the natural decay product of the plasmon-electronic heatdirectly into a voltage through the thermoelectric effect. 16,17 We employ two local gates to fully tune the thermoelectric and plasmonic behaviour of the graphene. High-resolution real-space photocurrent maps are used to investigate the plasmon propagation and interference, decay, thermal diffusion, and thermoelectric generation.

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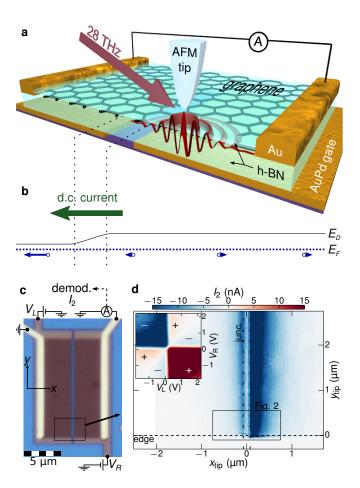
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Graphene plasmonics is an emerging platform for ter-36 ahertz to infrared nano-optics, attractive due to the $_{37}$ long intrinsic lifetime of > 0.5 ps and the strong tun-38 able broadband electrodynamic response of its Dirac electrons.^{6,18} Typically, graphene plasmons are sensed by out-coupling to light, which is inefficient due to one of the 41 key features of graphene plasmons: their extremely short wavelength ($\sim \frac{1}{100}$ that of free space light). While plasmon resonances have been exploited to enhance absorption and thereby enhance far-field photodetection, ^{19,20} the concept of an on-chip plasmon receiver has not yet been realized.

The presented experimental device is built around the 48 state-of-the-art plasmonic medium of graphene encapsulated in hexagonal boron nitride (hBN), which we have 50 recently demonstrated to support high quality propagat-₅₁ ing plasmons in the mid infrared.²¹ As a key innovation over previous plasmon studies, the induction of free

Controlling, detecting and generating prop- 56 ditional SiO₂ layer. The use of local gating allows to agating plasmons by all-electrical means is at 57 spatially modulate the charge carrier density and polarthe heart of on-chip nano-optical processing. 1-3 58 ity across the device, as well as providing lower voltage Graphene carries long-lived plasmons that are 59 operation and reduced charge trapping effects. 22 As we 17 extremely confined and controllable by electro- 60 will show below, the junction induced by the two gates 18 static fields, 4-7 however electrical detection of 61 can be used as a thermoelectric detector for the plasmons. Figures 1a,b show a schematic of the operating princibeen realized. Here, we present an all-graphene 63 ple of the detector. In lieu of an on-chip plasmon source, 64 we generate plasmons using the conventional scattering 65 scanning near field microscopy (s-SNOM) technique. ^{23,24} 66 The s-SNOM apparatus consists of a scanning metal 67 probe under illumination from a continuous wave laser 68 at mid infrared frequency. A laser frequency of 28 THz 69 (10.6 µm free space wavelength) was chosen to avoid com-70 plications from the hBN phonons. 21 In conventional plas-71 monic s-SNOM experiments, the signal of interest is the 72 out-scattered light, containing information about local 73 dielectric properties and plasmonic modes. Here, we in-74 stead measure a quantity I_2 , known as near field pho-75 tocurrent, from the current exiting the device electrodes ⁷⁶ (Fig. 1c). ²² This is the component of total measured cur-77 rent that oscillates at the second harmonic (~500 kHz) 78 of the probe tapping frequency (~ 250 kHz). As the 79 graphene shows a linear photocurrent response, I_2 can 80 be understood as the photocurrent arising only from the ~60 nm-sharp near fields of the tip, isolated from the 82 background photocurrent directly induced by the inci-83 dent light. For simplicity, in the remainder of this paper 84 we refer to I_2 simply as "the photocurrent" and treat it 85 as if it arises from an effective nanoscale light source.

The studied device and circuit schematic is shown in 87 Fig. 1c. By applying different voltages $V_{\rm L(R)}$ to the 88 left (right) gates, we induce a localized photosensitive 89 region, e.g., a p-n junction as studied in Fig. 1d. The 90 six-fold photocurrent pattern observed when both gates 91 are scanned (figure inset) is considered as evidence of a 92 thermoelectric generation mechanism, where the pattern 93 arises due to the nonmonotonic dependence of Seebeck ⁹⁴ coefficient on gate voltage. ^{25–28} For a simple junction, the ₉₅ thermoelectric current is $I = (S_{\rm R} - S_{\rm L}) \overline{\Delta T}^{\rm junc} / R$, where carriers in the graphene is achieved through the use of $_{96}$ $S_{\rm L(R)}$ is the left (right) Seebeck coefficient, $\overline{\Delta T}^{\rm junc}$ is the 54 separate local gates directly underneath the hBN, rather 97 junction-average rise in electronic temperature relative than the conventional global back gating through an ad- $_{98}$ to ambient, and R is the circuit resistance. The gate de-



Concept and device. a, Schematic cross-section of device and measurement technique. Continuous-wave laser light scatters at a movable metallized AFM tip, launching plasmons in the hBN-graphene-hBN heterostructure. **b**, Schematic of thermoelectric detection mechanism in a microscopic picture. The plasmon decay energy drives an outward majority carrier diffusion, in this case hole carriers. A gate-induced homojunction (seen as variation in the graphene Dirac point energy level $E_{\rm D}$ relative to the Fermi level $E_{\rm F}$) imbalances this diffusion, resulting in a nonzero net dc current. c, Optical micrograph of presented device and circuit diagram. Two metal electrodes (light yellow) contact an encapsulated graphene sheet (dark rectangle) which lies above a split metal gate layer (light brown). Split gate voltages $V_{\rm L}$ and $V_{\rm R}$ create the homojunction, while tip-induced currents near field photocurrent I_2 . **d**, Near-field photocurrent map of the entire device, showing the photosensitive junction created by applying different gate voltages. Inset: The sign of the photocurrent (measured with the tip over the junction) shows a six-fold pattern characteristic of thermoelectric effects.

99 pendence allows to identify the charge neutrality point 100 of the graphene in this device (occuring at a gate voltage offset of +0.09 V). Hereafter we use this offset and the 102 calculated gate capacitance to convert the gate voltages 122 ploy a simplified two-dimensional model that takes into $V_{L,R}$ into carrier densities $n_{L,R}$.

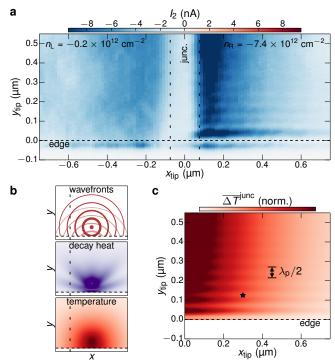


FIG. 2. Plasmon photocurrent spatial maps. a, A highresolution photocurrent map near the edge of the graphene, containing interference fringes. b, Modelled fields for a given $x_{\rm tip}$, $y_{\rm tip}$ position: wave propagation and interference (upper panel: strong red curves are launched wavefronts, faint red curves are reflected wavefronts), decay to heat (middle panel), and thermal spreading (bottom panel). Parameters used were $k_{\rm p}=(56+1.8i)~{\rm \mu m}^{-1},~r=0.4e^{0.65\pi i},~l_T=0.25\,{\rm \mu m}.$ **c**, The modelled average temperature rise along the junction (along the vertical dashed line in (b)), $\overline{\Delta T}^{\text{junc}}$, and its dependence on $x_{\rm tip}$, $y_{\rm tip}$. The \star symbol marks the case shown in panel **b** of this figure.

106 rier density (Fig. 2a), where interference fringes can be $_{107}$ observed in I_2 near the graphene edge. These fringes 108 can be unambiguously attributed to plasmon reflections, $_{109}$ as they match the half-wavelength periodicity seen in 110 the s-SNOM optical signal that is conventionally used 111 to characterize graphene plasmons. 21 The extracted plasare captured at the electrodes and demodulated to obtain the 112 mon wavelength of $\lambda_{\rm p}=112$ nm in this scan is close to the expected value of 114 nm, and consistent with a previous study of a similar hBN-graphene-hBN device.²¹

To explain the spatial I_2 pattern and the detection mechanism, we consider the following sequence, sketched 117 in Fig. 2b: Plasmons radiate away from the tip and re-118 flect at the edge; the self-interfered plasmon wave decays # into heat in the graphene electron gas; subsequent electronic diffusion spreads the heat to the junction, determining $\overline{\Delta T}^{\mathrm{junc}}$. To justify this interpretation, we em-# account each of these effects. The model, summarized Strong evidence of plasmons mediating the photocur104 Strong evidence of plasmons mediating the photocur105 rent is visible in photocurrent maps obtained at high car125 yields the value of $\overline{\Delta T}^{\text{junc}}$ (up to a normalization) for a

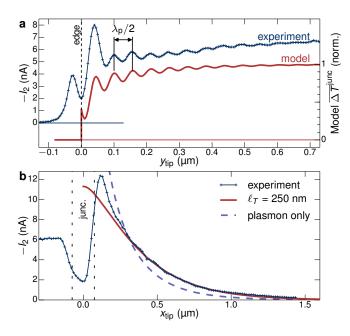


FIG. 3. Lineauts along x_{tip} and y_{tip} . a, Lineaut of photocurrent map perpendicular to the edge, obtained by averaging the data in Fig. 2a over the interval $x_{\rm tip} = 0.2 \cdots 0.4 \, \mu \text{m}$. The lower curve (right axis) shows the corresponding model linecut (from Fig. 2c). b, Linecut of photocurrent across the junction, far from the edge, obtained by averaging Fig. 2a over the interval $y_{\rm tip} = 0.6 \cdots 0.7 \, \mu \text{m}$. The solid red curve 177 from $\sim 500 \, \text{nm} \, (\text{low} \, |n|)$ to $\sim 250 \, \text{nm} \, (\text{high} \, |n|)$ is needed. shows the corresponding model linecut (from Fig. 2c), and the dashed curve shows the model result considering only plasmon propagation (without thermal diffusion); model curves have been vertically scaled for comparison with the data.

126 plasmon source located at any position $x_{\rm tip}$, $y_{\rm tip}$. The 184 tocurrent, due to the sign change of Seebeck coefficient three critical model parameters are plasmon wavevector the figure appears to follow less $k_{\rm p}$, reflection coefficient r, and electron cooling length $l_{\rm 186}$ $\frac{1}{2}\lambda_{\rm p} \propto |n|^{1/2}$ as expected for graphene plasmons. Most less $l_{\rm T}$. By varying these, we obtain a map (Fig. 2c) that fits less three critical model parameters are plasmon wavevector less differences. The fringe spacing appears to follow less $k_{\rm p}$, reflection coefficient r, and electron cooling length less $l_{\rm 187}$ strikingly, the photocurrent shows two regimes of strong 130 to the data, capturing the essential physics behind the 188 magnitude, at high |n| and low |n|, separated by a region 131 observed spatial pattern. Note that this model neglects 189 of weak photocurrent from $|n| \sim 1-4 \times 10^{12}$ cm⁻². We junction.

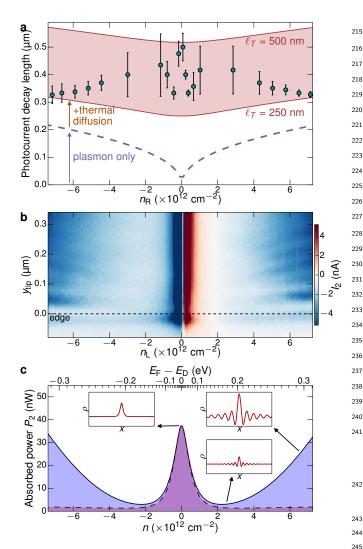
138 ing, Im[k_D] encodes the plasmon decay length and de-196 to variation in plasmon wavelength: plasmons with small 140 lar, the fringes decay according to an envelope function 198 confinement in the top hBN layer and the limited range $\exp(-y_{\rm tip}\,{\rm Im}[k_{\rm p}])/\sqrt{y_{\rm tip}}$, identical to that of the optical 199 of spatial frequencies probed by the round tip. Direct signal. The reflection coefficient r is relevant for setting beating on the other hand is strongest for low |n| due to and arg(r) respectively. The subunity value of |r|=0.4 202 point $E_{\rm D}$ is within about $\hbar\omega/2~(=58~{\rm meV})$ of the Fermi also leads to a drop in power as the tip is brought near 203 level $E_{
m F}$. 146 the edge, since in this model the unreflected plasmon $\#_{04}$ power is lost. A similar drop is seen in the data, suggest- 205 our detector. According to an estimation based on 148 ing that the unreflected plasmon power is not converted 200 the known thermoelectric properties of our device (see 149 to electronic heat in the same way as for plasmon decay 207 Supplementary Discussion 2), the junction responsivity 150 elsewhere.

The electron cooling length, l_T , is important for match-152 ing the photocurrent decay away from the junction, shown in Fig. 3b. This l_T is the typical distance of elec-154 tronic thermal diffusion before the heat is conducted out 155 of the electronic system, and hence correponds to the 156 effective length over which the junction is sensitive to 157 heat inputs (in this case, plasmon decay heat). At this 158 point it is worthwhile to compare to other hypotheti-159 cal non-thermal detection mechanisms, where the junction would sense directly the incident plasmon power. In that case, the signal would be proportional to the average plasmon intensity precisely at the junction, and hence proportional to the un-diffused decay heat along the junction. As we show in Fig. 3b, such mechanisms would produce a too-short decay length, determined only by the plasmon energy propagation length, $\text{Im}[2k_{\rm p}]^{-1}$.

The requirement of some diffusion to match the data confirms our picture that the detection mechanism does 169 not rely on direct rectification of the plasmon at the junc-170 tion, but instead is based on sensing the temperature rise 171 from plasmon decay. Further evidence along this line is 172 shown in Fig. 4a, where we have analyzed the photocur-173 rent decay by a fitted exponential decay length, at sev- $_{\rm 174}$ eral different carrier densities. This dependence disagrees 175 both quantitatively and qualitatively with a direct detec-176 tion mechanism. Instead, a density-dependent value of l_T

Next, we show tunability of the nature and strength 179 of the plasmon launching, with varying carrier density 180 (Figs. 4b,c). Figure 4b shows the dependence of the 181 photocurrent on the gate voltage under the tip. The 182 data show several features simultaneously evolving with 183 carrier density. There are two sign changes in phodirect three-dimensional near field coupling effects, giv- 190 attribute these to the two ways that graphene can absorb ing some disagreement within ~ 100 nm of the edge and $_{191}$ power from the tip: direct heating or plasmon launching, 192 which are both captured in our quantitative electrody-Two complex parameters, $k_{\rm p}$ and r, are key for match- 193 namic calculations of the absorbed power (Fig. 4c, details ing the y_{tip} dependence, examined in detail in Fig. 3a. $\frac{\#}{154}$ in Methods and Supplement). The launched plasmon Whereas $Re[k_p] = 2\pi/\lambda_p$ determines the fringe spac- 195 power grows strongly with carrier density primarily due termines the number of visible fringes. In particu- 197 values of λ_p couple poorly to the tip due to their strong the overall magnitude and phase of the fringes, 29 from |r| 201 unblocked interband transitions, possible when the Dirac

> Finally, we quantitatively evaluate the sensitivity of 208 should be of order 20 mA/W indicating that the ob-



for various carrier densities.

209 served 10 nA-level signals arise from a 500 nW absorbed 210 power. This suggests that the calculation of Fig. 4c has underestimated the absorbed power, not surprising 268 ulated at a frequency of approximately 250 kHz with ampli- $_{212}$ due to the large uncertainty in estimating the magni- $_{269}$ tude of 60 nm. The location of the etched graphene edge 213 tude of the electric fields excited at the tip. Still, a 270 $(x_{\rm tip}=0)$ was determined from the simultaneously-measured 214 straightforward calculation of the device noise (Johnson—271 topography.²¹

Nyquist noise of $\sim 1 \,\mathrm{k}\Omega$) indicates a noise equivalent power of 400 pW/ $\sqrt{\text{Hz}}$, a number which may be signifi-217 cantly improved by optimization of device geometry and 218 contact resistance. This can be compared to proposed 219 plasmon sources such as the graphene thermal plasmon ₂₂₀ radiator, ³⁰ which is a hot graphene strip (at $\sim 500 \text{ K}$) 221 adjacent to a room temperature graphene channel. Such a source would emit plasmon power on the order of tens of nanowatts, 30 which would be detectable using a graphene junction device such as ours. The fast cooling time intrinsic of the graphene electron gas also points towards the possibility of high speed ($\sim 50 \text{ GHz}$) plasmonic detection.31

In conclusion, we have shown that a graphene ho-229 mojunction serves as an electrical detector for the mid-230 infrared plasmons that are carried by the graphene itself. 231 The available evidence strongly indicates that thermo-232 electric action is detecting the energy of the plasmon af-233 ter it has decayed and that thermal diffusion plays an important role in spreading the decay energy. The pre-235 sented concept opens the door to graphene plasmonic 236 devices where inefficient plasmon out-coupling to light is 237 unnecessary. We anticipate in the future that this detec-238 tor may be paired with a local plasmon source such as 239 those based on thermal³⁰ or tunneling emission,³² result-240 ing in an end-to-end mid infrared optical system at sizes 241 far below the light diffraction limit.

METHODS

Device fabrication started with an 10 nm, low surface 244 roughness AuPd alloy gate film patterned by electron beam 245 lithography, on top of an oxidized Si substrate. The gap FIG. 4. Gate dependence. a, Carrier density dependence 246 separating the gates from each other was 150 nm, as inof photocurrent decay length away from junction. The de- 247 dicated in the figures. An hBN-graphene-hBN stack was cav length was obtained by an exponential fit to $I_2(x_{\rm tip})$ for $_{248}$ then prepared by the van der Waals assembly technique, 33 $x_{\rm tip} > 0.3 \,\mu{\rm m}$, far away from the edge. This was done for $_{249}$ and placed on top of the AuPd gate layer. The bottom several values of $n_{\rm L} \sim -2 \cdots 2 \times 10^{12} \,{\rm cm}^{-2}$, resulting in the $_{250}$ hBN film (between graphene and metal) thickness of 27 nm error ranges shown. The solid curves show the corresponding $_{251}$ was chosen to isolate the plasmonic mode from interacting decay in our model assuming the indicated thermal lengths, 252 with the gate metal, while still allowing for strong gate efand the dashed curve shows the result neglecting thermal dif- $_{253}$ fect. The top hBN film was made thin (9 nm) to allow fusion. b, Dependence of photocurrent on $n_{\rm L}$, at various tip $_{254}$ for plasmon launching by the s-SNOM method. The depositions away from the graphene edge. This scan was taken $_{255}$ vice geometry as well as the edge contacts were defined by 300 nm left of the junction with $n_{\rm R}=0.26\times10^{12}\,{\rm cm^{-2}}$. c, $_{256}$ dry etching and electron beam evaporation in the method Power absorbed in graphene, calculated from a rounded-tip 257 of Ref. 33. The dry etching depth was only 11 nm, leaving electrodynamic model. The dashed curve shows the absorbed $_{258}$ most of the bottom hBN thickness remaining in order to avoid power as it would be with only the real part of the graphene 259 leakage. Gate voltages were converted to carrier sheet denconductivity retained (i.e., without plasmons). Insets: The 260 sity via $n_{\rm L,R} = (0.73 \times 10^{16} \, {\rm m}^{-2} \, {\rm V}^{-1})(V_{\rm L,R} - 0.09 \, {\rm V})$, where induced charge density oscillation in the graphene calculated 261 the offset was determined by examining gate dependences and 262 the coefficient was calculated as the static capacitance of the ²⁶³ 27 nm hBN layer with dielectric constant 3.56.²¹

> The s-SNOM used was a NeaSNOM from Neaspec GmbH, 265 equipped with a CO₂ laser. The probes were commercially-266 available metallized atomic force microscopy probes with an 267 apex radius of approximately 25 nm. The tip height was mod

In Fig. 2, we solve the Helmholtz wave equation

$$k_{\rm D}^2 \rho(x, y) + \nabla^2 \rho(x, y) = f(x, y),$$
 (1)

273 for a localized sourcing distribution f(x,y) (concentrated at $x_{\text{tip}}, y_{\text{tip}}$, where k_{p} is the complex plasmon wavevector. Here ρ represents the spatial dependence of the oscillating charge density $\operatorname{Re}[\rho(x,y)e^{-i\omega t}]$. The reflective boundary at y=0 is asserted by the method of images: solving (1) for free space, 327 adding a virtual copy at $-y_{\rm tip}$ multiplied by a complex reflection coefficient r, and discarding the virtual solution below y=0. Dissipation in the graphene converts the plasmon to a decay heat distribution proportional to $|\vec{\nabla}\rho|^2$. Note that the direct product of plasmon absorption is actually a singleparticle excitation, but this can only travel a < 50 nm distance before thermalizing with the collective graphene elec- ${
m tron~gas.}^{31}$ This heating distribution is diffused,

$$l_T^{-2}(T(x,y) - T_0) - \nabla^2 T(x,y) \propto |\vec{\nabla}\rho(x,y)|^2$$
 (2)

286 to yield T(x,y), the local temperature distribution, with edge boundary condition $\partial_y T|_{y=0} = 0$. The parameter l_T is the characteristic length of lateral heat spreading before sinking 289 to the substrate at temperature T_0 . Finally the average tem-290 perature rise on the junction, which drives the thermoelectric ²⁹¹ effect, is represented by the quantity $\overline{\Delta T}^{\mathrm{junc}}$:

$$\overline{\Delta T}^{\text{junc}} = \frac{1}{W} \int_0^W \mathrm{d}y \, T(0, y) - T_0 \tag{3}$$

factors drop out due to normalization. The case of direct plasmon detection is found in the limit $l_T \to 0$, in which case the signal is determined by the y-integral of $|\vec{\nabla}\rho|^2$.

The solid curves in Figure 4a result from performing an exponential fit to the modelled $\overline{\Delta T}^{\text{junc}}$. For each |n| we estimated $k_{\rm p}$ using the fitted $k_{\rm p}$ from high |n| (Figs. 2,3) and the trend $1/k_{\rm p} \propto \sqrt{|n|}$ found in our previous study.²¹

Our electrodynamic calculation (Fig. 4c) consists of a tip charge distribution, calculated via a regularized boundaryelement electrostatic model,³⁴ fed into a multilayer transfer matrix calculation for the hBN-graphene-hBN-metal stack. An incident field of 0.3 MV/m was estimated from the experimental 10 mW incident laser power, which is focussed to a diffraction-limited spot (NA 0.5, 10.6 µm wavelength). The tip surface was taken as a circular hyperboloid of 50° opening angle and a 25 nm curvature radius at the apex, with a 5 μm length yielding a 45× tip electric field enhancement factor over the incident field. The 3D charge distribution was remapped to a 2D charge distribution located a distance z_{tip} from the top hBN surface and this distribution was oscillated at 28 THz, with accompanying in-plane currents. The absorbed power in the graphene, $\frac{1}{2} \operatorname{Re}[\vec{J}^* \cdot \vec{E}]$, was calculated $\frac{\#}{\$61}$ R.H. is co-founder of Neaspec GmbH, a company pro-316 for 36 different z_{tip} values from 0 to 60 nm, and this height 362 ducing scattering-type scanning near-field optical microscope 317 dependence was then used to simulate the second harmonic 363 systems such as the ones used in this study. All other authors 318 demodulation process, arriving at a second-harmonic power 364 declare no competing financial interests.

 $_{319}$ P_2 that best corresponds to the studied current I_2 . The hBN $_{320}$ relative permittivity at this frequency was taken as 8.27+0.16i $_{321}$ in-plane and 1.88+0.04i out-of-plane. 21 The graphene conduc-322 tivity used was the local finite-temperature RPA conductivity formula, 35 taking care to map from $(E_F - E_D)$ to n using the appropriate Fermi-Dirac integral.

Extended discussion on these models can be found in the 326 Supplementary Material.

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CONTRIBUTIONS

M.B.L. performed the measurements, analysis, modelling, and wrote the manuscript. Y.G. and C.T. fabricated the sam-355 ples. A.W. and P.A.-G. helped with measurements. K.W. and 356 T.T. synthesized the h-BN samples. J.H., R.H. and F.H.L.K. 357 supervised the work, discussed the results and co-wrote the 358 manuscript. All authors contributed to the scientific discus-359 sion and manuscript revisions.

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