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Population density controls on microbial pollution across the Ganga catchment

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1 2	Population density controls on microbial pollution across the Ganga catchment
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#### 21 Abstract

22 For millions of people worldwide, sewage-polluted surface waters threaten water security, 23 food security and human health. Yet the extent of the problem and its causes are poorly 24 understood. Given rapid widespread global urbanisation, the impact of urban versus rural 25 populations is particularly important but unknown. Exploiting previously unpublished 26 archival data for the Ganga (Ganges) catchment, we find a strong non-linear relationship 27 between upstream population density and microbial pollution, and predict that these river 28 systems would fail faecal coliform standards for irrigation waters available to 79% of the 29 catchment's 500 million inhabitants. Overall, this work shows that microbial pollution is 30 conditioned by the continental-scale network structure of rivers, compounded by the location 31 of cities whose growing populations contribute c. 100 times more microbial pollutants per 32 capita than their rural counterparts.

33

#### 34 Highlights

Faecal coliform concentration is strongly related to upstream population density [80]
Local rivers predicted to fail WHO irrigation standards for 79% of the population [83]
Rivers receive c. 100 times more sewage per capita from urban than rural populations [84]
Microbial pollution is conditioned by river network structure and settlement pattern [84]
Himalayan headwaters continue to dilute microbial pollution far downstream [74]

41 **Keywords:** faecal coliform; river network; population density; catchment-scale;

42

#### 43 Background

Rising demands on water resources raise concerns about the sustainable provision of clean
water worldwide. Unclean water poses significant risks of diarrhoea, opportunistic infections,
and consequent malnutrition accounting for ~1.7 million deaths annually; of which >90% are

in developing countries and almost half are children [Prüss-Ustün et al., 2014]. These deaths
are primarily due to ingestion of faecal pathogens from humans or animals [Ashbolt et al.,
2004; Kotloff et al., 2013; Prüss-Ustün et al., 2014].

50 India's growing population and economy are driving rapid urbanisation (30% of the 51 population now live in urban areas [Census of India, 2011a]) and exerting increased pressure 52 on surface and groundwater availability. In rural areas  $\sim 67\%$  of the population defecate in the open [Census of India, 2011b], a practice that poses severe risk to health and safety [Clasen 53 et al., 2010; Mara et al., 2010; Ziegelbauer et al., 2012; Kotloff et al., 2013]. While in urban 54 55 areas ~80% of the population have access to a toilet [Census of India, 2011b], but only ~30% 56 are connected to a sewage pipeline and few pipelines are connected to a treatment plant 57 [Narain, 2012]. The impact of these sanitation problems on surface water quality has been 58 documented for many years at individual sample locations or river reaches across India [Bhargava, 1983; Mukherjee et al., 1993; Baghel et al., 2005; Mishra et al., 2009; Central 59 60 Pollution Control Board, 2010]. However, there has been no catchment-wide quantification 61 of the problem and limited indication of what is driving it. The former is essential to fully understand the scale of intervention required, while the latter might inform decision-making 62 on 'what to do where'. Urban areas often dominate the microbial pollution signal in rivers 63 64 [Tchobanoglous et al., 1991; Kay et al., 2008; McGrane et al., 2014] but there is little consensus on the extent to which this reflects an increased impact per capita or simply a 65 66 larger population and thus source. This difference is important since a higher per capita impact indicates reduced attenuation, perhaps due to more efficient delivery to the river 67 system or less efficient treatment. If the difference can be attributed to per capita contribution 68 69 this will define the extent to which urban or rural focused interventions will improve surface 70 water quality.

We address this question using archival water quality data from across the Ganga (Ganges) catchment and show the pattern of microbial pollution in its major rivers. We compare instream concentrations of a pollution proxy with upstream densities of the two major sources of faecal pathogens (humans and livestock) at 100 sites spanning an approximate surface area of 10<sup>6</sup> km<sup>2</sup>.

76 Faecal pathogens are difficult to measure, however thermo-tolerant coliforms, which originate in faeces (i.e. faecal coliforms, FC), are easily detectable and routinely monitored as 77 indicator organisms [Ashbolt et al., 2001]. FCs are not a perfect predictor of human pathogen 78 79 presence, rather they establish connectivity between defecation and some receiving 80 environment which could be contributed to by a pathogen carrier. New host-specific tracing 81 techniques allow more precise tracking of microbial pollution sources that can help to better 82 assess risks to human health [Harwood et al., 2014, Field and Samadpour, 2007]. However, such techniques are not used within routine monitoring in India and thus do not have the 83 spatial coverage required for our analysis. Furthermore, the use of FCs for monitoring 84 85 pollution is still regarded as a viable measure of drinking and irrigation water quality [WHO, 86 2017].

Two key issues that must be addressed are: 1) the extent to which the FC signal that we 87 88 observe reflects human sources; and 2) the potential impact of FC die-off in our pollution tracer. Upstream livestock and human population densities are strongly correlated at the 89 90 catchment scale limiting our capacity to identify the source of the pollution signal. To address 91 this, we seek to de-correlate the predictor variables by using a mixing model to estimate 92 contributions from each non-overlapping segment of the catchment (our sub-catchments). To 93 address the impact of die-off in our pollution tracer we adjust the population and livestock 94 densities using a distance decay function then seek decay parameters that will maximise performance of our statistical model. 95

In the sections that follow we first introduce our null hypothesis that pollution should be linearly related to source density (both with and without accounting for die-off). We then detail our data sources and methods for their analysis; and introduce the mixing model that we use to calculate effective FC concentrations and source densities for each sub-catchment (the non-overlapping segments of the catchment).

101

### 102 Theory: Expected relationship between FC concentration and upstream source density

#### 103 with and without die-off

104 The FC concentration ( $C_{FC}$ ) at a given location is defined by the ratio of the FC flux ( $Q_{FC}$ ) to 105 the water flux ( $Q_w$ ):

$$106 \qquad C_{FC} = \frac{Q_{FC}}{Q_w}$$

107 Under the assumption that there is no die-off in FCs over time, the FC flux is calculated108 from:

109 
$$Q_{FC} = (P_h N_h + P_a N_a) = (P_h \rho_h + P_a \rho_a) A$$
 (2)

110 where:  $P_h$  is the production rate of FCs per human head [MPN #<sup>-1</sup> T<sup>-1</sup>];  $P_a$  is the production 111 rate per head of livestock [MPN #<sup>-1</sup> T<sup>-1</sup>];  $N_h$  and  $N_a$  are the total upstream population of 112 humans and livestock respectively [#];  $\rho_h$  and  $\rho_a$  are the upstream population densities of 113 humans and livestock respectively [# L<sup>-2</sup>]; and A is the catchment area [L<sup>2</sup>]. Under the 114 assumption of spatially uniform and time invariant runoff  $R_w$  [L T<sup>-1</sup>] the water flux  $Q_w$  [L<sup>3</sup> T<sup>-</sup> 115 <sup>1</sup>] is calculated from:

$$116 \qquad Q_w = R_w A \tag{3}$$

Substituting equations 2 and 3 into equation 1 gives the following equation for FCconcentration at each measurement point as a function of upstream population density.

119 
$$C_{FC} = \frac{(P_h \rho_h + P_a \rho_a)}{R_w} = k_h \rho_h + k_a \rho_a$$
 (4)

120 where:  $k_{h=}P_{h}/R_{w}$  and  $k_{a}=P_{a}/R_{w}$ . It is clear from this relationship that under these assumptions 121  $C_{FC}$  should be a linear function of upstream population and livestock density with the 122 gradients defined by the ratio of production rate, P, to runoff,  $R_{w}$ .

123

The assumption of no FC die-off is unlikely to be true but controls on die-off remain poorly understood. Given the uncertainties, die-off is most often represented using an exponential decay based on first order kinetics [Crane and Moore, 1986; Sadeghi and Arnold, 2002; Cho et al., 2012]:

128 
$$Q_{FC} = Q_0 e^{-k_1 t}$$

where:  $Q_0$  is the FC flux at time  $t_0$  (the time of exit from the gut) [MPN T<sup>-1</sup>], t is time since exit [T],  $k_1$  is a decay coefficient [T<sup>-1</sup>]. Assuming uniform time invariant FC velocity from source to measurement point the FC flux  $Q_{FC}$  can be expressed as a function of distance:

(5)

132 
$$Q_{FC} = Q_0 e^{-k_1 \left(\frac{x}{v}\right)}$$
 (6)

where: x is the travel distance from source to measurement point [L] and v is the characteristic velocity [L T<sup>-1</sup>]. Changing population (of people or livestock) with distance x upstream of the sampling point can be calculated as the derivative of N(x):

136 
$$n(x) = -\frac{dN}{dx} = -\rho(x)\frac{dA}{dx} - A(x)\frac{d\rho}{dx}$$
(7)

Assuming that FC production rates are time invariant and incorporating characteristic velocity into the decay coefficient to express decay in terms of distance, the FC flux can be calculated by combining equations 2, 6 and 7 and integrating over the range of travel distances from the measurement point to the furthest point upstream:

141 
$$Q_{FC} = \int_{0}^{x} \max\left( \left( P_h \, n_h(x) + \, P_a \, n_a(x) \right) \, e^{-k \, x} \right) \, dx \tag{8}$$

where change in population (for both humans and livestock) and area are a function of travel distance; and  $k=k_1/v$  the distance decay coefficient [L<sup>-1</sup>]. Substituting equations 3 and 8 into equation 1 gives the following equation for FC concentration:

145 
$$C_{FC} = \int_{0}^{x} max \left( \frac{(P_h n_h(x) + P_a n_a(x)) e^{-kx}}{R_w A} \right) dx$$
 (9)

This can be implemented in discrete form by summing over the ncells upslope of the measurement point where for each cell the flow path lengths and routes are derived from digital elevation data, and human and livestock population data from the sources described below.

150 
$$C_{FC} = \sum_{i=1}^{ncells} \left( \frac{(P_h \rho_{hi} + P_a \rho_{ai})A_i e^{-kx_i}}{R_w A_i} \right)$$
 (10)

151 where:  $\rho_{hi}$  and  $\rho_{ai}$  are the density of human and animal populations respectively in cell i;  $A_i$  is 152 the area of cell i; and  $x_i$  is the average flowpath length from cell i to the measurement point. 153 Rearranging and simplifying equation 10 gives:

154 
$$C_{FC} = k_h \Sigma_{i=1}^{ncells} \left( \rho_{hi} e^{-k x_i} \right) + k_a \Sigma_{i=1}^{ncells} \left( \rho_{ai} e^{-k x_i} \right) \quad (11)$$

where:  $k_{h=}P_{h}/R_{w}$  and  $k_{a}=P_{a}/R_{w}$ . Re-arranged in this form, equation 11 shows that accounting for FC die-off,  $C_{FC}$  remains a linear function of population and livestock density transformed to account for flowpath length. As in the no die-off case (equation 4), the linear coefficients reflect the ratio of (human or livestock) production rate to runoff.

159

#### 160 Methods

We used water quality samples from 100 locations across the Ganga catchment (Figure 1), collected and analysed by six agencies following a uniform protocol. Total and faecal coliform concentrations were estimated using the standardised 9221B and 9221E multiple tube fermentation techniques [APHA, 1995] to establish the most probable number (MPN) of

165 faecal coliforms per 100ml. At each site, we collated 10 years of data (2002-2012). The 166 frequency with which this data was sampled varies between sites, from three samples per 167 year at the two most remote Himalayan sites, to quarterly for 24 more Himalayan sites and 168 one or two samples per month at the remaining sites. At ~30 sites, samples were collected at two locations across the river in some years in order to improve representation. This data was 169 170 quality checked for potential data entry or measurement errors. We removed a total of 63 observations where FC concentrations exceeded Total Coliform (TC) concentrations (since 171 FC is a subset of TC). We also removed two observations at a single site on the same date 172 where FC concentration exceeded  $10^{10}$  MPN / 100 ml. We consider this to be suspicious 173 given that the concentration is ~100 times the upper end of the range of observed 174 175 concentrations for sewage influent [Tchobanoglous et al., 1991]. Removing suspicious 176 observations results in a loss of <0.5% of the full dataset and <3% at any individual site. The error-checked FC data at each site were poorly approximated by a normal distribution but 177 were generally well approximated by a log-normal distribution, thus we used geometric 178 179 means to summarise FC concentration for each site throughout our analysis.

180 To estimate upstream population density we used the GPWv3 gridded synthesis of census data from 2000 [Balk and Yetman, 2004; Balk et al., 2010]. To estimate livestock density we 181 182 used the FAO global gridded livestock density data [Wint and Robinson, 2007; Robinson et al., 2014], weighted by estimates of FC production rates for each livestock type (cow and 183 buffalo: 10<sup>11</sup> MPN/# day; goats and sheep: 1.2 x 10<sup>10</sup> MPN/# day; pigs: 1.1 x 10<sup>10</sup> MPN/# 184 day; poultry: 1.4 x 10<sup>8</sup> MPN/# day) [ASAE Standards, 1998]. Upstream area, upstream 185 population density (UPD) and upstream livestock density (ULD) for each sample point were 186 calculated using a D8 flow routing algorithm [O'Callaghan and Mark, 1984; Schwanghart 187 188 and Scherler, 2014] and the hydrologically corrected 90 m SRTM DEM [Farr et al., 2007]. To examine the influence of coliform die-off in transit and thus relax the assumption that 189

190 coliforms behave as conservative tracers we introduced an exponential decay in coliform 191 concentration with distance from the source. We sampled the shape parameter that defines 192 the rate of distance decay at 500 logarithmic intervals from 10<sup>-8</sup> to 10<sup>-1</sup> km<sup>-1</sup> testing model 193 performance in each case using ordinary least squares regression.

194

### 195 Mixing model

196 The observation locations form a nested set of catchments where 82% of observation sites 197 have at least one observation site upstream. We deal with this nested sampling in two ways. 198 First, by assessing the results for only non-nested (independent) catchments, however this 199 considerably reduces the number of available observations. Second, by performing the 200 analysis using sub-catchments, where these are defined as the part of the catchment that drains to the current sample site without first draining through any upstream sample site. The 201 202 result of this definition is segmentation of the entire Ganga catchment into 100 non-overlapping 203 sub-catchments.

Effective source density and FC concentration are then calculated for each sub-catchment using an approach similar to that of Granger et al. [1996] and Portenga et al. [2015] for effective erosion rates in nested catchments. To do this we assume that catchment area can be used as a proxy for discharge (equation 3) and use a mixing model to calculate the concentration of the FC input for the sub-catchment ( $C_{FCr}$ ) given the catchment area and FC concentration at the upstream and downstream boundaries:

210 
$$C_{FCr} = \frac{Q_{FCr}}{A_r} = \frac{C_{FCd} A_d - \sum_{i=1}^n (C_{FCui} A_{ui})}{A_d - \sum_{i=1}^n (A_{ui})}$$
(12)

where:  $C_{FCui}$  is the FC concentration at upstream boundary i; and  $C_{FCd}$  is the FC concentration at the downstream boundary of the sub-catchment;  $A_{ui}$  is the catchment area for upstream boundary i;  $A_d$  is the catchment area for the downstream boundary of the sub-catchment; and

n is the number of upstream boundaries. We repeat the same process to calculate the human and animal populations densities ( $\rho_r$ ) within the catchment area that drains into this subcatchment:

217 
$$\rho_r = \frac{N_r}{A_r} = \frac{\rho_d A_d - \sum_{i=1}^n (\rho_{ui} A_{ui})}{A_d - \sum_{i=1}^n (A_{ui})}$$
(13)

218 where:  $\rho_{ui}$  is the upstream population density of upstream boundary i; and  $\rho_d$  is the upstream 219 population density of the downstream boundary of the sub-catchment.

220

#### 221 Results

### 222 Observed pattern of FC concentrations

223 Our results suggest that high FC concentrations previously reported at the reach and subcatchment scale [Mukherjee et al., 1993; Baghel et al., 2005; Mishra et al., 2009; Central 224 225 Pollution Control Board, 2010] do not reflect isolated pockets of poor water quality but 226 extensive pollution across the catchment. Decadal mean FC concentrations at sites across the Ganga catchment range from 3x10° to 2.5x10° MPN/100ml. 70% of sites fail Indian 227 Government desirable bathing limits [Central Pollution Control Board, 2008] with those that 228 229 pass located almost exclusively in the sparsely populated catchment headwaters. On the more 230 populous plains, 70 of the 80 sites fail the desirable limits and 63 of the 80 sites fail the 231 maximum permissible 2500 MPN/100ml limit [Central Pollution Control Board, 2008]. Locally high FC concentrations are generally associated with large population centres (Figure 232 1), most markedly for rivers with smaller catchment areas (e.g. the Varuna at Varanasi). FC 233 234 concentrations are moderately reduced downstream of the Yamuna-Ganga confluence as 235 tributaries with lower FC concentrations dilute the main stem. Further downstream, even large cities (e.g. Patna) have limited influence and many samples on the main stem have very 236 237 similar FC concentration, reflecting the central tendency of water quality with increasing 238 catchment area for nested catchments.

240 Catchment scale relationships between FC concentration and upstream source density 241 Since people and livestock are the primary sources of FCs, we expect FC concentration to 242 increase with the upstream density of these sources. Figure 2 suggests that the data fit this expectation. If FC production per capita is spatially uniform, delivery to the river is 243 244 independent of population density, and if water flux is a linear function of catchment area, 245 then we expect FC concentration to be a linear function of upstream source density with the 246 form  $y=\beta x$  (see equations 1-4 for a full derivation). Variability in delivery to the river network or transit time through the river network that is uncorrelated with population density 247 248 will introduce scatter to the relationship but should not alter its functional form. However, comparing the data with linear contours in Figure 2 shows that the data are not a good fit to a 249 linear function ( $r^2 < 0.1$ ). Power functions are a better fit ( $r^2=0.69$  for UPD and 0.62 for ULD) 250 251 but over-predict high and low FC values and under-predict central values with both UPD and 252 ULD. Quadratic relationships offer a further improvement ( $r^2=0.71$  for UPD and 0.68 for ULD) suggesting positive curvature in log-log space but have a physically unreasonable 253 negative slope at low population densities. Residuals from the quadratic function, fitted by 254 ordinary least squares regression, for both population and livestock show some 255 256 heteroscedasticity, though White [1980] and BPK [Breusch and Pagan, 1979; Koenker, 1980] 257 tests return p-values that are always below 0.1. Given this moderate heteroscedasticity and 258 the insensitivity of ordinary least squares coefficients to heteroscedasticity we do not pursue more complex variance weighted analyses. UPD alone explains slightly more of the variance 259 260 in FC concentration than ULD, but there is little difference between the explanatory power of 261 these predictors, and their combination in a multiple quadratic regression offers little 262 improvement ( $R^2=0.71$ ). This is consistent with the strong correlation between upstream population and livestock densities (Figure 2c). A cubic function constrained to monotonic 263

264 increase over the range of the data gives a similar performance to the quadratic ( $r^2=0.71$  for 265 UPD and 0.68 for ULD). A linear spline (in log-log space) with a single interior knot (i.e. 266 piecewise power function) is the best-fit for both individual predictors ( $r^2=0.73$  for UPD and 267 0.71 for ULD), suggesting a threshold rather than continuous change in power relationship between UPD and FC concentration. Finally, we test one further null hypothesis that there are 268 269 two ranges of source density (population or livestock) with FC concentrations represented by their average value over each range. This model is important to exclude given the appearance 270 of clustered points within Figure 2 but has difficult physical implications. It implies a step 271 272 change in contribution at some source density and a constant contribution independent of 273 source density change (i.e. a declining per-head contribution) within each range. The 'step 274 model' (r<sup>2</sup>=0.69) does not outperform any of the curved functions (quadratic, cubic or linear 275 spline) for UPD, though it is a slight improvement on quadratic and cubic spline functions for 276 ULD (r<sup>2</sup>=0.70). These results demonstrate that there is positive curvature to the FC-UPD relationship independent of the particular functional form (quadratic, cubic or linear spline) 277 278 under consideration; and that the FC-ULD relationship also contains positive curvature but 279 can be almost as well described as two FC distributions at high and low population density.

280

#### 281 Sub-catchment relationships between FC concentration and upstream source density

As for catchment analysis, the sub-catchment analysis suggests that people and livestock are the primary sources of FCs with FC concentration increasing with the upstream density of these sources (Figure 3). The relationship between source density and FC concentration is not linear for sub-catchment based analysis or catchment based analysis. Figure 3 shows that as in the catchment analysis the data are not a good fit to any linear model ( $r^2 < 0.2$ ). Power functions are a better fit ( $r^2=0.54$  for UPD and 0.17 for ULD) but over-predict high and low FC values and under-predict central values for UPD. For ULD the fit is very poor, suggesting

289 that in the sub-catchment based analysis livestock density is only a weak control on FC 290 concentration. Quadratic relationships (in loglog space) offer further improvement for UPD 291  $(r^2=0.63)$  but not for ULD  $(r^2=0.16)$ . UPD alone explains considerably more of the variance in FC concentration than ULD. Their combination in a multiple quadratic regression offers 292 some improvement (R<sup>2</sup>=0.72). This reflects the reduced correlation between UPD and ULD 293 294 for sub-catchment ( $r^2=0.66$ ) rather than catchment ( $r^2=0.95$ ) analysis (compare Figure 2c with 3c). The linear spline with a single knot (i.e. piecewise power function) or cubic function (in 295 loglog space) constrained to monotonic increase result in similar fits relative to a quadratic 296 for both UPD ( $r^2=0.63$  in both cases) and ULD ( $r^2=0.16$  and 0.15 respectively). As in the 297 298 catchment analysis this suggests that there is not clear evidence for a threshold rather than 299 continuous change in power relationship between UPD and FC concentration when examined 300 at the sub-catchment scale. The results from these three (quadratic, cubic and linear spline) 301 approaches demonstrate that there is positive curvature to the FC-UPD relationship independent of the particular functional form under consideration. They also demonstrate that 302 303 UPD is a far better predictor than ULD for sub-catchment scale analysis and that there is 304 some merit in considering the two in combination. This suggests that most instream FCs are 305 human derived.

306

### 307 Per capita impact on instream FC concentration

Positive curvature to the FC-UPD and FC-ULD relationships indicate that FC concentration increases with upstream source density at an increasing rate per unit increase in upstream source density. This can be interpreted as the change in FC per capita with increasing upstream source density. The gradient of the line in logarithmic space reflects its exponent in linear space thus: values >1 indicate positive curvature and increasing per capita impact, those <1 indicate negative curvature and decreasing per capita impact with increased source

density. At low upstream source densities (<10 people or 6 livestock per km<sup>2</sup>), FC concentrations are low and the gradient of all three best-fit curves is slightly less than one indicating a slight decline in per capita impact with increasing upstream source density. At source densities from 10-60 people or 6-30 livestock per km<sup>2</sup> the gradient of all three best-fit curves reaches then exceeds unity, indicating that per capita impact reaches a minimum and begins increasing with increasing upstream source density.

For population density, quadratic, cubic and linear spline fits all predict a very similar relationship between UPD and FC concentration for  $10^2 < \text{UPD} < 10^3 \ \text{\#/km^2}$  (Figure 2a). Over this range the predicted FC concentration increases by three orders of magnitude (from  $10^2$  to  $10^5 \text{ MPN}/100 \text{ml}$ ), indicating a 100-fold increase in per capita impact. Over the same range in population density ( $10^2 < \text{UPD} < 10^3 \ \text{\#/km^2}$ ) there is considerable variability in the per capita contribution from no change at the lower limit to a 10,000-fold increase at the upper limit.

326 A similar comparison can be made for individual sites, with the linear trend lines in Figure 2a acting as contours for per capita impact. For example, moving downstream from the 327 328 catchment with lowest population density, UPD increases 10-fold from Badrinath to Srinagar (7-77 #/km<sup>2</sup>) but FC concentration increases only three-fold (3-10 MPN/100ml), thus per 329 capita impact declines by a factor of 3. Continuing downstream from Srinagar to Kanpur 330 331 UPD increases by a factor of 6 (77 to 450 #/km<sup>2</sup>) while the FC concentration increases by a factor of 1600 (10 to 1.6x10<sup>4</sup> MPN/100ml), thus impact per capita increases by a factor of 332 333 300. Per capita impact increases by a factor of 60,000 from its minimum for the rural Pindar 334 catchment (B) to its maximum for the densely populated Yamuna at Delhi (A). These results 335 indicate that urban populations contribute more sewage to the river per capita than rural 336 populations and that this increase: 1) depends on the difference in population densities, rather 337 than changing sharply at a particular density; 2) is large on average (a factor of 100); and 3) 338 is highly, and asymmetrically, variable (ranging from a factor of 1 to 10,000).

### 340 **Discussion**

#### 341 The relative importance of human or livestock FC sources

342 Both UPD and ULD are good predictors of FC concentration based on catchment scale analysis. This may reflect the importance of both sources, but is also very likely due to the 343 344 strong positive correlation between UPD and ULD in the catchment based analysis (Figure 2c), which makes it difficult to distinguish between the sources based on these data alone. 345 346 When calculated over large areas population and livestock density are highly correlated. 347 However, at small scales population and livestock density can become de-correlated (e.g. in 348 cities, where population density is high but livestock density low). Our sub-catchment based 349 analysis breaks the catchment into smaller non-nested segments, disrupting the correlation 350 between UPD and ULD (Figure 3c). This analysis shows a small reduction in the percentage of variance in FC concentration explained by UPD and a large reduction in that explained by 351 ULD. In the sub-catchment based analysis UPD is a much better predictor of FC 352 353 concentration than ULD.

354 This is consistent with simple accounting estimates of export coefficients calculated using 355 population and livestock densities with estimated FC production rates for the loading terms 356 and observed FC concentration as the output. Assuming a human production rate of  $2 \times 10^9$ MPN/# day [Tchobanoglous, 1991] and livestock production rates detailed in the methods 357 section, livestock-derived FC loads produced on any given day range from 2x10<sup>10</sup> MPN/km<sup>2</sup> 358 359 day (for ULD =  $3 \#/km^2$ ) to  $1.5 \times 10^{13}$  MPN/km<sup>2</sup> day (for ULD =  $200 \#/km^2$ ) while population derived FC loads range from  $1.4 \times 10^{10}$  MPN/ km<sup>2</sup> day (for UPD = 7 #/km<sup>2</sup>) to  $2 \times 10^{12}$  MPN/ 360  $km^2$  day (for UPD = 1000 #/km<sup>2</sup>). Yet over this range of source densities FC concentrations 361 362 increase from  $2x10^{\circ}$  to  $1x10^{\circ}$  MPN/100 ml on average. This results in export coefficients 363 >100 times larger at high livestock and population densities than at low densities. It is

difficult to conceive of a mechanism for such an increase in export coefficient for livestock-derived FCs as a function of source density.

366

#### 367 The relative importance of local or non-local FC sources

UPD is a good predictor of instream FC concentrations across the Ganga catchment, 368 369 explaining 73% of the observed variance in decadal mean FC concentrations from a catchment scale analysis and 63% from a sub-catchment scale analysis (Figure 2a and 3a). 370 This is consistent with findings from catchments across the world [Tchobanoglous et al., 371 372 1991; Kay et al., 2008; McGrane et al., 2014], and with previous reach-scale findings in the 373 Ganga Catchment [Mukherjee et al., 1993; Baghel et al., 2005; Mishra et al., 2009; Central 374 Pollution Control Board, 2010]. However, there remains considerable variance in FC 375 concentration unexplained by either UPD or ULD, particularly at high population densities, >100 people/km<sup>2</sup> (Figures 2 and 3). Previous reach-scale studies did not account for the 376 upstream boundary condition either in terms of FC flux or upstream population [Mukherjee et 377 al., 1993; Baghel et al., 2005; Mishra et al., 2009]. These studies implicitly assumed that 378 379 point sources proximal to sample sites dominated the FC signal (perhaps due to coliform dieoff in transit). However, while many of our sites near larger settlements have high coliform 380 381 concentrations, these concentrations are better explained by upstream population density ( $r^2$ >0.7) than population of the nearest settlement (r<sup>2</sup>=0.25). Examining paired samples above 382 and below settlements suggests that in some cases, positive residuals (where FC 383 384 concentration is greater than predicted) may reflect sites immediately downstream of population centres. However, including a distance-decay function in our analysis did not 385 386 improve our ability to predict FC concentrations. Figure 4 shows that model performance is initially stable as the rate at which FCs decay with distance increases, but that the 387 388 performance is never better than that without distance decay, and that performance declines

389 markedly for decay rates greater than 0.01 %/km. This reduction in performance relates to a 390 reduction in decay-adjusted population density primarily at sites with intermediate or dense 391 populations (Figure 5). These results suggest that, UPD is an important but not singular 392 factor in defining the connectivity between sources and receiving waters that defines the timescales and thus efficiency of delivery. Our approach neglects many processes that should 393 394 be important in the transport of coliforms from source to the point of measurement (e.g. weather dependent die-off rates, hydrological connectivity, hydraulics at the cross section and 395 reach scale). However, it is encouraging that even our simple empirical model explains a 396 397 large fraction of the variance in microbial pollution concentrations.

398

### 399 Implications of the FC-UPD relationship

400 The increase in per capita impact as UPD increases likely reflects an increase in the 401 efficiency of delivery rather than FC production, perhaps due to changes in individual or corporate waste management decisions as population density increases. At low population 402 403 densities, much of the population defecate in the open or in pit latrines [Census of India, 404 2011b] where faeces are less likely to be washed into the river and FCs are more likely to die 405 *in situ*. As population density increases and towns and cities grow, the distance to open fields 406 increases and there is a need for an alternative strategy to manage faeces. This problem has historically confronted communities across the world, leading to degradation of sanitary 407 408 conditions and construction of sewers [Gandy, 2004; Allen, 2008; Benzerzour et al., 2011]. 409 Sewage systems vary in sophistication but generally involve the movement of excreta by water out of the population centre; often made possible by piped domestic water. The faeces 410 411 have a much shorter residence time in the environment and FCs will be removed primarily by 412 sewage treatment rather than die-off in the environment. In many Indian cities, the flux of 413 sewage that is, and must be, removed from the population centre through a growing network

414 of sewers and storm water drains is many times higher than the capacity of the sewage 415 treatment facilities [Ansari et al., 2000]. In this case the predominant impact of the sewage 416 network is to remove the sewage from the population centre and rapidly deliver it to the river 417 untreated. Sewage removal is essential for the public health of the city, but without effective treatment it comes at the cost of accentuated river pollution with associated public health 418 419 implications for the population downstream. Here we demonstrate as others have [Central Pollution Control Board, 2010] the severe river pollution that results. The extent to which this 420 can be addressed by following the same trajectory towards centralised 'end-of-pipe' sewage 421 422 treatment has been called into question for practical and economic reasons [Jha, 2003; 423 Bracken et al., 2007; Katukiza, 2012]. However, there is a growing range of innovative, 424 water and energy efficient, on-site alternatives [Jha, 2003; Bracken et al., 2007; Gates 425 Foundation, 2016] as well as a growing recognition that this is a social as well as physical or 426 technical issue [Burra, 2003; Sharma and Bhide, 2005; McFarlane, 2008].

It is important to emphasise that our results do not imply that open defecation is a safe approach to sewage management. Water is not the only vector for faecal-oral disease; transmission can also occur through food, insects, and direct contact [Wagner and Lanoix, 1958]. Thus safely disposing of faeces involves more than simply ensuring that they do not enter the watercourse. There is good evidence to suggest that open defecation is extremely problematic for public health and safety [Clasen et al., 2010; Mara et al., 2010; Ziegelbauer et al., 2012].

434

#### 435 Network structure controls the spatial pattern of microbial pollution

The relationship between upstream population density and FC concentration enables a simple predictive relationship, albeit with considerable scatter. This model predicts that 33–48% of rivers in the Ganga catchment fail the Indian Government's safe bathing standards,

depending on the choice of standard (Figure 6). However, many of those rivers that pass are
in sparsely populated headwaters. For 70-85% of the catchment's population, their nearest
river fails safe bathing standards [Central Pollution Control Board, 2008]; for 79% it should
not be used for flood irrigation, irrigation of crops eaten raw or where children are involved
in farming [WHO, 1989; Blumenthal et al., 2000]; and for 51% it should not be used for

The pattern of predicted FC concentration from this empirical model is strongly influenced 445 446 by the catchment's network structure (Figure 6). Sparsely populated Himalayan headwaters produce high discharges of clean water suppressing FC concentrations far downstream; 447 448 without this discharge, plains-fed rivers (e.g. Kali) have high FC concentrations throughout. 449 The most polluted reach of the Ganga is predicted to be between Kanpur and Allahabad. 450 Upstream of Kanpur the diluting effect of the headwaters persists while downstream of 451 Allahabad the Ganga is diluted first by the less polluted Yamuna (strongly influenced by the Chambal) and then by the large left bank tributaries with their headwaters in the Himalaya. 452 453 This may be the result of not only the topology but also the geometry of the network, since 454 the Ganga at Allahabad is at its furthest point from the mountain front meaning cleaner Himalayan water must travel over a larger expanse of populated plain to reach that point. 455

Interventions high up the river network have the highest potential for impacting FC concentration for a given FC flux reduction because: 1) lower discharge on these rivers means that the same FC flux reduction will lead to a larger concentration reduction; and 2) rivers are directed networks (i.e. they accumulate) thus a reduction in FC flux at a given location will impact only reaches downstream of it. Decisions of what to do where are difficult and necessarily political, with many drivers [Bulkeley and Mol, 2003], but the findings of this study can help guide strategic investment in pollution reduction.

#### 464 Conclusions

465 The rivers of the Ganga catchment are subject to widespread and, in places, severe microbial 466 pollution. 52-67% of measured sites fall below the Indian Government's upper and desirable limits for safe bathing; and for 61-70% of the population, model results suggest that their 467 nearest river falls below these same bathing standards. The network structure of the Ganga 468 469 catchment pre-conditions certain rivers to be highly polluted, and others (with large Himalayan headwaters) to be more robust against pollution, despite their location on the 470 471 densely populated plains. The entire population upstream (not only those nearby) contribute 472 to microbial river pollution but urban populations contribute more pollution per capita than 473 rural populations. How much more depends on their respective population densities. A 474 person living in an area with 1000 #/km<sup>2</sup> contributes on average 100 times more pollution to 475 the river than they would in an area with  $100 \ \text{#/km}^2$ . While this is an average in the presence 476 of considerable (asymmetric) variability, the denser population in this case contribute at least as much pollution per capita at the lower limit and up to 10,000 times more at the upper limit. 477 478 Densely populated areas dominate surface water pollution in the Ganga catchment not only 479 because of they contain many people but because their faeces are more efficiently delivered to the river network. We suggest that this increasing efficiency reflects: the transmission 480 481 speed of urban sewerage systems, delivering the coliforms to the river more quickly with less die-off; and the limited capacity for sewage treatment within these systems. Addressing this 482 483 problem requires investment in both sewage removal and treatment whether by increasing 484 existing sewerage capacity or implementing decentralised treatment solutions.

485

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#### 659 Figures



Figure 1. Network graph of decadal mean FC concentrations (circle colour) and catchment area (circle size). Large red circles indicate high FC concentration and water discharge (thus high FC flux); smaller green circles indicate lower concentration and discharge (thus low FC flux). Sites with thick blue outlines pass Indian Government desirable standards of <500 MPN / 100 ml; those with thin blue outlines pass the upper limit of <2500 MPN / 100 ml [Central Pollution Control Board, 2008]. Rivers are labelled in blue; cities are labelled in black, with approximate populations, in millions, in brackets and grey boxes to show approximate extent. Inset shows a location map of the Ganga catchment.



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Figure 2: catchment scale analysis of faecal coliform concentration against: a) upstream population density; b) upstream livestock density adjusted for variable coliform production rates; c) co-variation between upstream population and livestock density. Trend lines show quadratic (solid), cubic (dotted) and linear spline (dashed) regressions for a and b, and linear regression for c. Solid circles show non-nested (i.e. independent) observations, n=18; crosses show the full dataset, n=100. Labelled points are: A) Yamuna catchment at Delhi; and B) Pinder catchment at Karanprayag.



676 677 Figure 3: sub-catchment based Faecal Coliform concentration against: a) upstream population density and b) 678 upstream livestock density adjusted for variable coliform production rates; c) co-variation between upstream 679 population and livestock density; d) predicted v observed coliform concentrations from multiple cubic 680 regression with upstream population and livestock density. Trend lines show quadratic (solid), cubic (dotted) 681 and linear spline (dashed) regressions for a and b, and linear regression for c. Solid circles show non-nested (i.e. 682 independent) observations, n=18; crosses show the full dataset, n=100. Labelled points are: A) Yamuna 683 catchment at Delhi; and B) Pinder catchment at Karanprayag. Contours in c show prediction surface from 684 multiple regression.



Figure 4. Model performance (Adjusted  $r^2$  for FC concentration v decay-adjusted UPD) with varying distance decay coefficient (k) for the three empirical functions fitted in Figure 2. Best performance is always for no decay (k=0); small coefficients (k < 10<sup>-4</sup>) have little effect; larger coefficients result in a breakdown in model performance.





### Decay Adjusted Upstream Population Density [#/km<sup>2</sup>]

692Figure 5. Scatter plots of faecal coliform (FC) concentration against upstream population density (UPD)693adjusted with an exponential distance decay using a range of decay coefficients (k). Panels reflect decay rates694of: a) 0 %/km, b) 0.01 %/km, c) 1 %/km and d) 10 %/km. Best model performance is for no decay (k=0); small695coefficients (k < 10<sup>-4</sup>) have little effect; larger coefficients result in a breakdown in the relationship between696UPD and FC concentration.



699 Figure 6. Spatial pattern of predicted coliform concentration. Dark blue areas have concentrations below 500 700 MPN/100ml, the Indian Government's desirable limit for safe bathing [Central Pollution Control Board, 2008]; 701 light blue areas have concentrations below 2500 MPN/100ml, the upper limit for safe bathing [Central Pollution 702 Control Board, 2008]. Inset shows the fraction of the river network (blue) and population (red) for which the 703 nearest river has an FC concentration less than the x-axis value. Letters signify: (a) USA limit for safe bathing 704 [U.S. EPA, 1976]; (b) Indian government desirable limit for safe bathing [Central Pollution Control Board, 705 2008]; (c) WHO recommended limit for flood irrigation, or for crops eaten raw, or where children are involved 706 in farming [WHO, 1989; Blumenthal et al., 2000]; (d) Indian government upper limit for safe bathing [Central 707 Pollution Control Board, 2008]; (e) WHO limit for sprinkler irrigation [Blumenthal et al., 2000].

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