Hazard/Risk Assessment

WORLDWIDE ESTIMATION OF RIVER CONCENTRATIONS OF ANY CHEMICAL ORIGINATING FROM SEWAGE-TREATMENT PLANTS USING DILUTION FACTORS

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Abstract: Dilution factors are a critical component in estimating concentrations of so-called "down-the-drain" chemicals (e.g., pharmaceuticals) in rivers. The present study estimated the temporal and spatial variability of dilution factors around the world using geographically referenced data sets at $0.5° \times 0.5°$ resolution. Domestic wastewater effluents were derived from national per capita domestic water use estimates and gridded population. Monthly and annual river flows were estimated by accumulating runoff estimates using topographically derived flow directions. National statistics, including the median and interquartile range, were generated to quantify dilution factors. Spatial variability of the dilution factor was found to be considerable; for example, there are 4 orders of magnitude in annual median dilution factor between Canada and Morocco. Temporal variability within a country can also be substantial; in India, there are up to 9 orders of magnitude between median monthly dilution factors. These national statistics provide a global picture of the temporal and spatial variability of dilution factors and, hence, of the potential exposure to down-the-drain chemicals. The present methodology has potential for a wide international community (including decision makers and pharmaceutical companies) to assess relative exposure to down-the-drain chemicals released by human pollution in rivers and, thus, target areas of potentially high risk. *Environ Toxicol Chem* 2014;33:447–452. © 2013 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals, Inc. on behalf of SETAC. This is an open access article under the terms of the Creative Commons Attribution-NonCommercial-NoDerivs License, which permits use and distribution in any medium, provided the original work is properly cited, the use is non-commercial, and no modifications or adaptations are made.

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INTRODUCTION

Over recent decades, scientists and regulators have had increasing concerns over the extent of the threat posed by chemicals discharged to water from domestic sources as opposed to industry or agriculture. Such chemicals include pharmaceuticals and personal care products, natural hormones (e.g., estrogens), and engineered nanoparticles (e.g., nanosilver, present in a variety of household products). Most of these substances enter freshwaters via wastewater disposal after consumer use; they are thus commonly referred to as "down-the-drain" chemicals. Scientists worldwide study the impact of the presence of such substances in freshwaters on the surrounding wildlife. For example, the magnitude of endocrine disruption in wild fish has been strongly related to steroid estrogen excretion from human population centers [1].

For most policy makers, regulators, and indeed the public, 2 important questions have arisen: What level of exposure is the aquatic wildlife subjected to, and what will be the impact in their country? The response of many countries to assess their particular situation, such as with endocrine disruption, has been to commission large-scale chemical and biological monitoring programs [1–3]. These exercises are very expensive and time-consuming. The answer they provide may come many years after the issue was first raised, and the results may be ambiguous.

There are many modeling approaches that could be applied to estimate predicted environmental concentrations of down-the-drain chemicals in rivers [4]. It was demonstrated that the temporal variability of down-the-drain chemical concentration in surface waters is driven mainly by the seasonal variability of river flows [5,6].

Some higher-tier models have been developed to better represent local geographic conditions and provide more accurate estimates of concentrations (e.g., Low Flows 2000 Water Quality eXtension model [7], QUAL2E [8], GREAT-ER [9]). Unfortunately, such models are rarely applicable at national or continental scales as a result of a lack of data. Many countries do not have measured or modeled hydrological data, such as river flows; additionally, location and size of sewage treatment plants (STPs) are often unavailable. It was recently demonstrated that despite such a lack of local data, there is sufficient global-scale data and information to estimate global threats to human water security and river biodiversity [10].

Thus, the dilution factor—the ratio between the volume of freshwater available and the domestic sewage discharge—can be used as a surrogate to compare risk levels caused by chemical exposure between and within countries [11]. We propose that a quantification of the national dilution factor for domestic effluent should be the first step in estimating the extent of the freshwaters that are at risk from domestically sourced chemicals. This factor would be relevant to assessing aquatic exposure to all down-the-drain chemicals.

In the present study, global grids of dilution factors at annual and monthly resolutions were generated using data sets of annual and monthly runoff and population. The present approach, similar to that of Vörösmaty et al. [10], builds on the method developed by Keller et al. [12] to estimate spatial variations of dilution factors at the global scale, using readily available $0.5° \times 0.5°$ gridded data ($\approx$55 km $\times$ 55 km at the equator) for the whole terrestrial land surface. The spatial and temporal variability of dilution factors within and between countries are then assessed using statistical measures such as the median. This approach was

designed to assess 1) differences between and within countries in terms of dilution of down-the-drain chemicals, 2) monthly variations within a country, and 3) suitability of providing a unique national dilution factor for a country.

MATERIALS AND METHODS

Modeling approach

The present method was designed specifically to assess the environmental level of exposure to down-the-drain chemicals in surface waters across the globe, even in countries where data are scarce. For a country, a crude estimate of the predicted environmental concentration in raw wastewater (PEC_{SEWAGE}; mg/L) of a chemical can be derived from daily per capita consumption (U; mg/cap/d)

$$PEC_{SEWAGE} = \frac{U}{W} \qquad (1)$$

where W is the daily per capita domestic water use (L/cap/d). Assuming no in-stream degradation and no background concentration, the river predicted environmental concentration (PEC_{RIVER}) immediately after mixing can then be defined as

$$PEC_{RIVER} = \frac{(1 - F) \times PEC_{SEWAGE}}{DF} \qquad (2)$$

where DF is the dilution factor and F is the fraction of chemical removed during wastewater treatment, which can be either measured or extrapolated from laboratory tests.

The dilution factor is defined as follows

$$DF = \frac{Q_r}{Q_{ww}} \qquad (3)$$

where Q_r (m^3/s) is the river flow at the outlet of the catchment and Q_{ww} (m^3/s) is the total domestic wastewater effluent generated within the catchment. Using gridded data, the river flow can be calculated from globally distributed runoff estimates (R; mm/yr). For a catchment with the outlet in cell i, the river flow in cell i (Q_{r_i}; m^3/s) is

$$Q_{r_i} = \sum_{j=1}^{i} \left(R_j \times \frac{1}{10^3} \times \frac{1}{31\,536\,000} \right) \times (A_j \times 10^6) \qquad (4)$$

where A is the cell area (km^2) and j is an index of all cells contributing to the catchment above cell i.

The total domestic wastewater effluent generated in a catchment is the total amount of water used for household purposes (e.g., food preparation, flushing toilets, bathing, and lawn watering) across the catchment. Using gridded data, for a catchment with the outlet in cell i, Q_{ww_i} (m^3/s) is estimated by combining population estimates (P) and national per capita domestic water use (W; m^3/cap/yr) estimates

$$Q_{ww_i} = \sum_{j=1}^{i} P_j \times W_j \times \left(\frac{1}{31\,536\,000} \right) \qquad (5)$$

For any grid cell i, the dilution factor (DF_i) is then

$$DF_i = \begin{cases} \sum_{j=1}^{i} \left(\dfrac{R_j \times A_j \times 10^3}{P_j \times W_j} \right) & \text{where} \quad \sum_{j=1}^{i} P_j > 0 \\ +\infty & \text{where} \quad \sum_{j=1}^{i} P_j = 0 \end{cases} \qquad (6)$$

The present approach assumes that all populations are connected to their nearby water courses, although sometimes a large proportion of a population may use septic tanks or a similar wastewater disposal in which the route to water is not direct. Such an assumption was made to provide a conservative estimate of the dilution factor; this estimate is likely to underestimate the dilution of chemicals discharged via humans and overestimate river predicted environmental concentration.

National summary statistics

The present approach may be applied at the catchment (within the limits of the grid size), national, or global scale using either annual or monthly runoff. Assessing the temporal and spatial variability in the dilution factor can address questions such as whether endocrine disruption in fish would be expected in only a few areas or be widespread across the nation. The median, mean, and a selection of percentiles—the interquartile ranges (25th and 75th percentiles), 5th, and 95th percentiles—were generated for each country; for each dilution factor map (annual and monthly), all cells within a country were identified and percentiles calculated across the selected grid cells. A percentile is the value of a variable below which a certain percentage of observations fall. The statistics within the present study were calculated using Microsoft Excel 2007.

While generating these statistics, the assumption was made that the dilution factor is only relevant where there is both river flow and wastewater effluent. Thus, for each country, these statistics were drawn only from cells where both the river flow and the total population in the upstream catchment were greater than 0. These statistics were calculated for both the annual and the monthly dilution factor grids.

Hydrological data

Runoff estimates can be derived using macro-scale hydrological models such as Macro-PDM [13], the variable infiltration capacity model [14], the water balance model [15], and Water-GAP2 [16,17]. A macro-scale model is a model that can be applied over a large geographic domain without calibration at the catchment scale [13]. Such models were applied over recent years to estimate present and future water resource availability at global, continental, and regional scales [18–20].

In the present study, the annual and monthly composite runoff field data sets produced by Fekete et al. [21] at a spatial resolution of $0.5° \times 0.5°$ were used. A climate-driven water mass balance was combined with observed river flow data to generate these long-term average runoff estimates. The water balance model uses climatologically averaged monthly air temperature and precipitation from an updated data set of Legates and Willmot climate fields [22,23]. Runoff is then predicted based on soil type and texture from the Food and Agriculture Organization/United Nations Educational, Scientific and Cultural Organization soil data bank [24], topographic data from the global elevation data set ETOPO5 [25], and a contemporary land-cover classification derived from overlaying cultivated areas from Olson's land-use classification [26] onto potential vegetation [27]. The water balance model is then combined with observed river flow data from the Global Runoff Data Centre [28] using the global Simulated Topological Network at 30-minute spatial resolution (STN-30p) [29]; the measured interstation runoff (difference between discharge downstream and discharge upstream), where available, was used to constrain the magnitude of the water balance modeled runoff. The time period of these river flow data varies for each gauging station; however, only those with at least 12 yr of records were

selected. Approximately 60% of the gauging stations selected had the common data period 1970 to 1980. In the present study, the STN-30p was used to accumulate the annual and monthly runoff to estimate cell-specific river flow Q_{r_i}.

Keller and Rees [30] assessed the goodness of fit of the flows derived from this approach using observed flow values from 670 gauging stations of the Global Runoff Data Centre. The measured flow data used in this assessment were not the same as the observed river flow data originally used by Fekete et al. [21] to estimate the composite runoff fields. The composite runoff fields method is most successful at simulating flows across Asia, North and Central America, Europe, and the Near East, with a mean bias between 0% and 10%. The method tends to overestimate observed flows across Africa and South America, with mean bias of 53% and 37%, respectively. For Australia and the Pacific, however, the method underestimates river flows (mean bias of –41%).

Population data

Population estimates are available from the SocioEconomic Data and Applications Centre. The Gridded Population of the World v.3 data set [31] is derived from population data issued by national bodies, such as national statistics offices, to generate gridded estimates of population, which are then adjusted to match United Nation totals. Here, the projected estimates for 2005 at $0.5° \times 0.5°$ resolution were used; these 2005 estimates made use of census data up to 2004.

The dilution factors for each grid cell were calculated by applying Equation 6. It was therefore important that the population and runoff coverages matched adequately. An apparent mismatch occurs mostly along the coastlines, where the population coverage is wider in places, by 1 cell at the most, than the runoff coverage. The total population within the Gridded Population of the World v.3 data set is approximately equal to 6.4×10^9 inhabitants; however, only 6.1×10^9 inhabitants overlapped with the runoff coverage. The discrepancy in population is approximately equal to 6% across the world, and similar discrepencies are observed at the national level (e.g., 3% for India and 5% for Egypt). Most statistics considered in the present study (median and 25th and 75th percentiles) are not sensitive to extreme values; thus, the impact of such a discrepency on the selected national dilution factor statistics was assumed to be negligible. However, the local dilution factor values along the coastlines must be handled with care as they might be underestimated (some of the coastal population discharge sewage effluent directly into the sea rather than into their local river; however, because of a lack of data, it was assumed that all coastal population discharges to rivers) or overestimated (as a consequence of the underestimated population [≈6%] when overlapping the runoff and the population coverage, the coastal population potentially discharging to rivers may be underestimated; this would result in underestimating the total domestic wastewater effluent and therefore overestimating the dilution factor [Equations 3 and 6]).

Domestic water use

In the present study, domestic water use was used as a proxy for wastewater discharge (Equation 1). There are significant variations in the amount of water used for domestic purposes between and within countries; these reflect differences in water availability as well as infrastructure, wealth, and habits. Although water-use data per country and per sector is among the most desired data in terms of water resources, the uncertainty within these data are often considerable. Furthermore, these data

are often estimated rather than measured, with varying methods across data sources [32]. These data should therefore also be handled with care.

Four main data sources for national per capita domestic water use were used to build a global data set: Gleick [32], Food and Agriculture Organization [33], World Resource Institute [34], and Organisation for Economic Co-operation and Development [35]. Where discrepancies arose, only the data for the year 2000 or later were retained, and from these the lowest estimate was selected to provide a higher pollution scenario as it maximizes predicted environmental concentration in raw wastewater (Equation 1). The resulting set of mean values of national per capita domestic water use (W) was mapped at the country level, which was then disaggregated at a $0.5° \times 0.5°$ resolution to produce gridded values of domestic water use across the globe (tabulated values in Supplemental Data, Table S1). It should be noted that the variability in stated per capita water consumption can be significant; for approximately 34% of countries with available data, there was at least a factor 2 between data sources, with 11 countries having more than a factor 5 between the lowest and the highest values.

At present, because of a lack of data, a single national average domestic water-use figure was used. However, in some countries, differences in domestic water use between urban and rural areas could be significant as a result of social and cultural factors such as household size, distance to a well, wealth, and education [36,37]. The implementation of such differences might increase the dilution factor within urban areas and decrease it in rural areas.

RESULTS AND DISCUSSION

Differences in dilution factors between nations

Predicted national annual median dilution factors vary across the globe (Figure 1; tabulated values in Supplemental Data, Table S2). The local dilution factors are derived using Equation 6 (map of locally predicted annual dilution factors in Supplemental Data, Figure S1); thus, low dilution factors may result from low runoff, high population density, or a combination of both. In engineering practice, a ratio of river flow to raw wastewater flow of 40 is recommended to prevent risk [38]. This ratio was used as a benchmark in the present study to visualize the levels of risk.

Across the globe, the differences in national annual dilution factors (and hence chemical concentrations) are extreme; for example, there are nearly 4 orders of magnitude between the annual median dilution factors in Canada ($\approx 33\,500$) and Morocco (≈ 5). Most countries with the lowest median dilution factors (<10) are in North Africa and the Middle East. These areas tend to correspond to very arid regions (e.g., the Sahara) with few rivers. Population density can also play a significant role, as is the case in Belgium, where the annual dilution factor is among the lowest across the globe. Belgium is one of the countries in Europe with the highest population density: 343 cap/km^2 in 2005 according to the United Nations Department of Economic and Social Affairs [39]. Countries where low national dilution factors result in large part from high population densities include India, Belgium, the United Kingdom, and the Republic of Korea.

Most countries with high dilution factors are in North and South America, northern Europe, northern and eastern Asia, and Australia. The annual dilution factor for Australia seems unexpectedly high. Across Australia, only 10% of the cells have a river flow; thus, the dilution factor is not calculated in
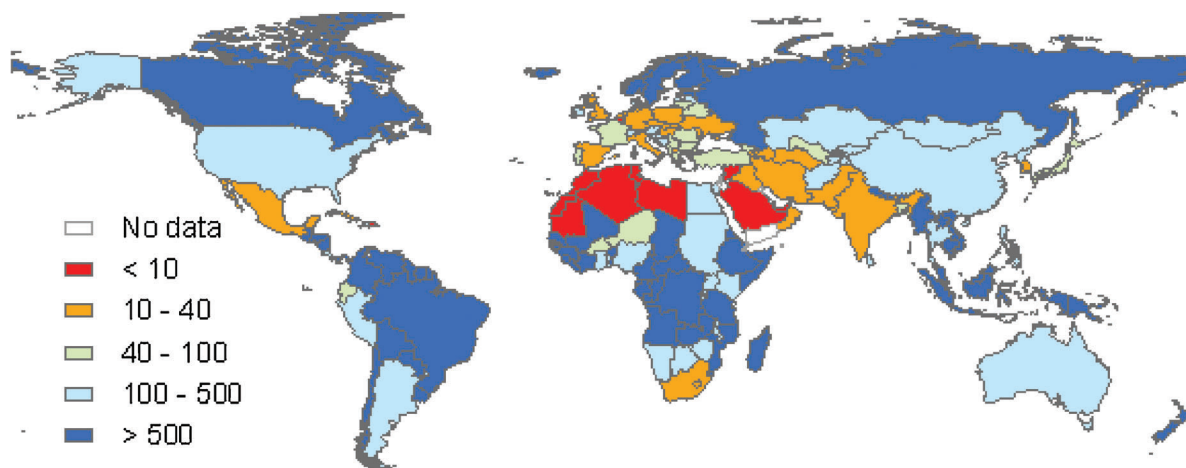
Figure 1. Predicted values of national annual median dilution factors. The median is a median across grid cells meeting inclusion criteria.

many parts of the country, in particular where population densities are relatively high and a low dilution factor might be expected. The national dilution factor for Australia, and many other countries, would therefore be much lower if the restriction on flow values was relaxed and cells with flow values of 0 m³/s were included in the calculation of the national statistics. A different type of problem is that associated with small countries as a result of the coarseness of the spatial resolution. At 0.5° resolution with an average cell size of 2250 km², higher uncertainties arise when estimating runoff and, thus, river flow in catchments smaller than 25 000 km² [29]. Although the concept of the dilution factor calculation remains valid when looking at smaller basins and therefore smaller countries such as Japan and the United Kingdom [40], a higher grid resolution would be more appropriate to reduce uncertainties in flow estimates [11].

Within-nation variability in dilution factors

National median values of annual dilution factor may be a useful starting point for comparison between countries of their potential exposure to down-the-drain chemicals. But the often unique spatial and temporal variabilities of dilution factor within a country must also be quantified because they can provide vital detail. The spatial variability of the annual dilution factor for

some countries (tabulated values for all countries in Supplemental Data, Table S2) is captured in a box-and-whisker plot (Figure 2).

Significant differences occur within a country. There often is 1 order of magnitude between the 25th and the 75th percentiles (e.g., Australia, Cambodia, France); however, there are 3 orders of magnitude in Venezuela (≈330 and 141 680), and some countries have 2 orders of magnitude (e.g., United States, United Kingdom, Mexico, Argentina, Russia). These differences in dilution values essentially have 2 components: seasonal flow variations and demographic variations. These differences in dilution factors can reflect the variety of climates within a country (e.g., United States, from desert in Arizona to humid subtropical in Florida) or differences in population density (e.g., United Kingdom, low density in the highlands of Scotland, high density in southeast England). The mean dilution value can be particularly misleading; this is because of the possible wide range of dilution factor values within a country with an average often skewed by the highest values. This is clearly the case for Canada, the United States, Venezuela, the United Kingdom, and India (Figure 2). These important national differences emphasize the limitations of using a single value of annual dilution factor when assessing chemical exposure levels within a country.
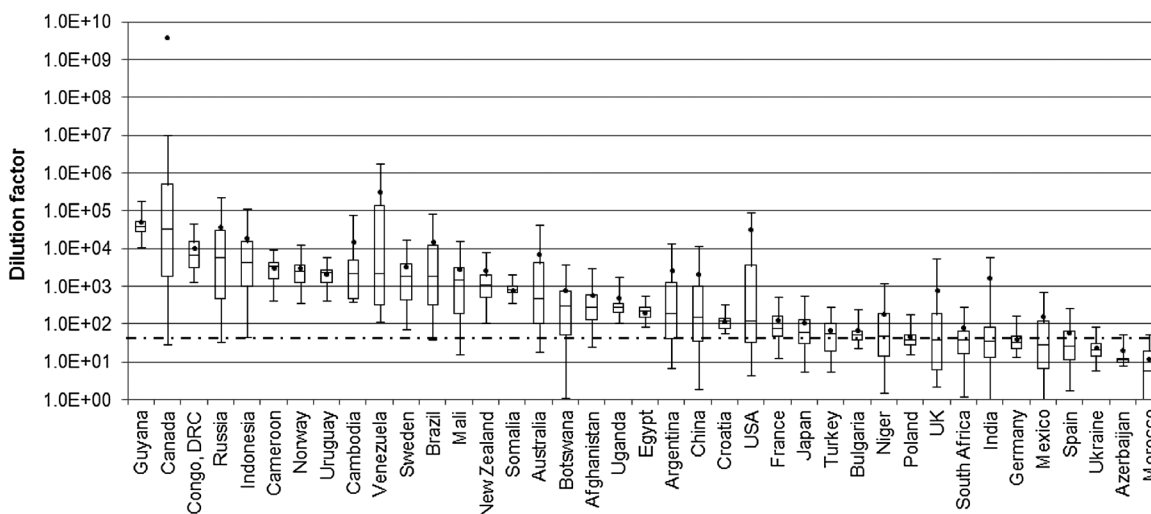

Figure 2. Spatial variability of the annual dilution factor between and within some countries. In each box-and-whisker plot, the box boundaries are the 25th and 75th percentiles, the line inside the box is the median, the whiskers are the 5th and 95th percentiles (where bottom whisker is missing, 0 < 5th percentile <1), and the dot is the mean. The dotted line represents a ratio of river flow to wastewater flow of 40, as recommended in engineering practice [38].
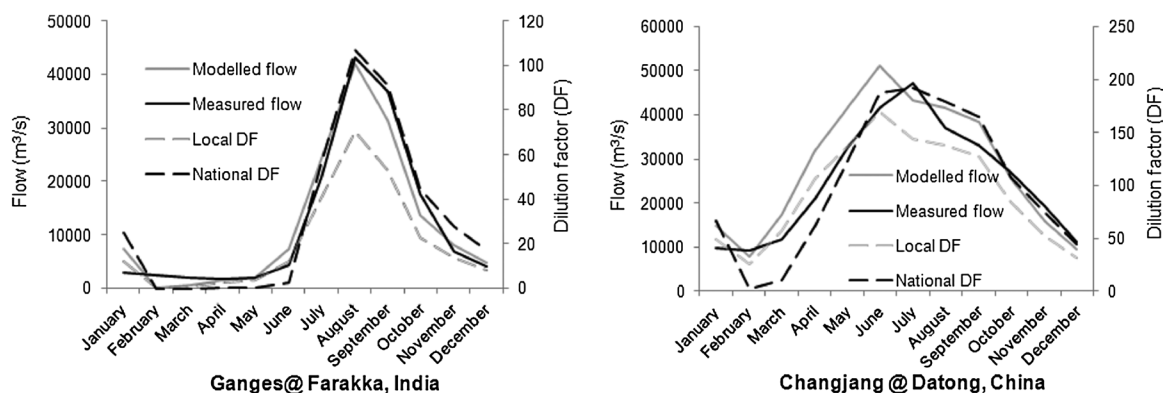
Figure 3. Temporal variability of mean monthly flow (measured and modeled at the gauging station) and median monthly dilution factor at the catchment and country scale for the river Ganges at Farakka in India (longitude = 87°92′, latitude = 24°83′) and the river Changjang at Datong in China (longitude = 117°61′, latitude = 30°76′). DF = dilution factor.

Temporal (seasonal) variation can in some countries be the greatest source of variation in the dilution factor. Across the globe, the maximum monthly difference in median dilution factor can vary between 0 and 7 orders of magnitude in a country. Most countries with the highest order of magnitude (>7) between minimum and maximum monthly national dilution factors lie between the Tropic of Cancer and the Equator (e.g., Mali, Chad, Ethiopia, Thailand, and India). These countries tend to be in arid climates (northern Africa), where rivers can dry up, or tropical climates with dry winters (e.g., Thailand, India, and Cambodia), where monsoons can transform a small stream into a huge river. In contrast, most countries in temperate regions (e.g., France, United Kingdom, United States, and Paraguay) have the lowest temporal variability (between 0 and 2 orders of magnitude). Countries with a polar climate tend to have 2 to 4 orders of magnitude between the minimum and maximum monthly dilution factors; the temporal variation in these countries is a consequence of snow melt.

An example of this type of variability where countries/ regions have a climate which can change from drought to torrential rains each year can be illustrated by flow regimes in the Ganges River (India) and the Changjang River (China) (Figure 3). The differences in flow, and hence dilution, between winter/spring and late summer/autumn is dramatic, changing more than 100-fold.

With this method, the temporal variability of the dilution factor is a reflection of the monthly variability in river flow as the population and per capita water use are assumed to be constant throughout the year. For these 2 large catchments, the modeled flows are in reasonable agreement with the observed flows. However, for the Changjang River, although the shape of the modeled hydrograph is reasonably similar, there is a temporal shift of about 1 mo. Such discrepancies are inevitable using global-scale hydrological models, but the essential observation is that the model has got the amount of flow available for dilution right in this case.

Using dilution factors

As an example, we can estimate the range of estrone (estrogenic female hormone) concentrations in rivers in Morocco and Canada using these dilution factor values. We will assume an excretion rate of 3.3 μg/d [7] and a daily per capita domestic water use for Morocco and Canada of, respectively, 96 L and 787 L [32,33] to estimate PEC_{SEWAGE} (Equation 1). No removal during wastewater treatment ($F = 0$, in Equation 2) is assumed, to provide a worst-case estimate.

Applying Equation 1, the PEC_{RIVER} for Morocco is approximately equal to 5220 ng/L, 6 ng/L, and 1 ng/L when using, respectively, the 5th percentile, 50th percentile, and 95th percentile annual dilution factor (Supplemental Data, Table S2). For Canada, PEC_{RIVER} is approximately equal to 0.15 ng/L when using the 5th percentile annual dilution factor and less than 1 pg/L for both the median and the 95th percentile.

CONCLUSIONS

The principle of using available dilution as an indicator of potential national exposure to down-the-drain chemicals from the human population is a reasonable one. The method described in the present study is not claimed to be either unique or exclusive. There is no reason that other approaches to calculating flow and quantifying the local human population to provide dilution factors could not be equally, if not more, effective. We do not believe, however, that different methods would come to an entirely different conclusion. The present study hopefully can illustrate to a wide audience that the exposure to down-the-drain chemicals, and hence the experience of local aquatic wildlife, will be extraordinarily different between nations across the globe. Given the dramatic range of dilution factors, we would argue these are far more important determinants of chemical concentration in rivers than other variations in fate and behavior. These dilution factor values provide the means to estimate the possible range of in-river concentrations for any down-the-drain chemical where a per capita excretion, consumption, or even production value is available.

Despite a relatively crude approach, the present method revealed significant differences between nations with regard to the dilution they offer to down-the-drain chemicals. For the median annual dilution factor, there can be up to 4 orders of magnitude between countries and up to 3 orders of magnitude within a country. Temporal variability is also significant: within a country, the maximum monthly difference in dilution can vary between 0 and 9 orders of magnitude. Because of the great variability in dilution factors within many countries, comparing nations on the basis of a single dilution factor could be misleading.

Within every nation, depending on the location and time of year, there will be hot spots of chemical exposure; however, the present set of statistics helps define how widespread the issue could be. The present methodology provides a means of assessing where and when levels of exposure from down-the-drain chemicals might be of concern and, therefore, finer-resolution data and/or models applied or measurements taken.

It is hoped that the present approach might prove useful both to scientists and to regulators of nation-states, particularly in developing countries where few data sets exist, as a guide to potential exposures and, hence, risks from chemicals. The present methodology and data set may be combined with data such as consumption and degradation rates and simple water-quality principles to predict concentrations in rivers for individual down-the-drain chemicals. The present approach may be valuable to chemical companies as they consider new markets for their products. It also provides the basis for implementing future climate scenarios and therefore the means to assess the possible impact of climate change on dilution factors across the globe.

SUPPLEMENTAL DATA

Tables S1 and S2.
Figure S1. (80 KB PDF).

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