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Quantifying the Extent of Anthropogenic Eutrophication of Lakes at a National Scale in New Zealand

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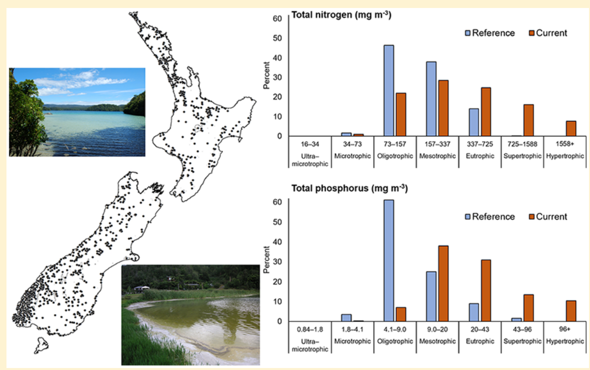
Supporting Information

ABSTRACT: Quantifying environmental changes relative to ecosystem reference conditions (baseline or natural states) can inform assessment of anthropogenic impacts and the development of restoration objectives and targets. We developed statistical models to predict current and reference concentrations of total nitrogen (TN) and total phosphorus (TP) in surface waters for a nationally representative sample of ≥1033 New Zealand lakes. The lake-specific nutrient concentrations reflected variation in factors including anthropogenic nutrient loads, hydrology, geology, elevation, climate, and lake and catchment morphology. Changes between reference and current concentrations were expressed to quantify the magnitude of anthropogenic eutrophication. Overall, there was a clear increase in lake trophic status, with the most common trophic status being oligotrophic under a reference state and mesotrophic under current conditions. The magnitude of departure from reference state varied considerably within the sample; however, on average, the mean TN concentration approximately doubled between reference and current states, whereas the mean TP concentration increased approximately 4-fold. This study quantified the extent of water quality degradation across lake types at a national scale, thereby informing ecological restoration objectives and the potential to reduce anthropogenic nutrient loads, while also providing a modeling framework that can be applied to lakes elsewhere.



INTRODUCTION

Developing policies to protect and restore ecosystems requires understanding the state and range of natural conditions.[1] The term "natural" can be subjective, particularly regarding the extent to which humans are considered part of an ecosystem.[2,3] Nonetheless, in the field of aquatic ecosystem management, the concept of reference conditions that correspond to the absence of human disturbance is fundamental to assessing the natural state of the environment and, more broadly, ecological integrity.[4,5] The extent to which an environmental attribute (e.g., the concentration of a nutrient that limits primary production in a waterbody) differs from a defined reference state provides a measure of the degree of anthropogenic change, although it should be recognized that environmental conditions fluctuate naturally, and the relationship between physicochemical variables and the ecological status of freshwater ecosystems is often nonlinear.[6] Estimates of reference conditions can therefore be used to inform freshwater management policies and the concept has been widely applied, including in the U.S. and Europe.[4,7]

Although it will often be neither desirable nor feasible to designate a reference state as an objective of aquatic ecosystem management,[3,7,8] quantifying such conditions provides a "line in the sand" against which current conditions and targets can be benchmarked. Characterizing reference conditions can therefore avoid the "shifting baseline" phenomenon,[9,10] which refers to society's erroneous and progressively worsening perceptions of what constitues a pristine ecosystem state, resulting in continuous ecological decline as management targets shift. Management objectives can then be established that meet the criteria of being both obtainable and protective.[11] More broadly,

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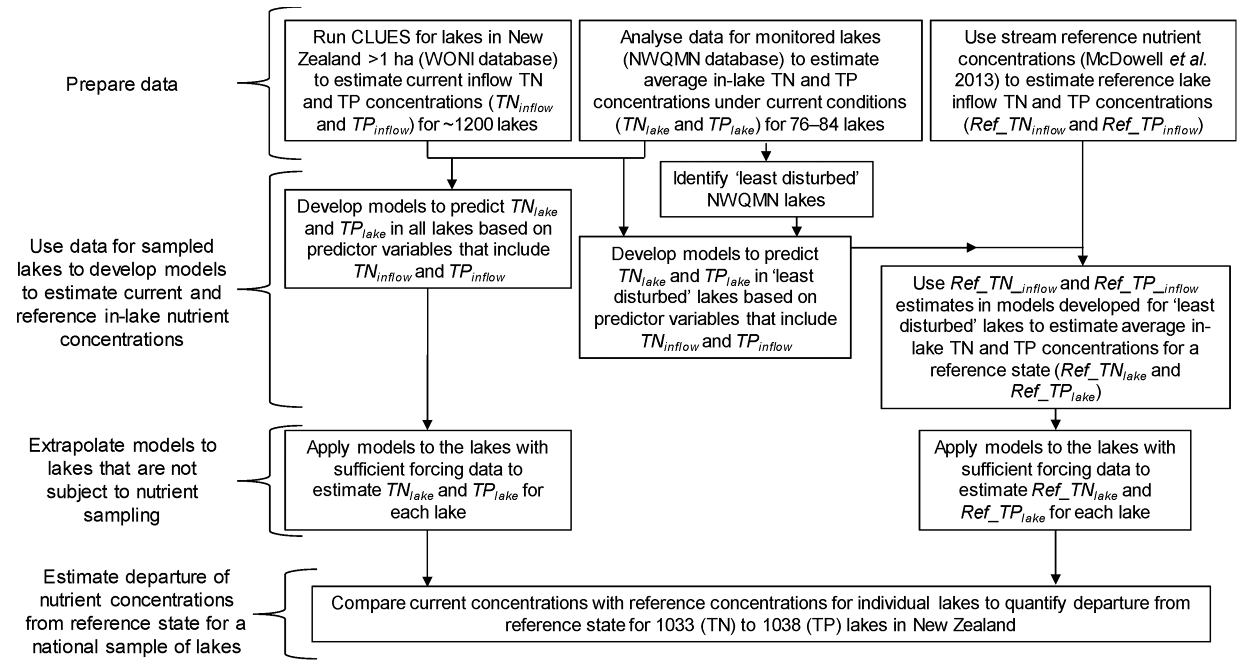

Figure 1. Summary diagram of the methods used to derive lake−specific reference total nitrogen (TN) and total phosphorus (TP) concentrations. See text for further details, including definition of acronyms.

comparison between estimated reference concentrations and current measurements helps managers to understand broad temporal trends in water quality, which can provide context for evaluating changes detected over shorter time scales.[12−14] Further, characterizing departure from reference states has wider value than only guiding the work of environmental managers; instead, it can promote awareness among the public of what a natural ecosystem "looks like" and the nature of anthropogenic changes that have occurred to natural resources.[9]

In freshwaters, eutrophication due to increased nitrogen (N) and phosphorus (P) concentrations is one of the foremost threats to ecological integrity.[12,15] Departure of nutrient concentrations from reference conditions therefore provides an indication of the ecological quality of an aquatic ecosystem. Even in the absence of human disturbance, nutrient concentrations in lakes vary widely due to factors such as catchment geology, lake morphometry, residence time, and climate and mixing regimes; these differences need to be considered when deriving reference nutrient concentrations.[16] To account for this natural variability, it is preferable to initially derive reference concentrations that are specific to individual lakes, although there may be scope to then use these lake-specific concentrations to quantify reference concentrations for broader categories of lakes.

A range of methods has been used to identify reference N and P concentrations for lakes and the European Commission[17] recommends that one or more of the following approaches be used, listed in order of preference:

i. analysis of data obtained from existing pristine sites,
ii. analysis of historical data relating to sites prior to disturbance,
iii. modeling,
iv. expert judgment.

The small number of remaining pristine sites precludes the use of option i for many regions, for example, most of lowland Europe,[6] as well as coastal areas in many developed regions (e.g., ref 18), which are at the downstream end of catchments and are therefore subject to pressures from human development upstream. Similarly, lack of historical data for most lakes constrains the use of option ii. This constraint can be partly overcome by applying paleolimnological techniques to analyze sediment cores and provide insights into historical nutrient status (e.g., ref 19); however, these require detailed field, laboratory and analytical work. Also, several factors can confound the application of paleolimnological methods for assessing historical nutrient concentrations, including unquantified effects from variables such as alkalinity, and a lack of consistent relationships (transfer functions) between diatom communities and nutrient concentrations,[20] particularly if the nutrient of interest does not consistently limit diatom biomass and deposition rates.[21] In the case of option iii, the most widely used approach for establishing reference conditions has been landscape-context statistical models that use spatially resolved data to derive statistical models to predict nutrient concentrations based on variables such as geology, soil type and proxies for human nutrient sources such as catchment area in agricultural land.[16] Reference concentrations may then be hindcast by setting the influence of human pressure variables to zero. The technique requires a large lake sample size; for example, in the U.S., Dodds et al.[22] used multiple regression analysis for 220 lakes at the regional scale (Kansas), whereas Herlihy et al.[23] used a similar approach for 1028 lakes at a national scale. The challenges of this approach relate to the ability to encompass a sufficiently large number of monitored lakes corresponding to the range of landscape characteristics necessary to robustly configure statistical models. Furthermore, the approach does not explicitly account for variability caused by in-lake processes (e.g., different rates of sedimentation), which can significantly affect the accuracy of predictions.[24,25]

The objective of this study was to quantify how current nutrient concentrations have changed from a reference state in a nationally representative sample of lakes in New Zealand (NZ). We consider "reference state" to be conditions prior to the

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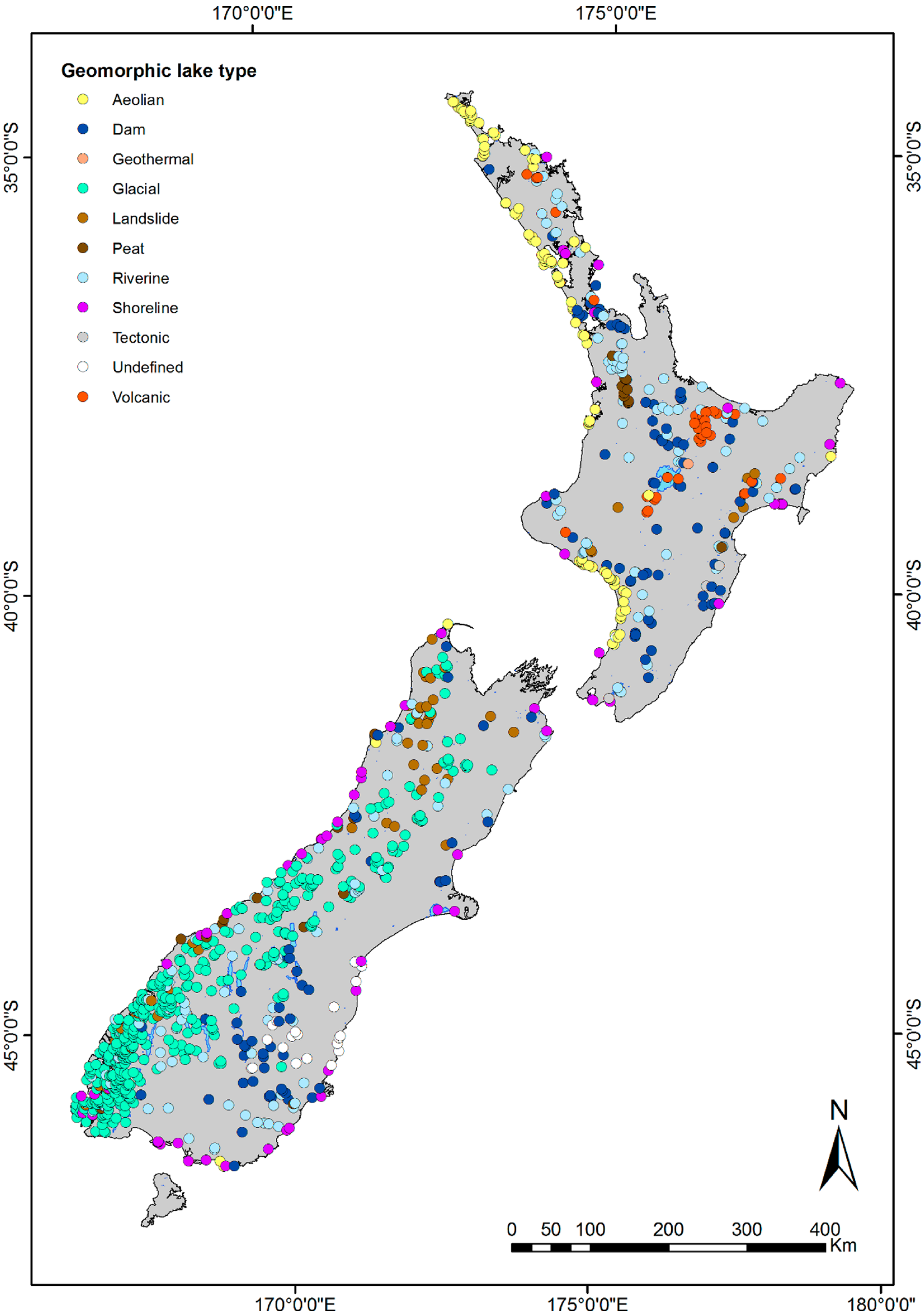

Figure 2. Locations and geomorphic type of the 1038 lakes for which changes in nutrient concentrations from a reference state were estimated.

initiation of widespread land development by European settlers (predominantly for agricultural purposes) that occurred in the mid-1800s,[26] recognizing that nutrient pollution associated with land use intensification in more recent decades has been a major pressure to water quality.[27] A study by McDowell et al.[28] established median reference concentrations for analytes in streams and rivers across NZ, accounting for climate, topography and geology. This work provided an opportunity to derive catchment-specific reference nutrient loads that can be used to predict lake-specific reference nutrient concentrations.

To achieve the study objective, we developed a novel approach that used parsimonious nutrient mass loading models to estimate lake nutrient concentrations. The models were applied using lake−specific estimates of nutrient concentrations in lake inflows that corresponded to reference and current states, output from a national−scale hydrological model that was used

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Table 1. Summary of Sub-Samples of the National Water Quality Monitoring Network Dataset (Post 2000) That Were Used to Develop Models to Predict Current in-Lake TN (TN_{lake}) and TP (TP_{lake}) Concentrations

sample	number of lakes	number of samples per lake (mean in parentheses)	minimum of mean concentrations (mg m^{-3})	maximum of mean concentrations (mg m^{-3})	mean of mean concentrations (mg m^{-3})	mean coefficient of variation
TN_{lake}	76	10−261 (64)	39	3407	662	47%
TP_{lake}	84	10−261 (62)	5.2	735.6	75.9	86%

to estimate lake inflow rate, and information about lake morphometry. The outcomes of the modeling were separate estimates of nutrient concentrations corresponding to reference and current states for a national sample of ≥1033 lakes. These concentrations were specific to individual lakes and reflected variability in landscape factors and dominant in-lake nutrient cycling processes. Modeled reference and current concentrations were then compared to quantify the extent that contemporary (post 2000) lake nutrient concentrations have departed from a reference state at a national scale.

■ MATERIALS AND METHODS

Overview. An overview of the methods used to estimate departure from reference state of mean total nitrogen (TN) and total phosphorus (TP) concentrations in nationally representative samples of 1038 (TN) and 1033 (TP) lakes is provided in Figure 1.

Methods are described below; in brief, we analyzed a National Water Quality Monitoring Network (NWQMN) database that included nutrient concentrations for lakes measured during 2000−2014. Based on this, we identified training data sets (n = 76−84) to develop statistical models to predict current TN and TP concentrations in lakes using variables that included flow-weighted TN and TP concentrations in lake inflows estimated using a catchment model (CLUES). Models to estimate reference in-lake nutrient concentrations were then developed by analyzing subsets of NWQMN lakes that were considered least disturbed by human pressures, in conjunction with estimates of flow-weighted TN and TP concentrations in lake inflows corresponding to a reference state (derived by McDowell et al.[28]). The models were then used to estimate current and reference nutrient concentrations in a larger and nationally representative sample of lakes included in the Waters of National Importance (WONI) database. This step provided current and reference TN and TP concentrations in ≥1033 lakes in NZ (Figure 2) that were compared to evaluate how nutrient concentrations in NZ lakes have departed from a reference state.

Current in-Lake Nutrient Concentrations (n = 76−84). Mean current total nitrogen (TN_{lake}) and total phosphorus (TP_{lake}) concentrations in the surface waters of the sample of NZ lakes were obtained from the NWQMN database compiled by NZ regional government agencies. Data were provided via the Land, Air, Water Aotearoa (LAWA) partnership and were typically collected at monthly frequency. Detailed sampling and analytical protocols are described in Burns et al.,[29] which require samples to be collected from the surface or within the epilimnion in stratified lakes (see Davies-Colley et al.[30] for a discussion of differences in lake sampling depths among monitoring agencies). Measurements in the NWQMN database (1976−2014) collected prior to 2000 were removed from the analysis and the data set was then screened to create two separate subsamples (Table 1) that contained only lakes with 10 or more measurements of TN or TP. These screening decisions reflected a compromise between an aim to ensure that data were representative of current lake water quality and an aim to maximize the sample size. The data set was then further screened to ensure that the subsamples (Table 1) contained only lakes with (1) valid water residence time (τ_w) estimates (years) derived from the WONI database (discussed below), and (2) valid estimates of inflow nutrient concentrations derived from a catchment model (discussed below). The screening yielded separate subsamples of 76 (TN) and 84 (TP) lakes (Table 1). Current in-lake annual mean TN and TP concentrations (TN_{lake} and TP_{lake}) were estimated for lakes in these subsamples by calculating the arithmetic mean of concentrations.

Current Lake Inflow Nutrient Concentrations. Geographic Information System software (ArcMap 10.0; ESRI) was used to identify all lake surface inflows included in the 2010 NZ River Environment Classification (REC). The REC is a classification scheme that includes all streams and rivers in NZ at a 1:50 000 mapping scale.[31,32] Mean annual discharge was retrieved from the REC for all river sections upstream of each lake.

A catchment model (the Catchment Land Use for Environmental Sustainability Modeling Framework; "CLUES")[33] was used to estimate median TN and TP concentrations in all mapped lake inflows. CLUES is national-scale and has been used to satisfactorily predict TN and TP concentrations for streams that drain a wide range of landscapes across NZ (see Supporting Information (SI) for background information about the CLUES modeling framework, including details of model performance). Median TN and TP concentrations in lake inflows were then combined with mean annual discharge estimates for each inflow to estimate annual loads. Discharge-weighted estimates of current median lake inflow nutrient concentrations in mg m^{-3} (TN_{in} and TP_{in}) were then estimated for each lake by calculating volumetric averages, based on loads to each lake. Median values were deemed preferable to other statistics (e.g., arithmetic means) to characterize typical conditions because they minimize bias associated with extreme conditions, for example, floods.

CLUES was used to estimate TN_{in} and TP_{in} for lakes in the NWQMN database with TN_{lake} and TP_{lake} estimates. These TN_{in} and TP_{in} estimates were evaluated to identify potential data quality issues; this included analyzing ratios of inflow concentration to in-lake concentration, based on the assumption that outlier values of these ratios may reflect erroneous inflow concentrations. Five lakes were omitted from the final NWQMN samples and adjustments were made to the inflow concentrations assigned to three lakes (see SI for details of these data review steps). Omitted lakes are not included in the final samples summarized in Table 1. CLUES was also used to estimate TN_{in} and TP_{in} for all other lakes on mainland NZ in the larger WONI database (described below).

Waters of National Importance (WONI) sample (n = 1038). The national WONI database was analyzed to obtain a large and nationally representative sample of lakes. The WONI database comprises spatial, land use and physical data for 3820 lakes in NZ that include lakes that have been mapped with area >1 ha.[34,35] The majority of these lakes are not monitored and all estimates of morphometric variables, including τ_w, are based on

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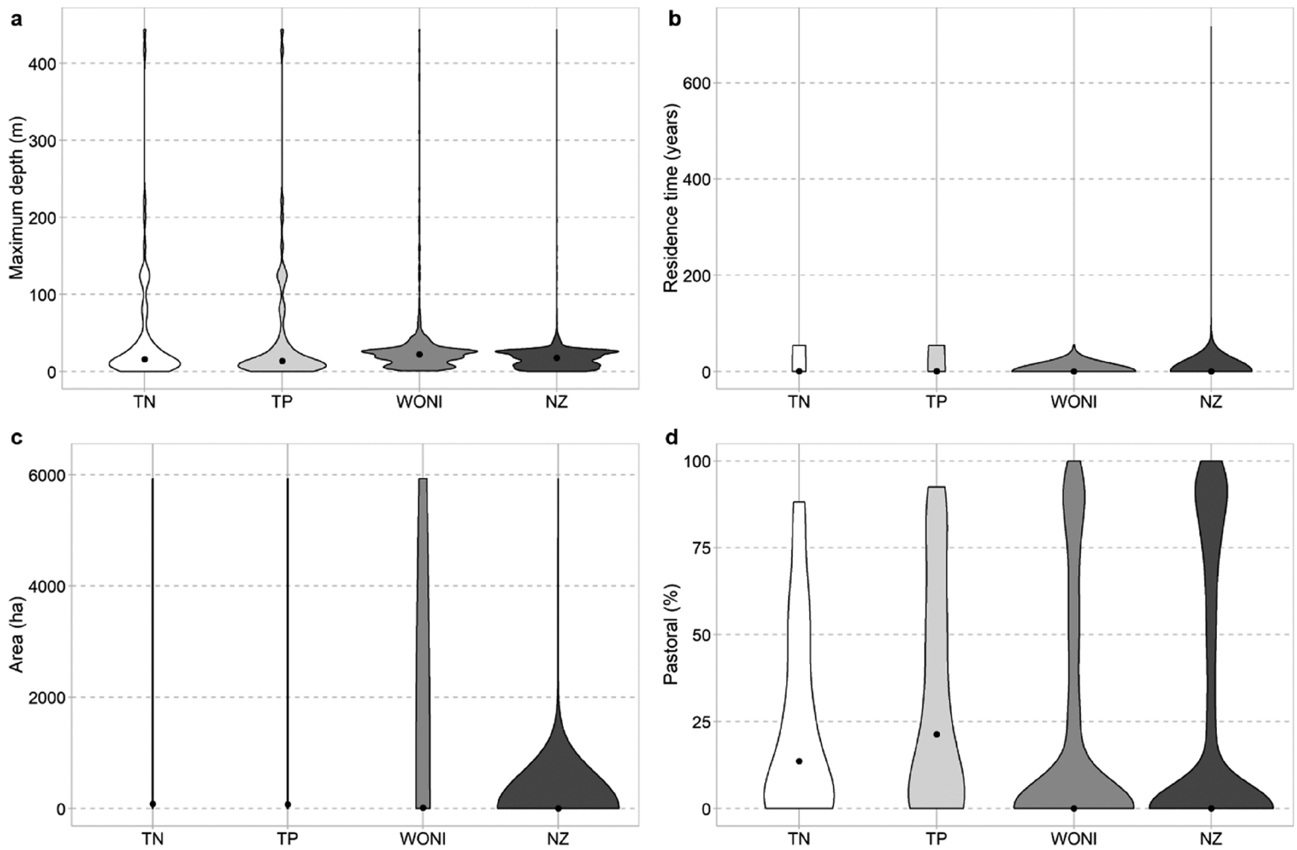

Figure 3. Violin plots of the relative frequency (kernel probability density) of (a) maximum depth, (b) annual mean residence time, (c) surface area, and (d) proportion of pastoral land use in surface catchments for lake groups. Plots relate to the "training" samples used to develop models to predict TN ($n = 76$) and TP ($n = 84$) concentrations; the Waters of National Importance (WONI) sample ($n = 1038$) of lakes for which reference and current nutrient concentrations were predicted in this study, and; the complete WONI sample of all 3820 mapped lakes >1 ha in NZ. Black circles denote median values.

modeling.[35] The WONI database was filtered to select only lakes that had mapped inflows (and therefore inflow nutrient concentrations could be estimated using CLUES; see above) and also had assigned values of predictor variables used to develop models (e.g., τ_w; see below). This yielded a WONI sample of 1038 lakes. The screening criteria were likely biased toward omitting the smallest lakes and seepage lakes that lacked surface inflows.

Morphometric characteristics of lakes included in the NWQMN samples (n = 76−84) used for model fitting (Table 1) were broadly representative of the larger WONI sample (n = 1038), although large (area > ~5 km^2) and also shallow (maximum depth < ~10 m) lakes were somewhat over-represented in the NWQMN samples (Figure 3). The distribution of surface area within the NWQMN samples (Figure 3c) was strongly influenced by the inclusion of Lake Taupō, which is the largest lake in NZ (613 km^2).

The distribution of lakes among different geomorphic types (obtained from the FENZ database[35]) was also similar between the WONI sample and the samples used for model fitting, although the proportions of aeolian (dune) lakes and volcanic lakes were lower in the WONI sample than the samples used for model fitting (Table 2). Glacial lakes were under-represented in the samples used for model fitting, relative to the frequency of this lake geomorphic type nationally (Table 2).

Table 2. Proportion of Lakes by Geomorphic Type in Samples of Lakes

geomorphic type	TN sample (n = 76)	TP sample (n = 84)	WONI sample (n = 1038)[a]	NZ (n = 3820)[a]
shoreline	5%	5%	6%	8%
aeolian	30%	29%	11%	9%
riverine	12%	17%	16%	21%
dam	4%	4%	12%	14%
volcanic	17%	14%	4%	3%
peat	1%	6%	2%	3%
glacial	28%	24%	44%	39%
landslide	3%	2%	5%	2%
geothermal	0%	0%	<0.2%	<0.2%
tectonic	0%	0%	<0.3%	<0.3%

[a]21 lakes (2%) in the WONI sample and 339 lakes (9%) in the NZ national data set had an undefined type; these were omitted from the analysis presented in this table.

Reference Lake Inflow Nutrient Concentrations. Nutrient concentrations in lake inflows that correspond to a reference state were calculated using median stream reference nutrient concentrations estimated by McDowell et al.[28] (SI Table S1). The authors used data collected from >1000 stream sites across NZ during 1989−2009 to develop models for different REC classes.[28] Specifically, the authors used mixed-effects models with random slopes and intercepts, and with a smoothing spline, to model the relationship between the log-transformed median values of each analyte and the proportion of intensively−farmed land in a catchment. Based on these models, the authors calculated reference concentrations that corre-

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sponded to concentrations under a scenario of no intensively–farmed land. This approach reflected the well–established dominant influence of intensive agriculture on nutrient concentrations in NZ streams;[36−38] for example, Julian et al.[38] showed that the Spearman rank correlation coefficient between cattle density and nutrient concentrations in a national sample of 77 NZ rivers was 0.85 for TN and 0.72 for TP.

Median stream reference TN and TP concentrations (and associated estimates of standard error) for classes of different climate by topography by geological combinations (SI Table S1) were assigned to all lake inflows based on the REC classification of the inflow river section. Due to sample size constraints, median reference stream concentrations were not available for all the REC classes represented by lake inflows, so values for similar classes or the hierarchical parent climate by topography REC class were substituted for those that had no estimated reference concentration (SI Table S2). For each lake, reference inflow nutrient loads were summed and then divided by the estimated total inflow discharge to calculate discharge–weighted estimates of median reference inflow TN (Ref_TN_{in}) and TP (Ref_TP_{in}) concentrations in mg m^{-3}. Uncertainty in these estimates due to propagation of uncertainty in the median reference nutrient concentrations for each lake inflow was accounted for using a bootstrapping routine (described below).

Empirical Nutrient Mass Loading Model: Current Nutrient Concentrations. The NWQMN data sets (Table 1) were used to develop models to predict TN_{lake} and TP_{lake} for lakes in the WONI sample. Candidate models were identified based on reviewing literature, and the final models used to predict current nutrient concentrations were then selected based on model performance. We focused on trialling various empirical nutrient loading models ("box models") that have a theoretical basis, yet include a small number of parameters, and are suited to our broad-scale application involving a large number of lakes with limited data. The models have narrow temporal and spatial constraints (i.e., they predict lake-wide annual mean nutrient concentration for the mixed layer or mixed water column condition) and assume equilibrium of nutrient fluxes (i.e., steady state conditions). Nonetheless, when assumptions are met, these models typically yield estimates of mean nutrient concentrations that are comparable to predictions made with more sophisticated models.[39]

The models stem from the work of Vollenweider[40] who showed that TP_{lake} may be estimated as

$$TP_{lake} = \frac{L}{z_{mean}(\rho + \sigma)} \tag{1}$$

where L is areal TP loading rate (mg TP m^{-2} year^{-1}), z_{mean} is mean depth (m), ρ is lake flushing rate (year^{-1}), and σ is a first order rate coefficient for TP loss from the lake (year^{-1}). This model has since been widely adapted and numerous authors (including R.A Vollenweider) have developed Vollenweider's model to replace L with TP_{in}. Four of these models were trialled; these are listed as Models 1 to 4 below and they are based on the expectation that the relationship between in-lake and inflow nutrient concentrations is modulated by residence time in years (τ_W). Model 4 was determined to be the best-performing model in a comparative study of multiple models by Brett and Benjamin.[25] Models 5 and 6 are linear regression models that were initially included for comparative purposes; these models comprise predictor variables included in Models 1−4, although they have weaker theoretical underpinnings. These six models were also trialled to predict TN_{lake}, with TN_{in} replaced for TP_{in} in the equations. We used z_{max} rather than z_{mean} in all models because this is the depth metric provided in the WONI data set. Piecewise versions of the models were trialled to investigate whether the model performance could be improved by refitting the models to subsets of the data based on z_{max} or geomorphic type, with the boundaries for this analysis guided by limnological theory (i.e., we strived to avoid "data dredging").

Model 1: (based on Vollenweider[24]) − Parameters: $\beta_0 \beta_1$

$$TP_{lake} = \beta_0 + \beta_1 \frac{TP_{in}}{1 + \sqrt{\tau_w}} \tag{2}$$

Model 2: (based on Brett and Benjamin[25]) − V is a fitted parameter that represents particle settling velocity

$$TP_{lake} = \frac{TP_{in}}{1 + \left(\frac{V\tau_w}{z_{max}}\right)} \tag{3}$$

Model 3: (based on Jones and Bachmann[41]) − Parameters: c_1, c_2

$$TP_{lake} = \frac{c_1 TP_{in}}{1 + c_2 \tau_w} \tag{4}$$

Model 4: (based on Brett and Benjamin[25]) − Parameters: k_1, k_2

$$TP_{lake} = \frac{TP_{in}}{1 + k_1 \tau_w^{k_2}} \tag{5}$$

Model 5: Linear regression model - Parameters: β_0, β_1, β_2, β_3

$$TP_{lake} = \beta_0 + \beta_1 TP_{in} + \beta_2 \tau_w + \beta_3 TP_{in} \tau_w \tag{6}$$

Model 6: Linear regression model - Parameters: β_4, β_5, β_6

$$TP_{lake} = \beta_4 + \beta_5 TP_{in} + \beta_6 z_{max} \tag{7}$$

Similar to Brett and Benjamin,[25] the following criteria were used to compare model performance: standard error of the estimate, sum of squared errors, r^2, Akaike's Information Criterion (AIC),[42] and the root-mean-square error (RMSE) of a 10-fold cross validation. This RMSE metric provides a good measure of the sensitivity of the model to the input data, thereby helping to identify the most robust predictive model. r^2 was calculated as the square of the Pearson's correlation coefficient between predicted and observed values; we recognize that r^2 has limitations if used to directly quantify goodness of fit for a nonlinear model. Bias was examined by visually inspecting observed and predicted values plotted alongside a 1:1 line. Variables were $\log_{10}$-transformed, which was identified as necessary to improve compliance with model assumptions. AIC (a measure of model fit and parsimony) and the RMSE metric (a measure of model error) were used as the primary metrics to select the optimum model. All analyses and data manipulation were conducted using the program R.[43] Nonlinear least-squares model validation was undertaken using nlstools.[44]

Detailed results of model evaluation and selection are shown in the SI, which includes performance statistics for all models that were trialled and diagnostic plots of residual values. The optimum model to predict $\log_{10}TP_{lake}$ in mg m^{-3} was

$$\log_{10}TP_{lake} = \frac{\log_{10}TP_{in}}{1 + (k_1 + \Delta k_1 d)\tau_w^{k_2}} \tag{8}$$

where k_1, Δk_1, and k_2 are fitted coefficients with values of 0, 0.44, and 0.13 respectively, and d = 0 for shallow lakes and d = 1 for deep lakes. This model (eq 8) is a piecewise version of Model 4 (eq 5) that permits the relationship between inflow and in-lake

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nutrients to vary between shallow and deep lakes. This difference has a strong theoretical basis as nutrient cycling processes that occur in shallow lakes (e.g., internal loading due to sediment resuspension) are less dominant in deep lakes.[45] A z_{max} threshold of ≤7.5 m was used to differentiate shallow lakes from deep lakes in eq 8. This threshold was identified by trialling z_{max} values between 0 and 20.0 m and comparing model performance statistics (see SI for results). r^2 was 0.69 and RMSE was 0.33 mg m^{-3} in $\log_{10}$ transformed units for this model (see SI for further details of performance). For shallow lakes, eq 8 predicts that TP_{lake} is approximately equal to TP_{inflow} as the fitted value of k_1 for shallow lakes is 0. Therefore, this result indicates that negligible net in-lake attenuation of TP occurs in the shallow lakes in the sample. In deep lakes, eq 8 predicts that increasing τ_W results in greater attenuation of TP_{inflow} in the lakes in the sample, consistent with theory.[24]

The optimum model to predict $\log_{10}TN_{lake}$ in mg m^{-3} was Model 5 (eq 6). When fitted parameter values are included, this is written as

$$\log_{10} TN_{lake} = 1.60 + 0.54\log_{10} TN_{in} - 0.41\log_{10} z_{max} \tag{9}$$

r^2 was 0.78 and RMSE was 0.24 mg m^{-3} in $\log_{10}$ transformed units (see SI for further details of performance) for this model. When the dependent variable is untransformed, eq 9 can be written as

$$TN_{lake} = 39.8\frac{TN_{in}^{0.54}}{z_{max}^{0.41}} \tag{10}$$

Thus, τ_W was not used to predict TN_{lake} since z_{max} was a markedly better predictor of TN_{lake} than τ_W. This indicates that, relative to P, particle settling (assumed to be inversely correlated with τ_W) exerts a minor role in attenuating inputs of N in deep NZ lakes. We recognize that eq 10 has a weaker theoretical basis than the box models that include τ_w as a predictor variable; however, selection of eq 10 to predict TN_{lake} in unmonitored lakes is supported by its superior performance.

Empirical Nutrient Mass Loading Model: Reference Nutrient Concentrations. Separate models were developed to predict in-lake annual mean TN and TP concentrations that correspond to a reference state (REF_TN_{lake} and REF_TP_{lake}). These models were developed using the same process described above for the TN_{lake} and TP_{lake} models, except the models were fitted to subsamples of lakes that were considered "least disturbed" (criteria described below). This was necessary because a proportion of the lakes in the NWQMN sample have undergone a substantial degree of cultural eutrophication. Consequently, nutrient cycling processes in these lakes are expected to have substantially changed from a reference state; for example, associated with increased anoxia-driven internal loading,[45] increased rates of denitrification and N fixation,[46] or occurrence of nutrient resuspension by introduced benthivorous fish.[47] As such, it was deemed inappropriate to use models fitted to current conditions in such lakes to hindcast reference conditions.

Selecting the "least disturbed" sample involved striking a balance between selecting only those lakes that were expected to be least departed from a reference state, while also ensuring that a sufficiently large sample size was selected to develop robust models. Following data exploration, the "least disturbed" samples were selected by choosing subsamples from the screened NWQMN sample that had assigned TN_{inflow} (for REF_TN_{lake} model fitting) or TP_{inflow} (for REF_TP_{lake} model fitting) estimates that were equal to or less than the maximum median stream reference concentrations developed by McDowell et al.[28] (see SI). That is, subsamples of lakes were selected for which TN_{inflow} < 362 mg m^{-3} or TP_{inflow} < 35 mg m^{-3}. Intuitively, this screening criterion seemed reasonable; that is, we only selected lakes that had inflow nutrient concentrations that were equal to or less than the maximum concentrations estimated to occur in NZ's streams under a reference state. Note though that the inflow nutrient concentrations assigned to these lakes were not necessarily less than the reference stream concentrations that corresponded to the respective third level REC classes in each lake catchment; this more restrictive filter would have resulted in an excessively small sample size. This approach yielded "least disturbed" samples to use for model fitting of 30 lakes for REF_TN_{lake} and 27 lakes for REF_TP_{lake}. These samples included lakes distributed throughout NZ that encompassed a wide range of morphometric characteristics (see SI for details of all lakes included in the least disturbed samples). The least disturbed samples included most of the main lake geomorphic types (Table 2), although no peat or shoreline lakes were included, while it was notable that the proportion of glacial lakes in the samples (56% in both samples) was higher than the proportion of lakes in NZ generally that are of glacial origin (39%; Table 2).

Following model evaluation (see SI), we selected the same models (albeit with different fitted parameter values) to estimate reference nutrient concentrations as those selected to estimate current nutrient concentrations. The optimum model to predict $\log_{10}$ REF_TN_{lake} in mg m^{-3} was

$$\log_{10} TN_{lake} = 0.89 + 0.81\log_{10} REF\_TN_{in} - 0.33\log_{10} z_{max} \tag{11}$$

r^2 was 0.55 and RMSE was 0.26 mg m^{-3} in $\log_{10}$ transformed units for this model (see SI for further details of performance).

The optimum model to predict $\log_{10}$ REF_TP_{lake} in mg m^{-3} was

$$\log_{10} REF\_TP_{lake} = \frac{\log_{10} REF\_TP_{in}}{1 + 0.27\cdot\tau_w^{0.29}} \tag{12}$$

r^2 was 0.20 and RMSE was 0.28 mg m^{-3} in $\log_{10}$ transformed units for this model (see SI for further details of performance). Note that, unlike eq 8, eq 12 is not piecewise; that is, the model fit does not vary between shallow and deep lakes. This is because there was deemed to be an insufficient sample size to robustly parametrize separate models; for example, there were only five lakes in the TP "least disturbed" sample with z_{max} < 7.5 m and seven lakes with z_{max} < 10.0 m. Residual analysis supported this approach as it did not show a systematic bias in the residuals (see SI). This is intuitive as differences between shallow and deep lakes in the magnitude of nutrient cycling processes may be expected to be less pronounced under a reference state; for example, due to lower fluxes of sediment-bound nutrient resuspension in shallow lakes under a reference scenario with lower fine sediment loads.

Estimating Uncertainty and Deriving Model Predictions. The optimum models were used to estimate TN_{lake} for 1038 lakes and TP_{lake} for 1033 lakes in NZ. This sample comprised the WONI sample (n = 3820) minus lakes without assigned τ_W estimates or modeled inflow concentrations (TN_{inflow} or TP_{inflow}). There were five fewer lakes in the TP_{lake}

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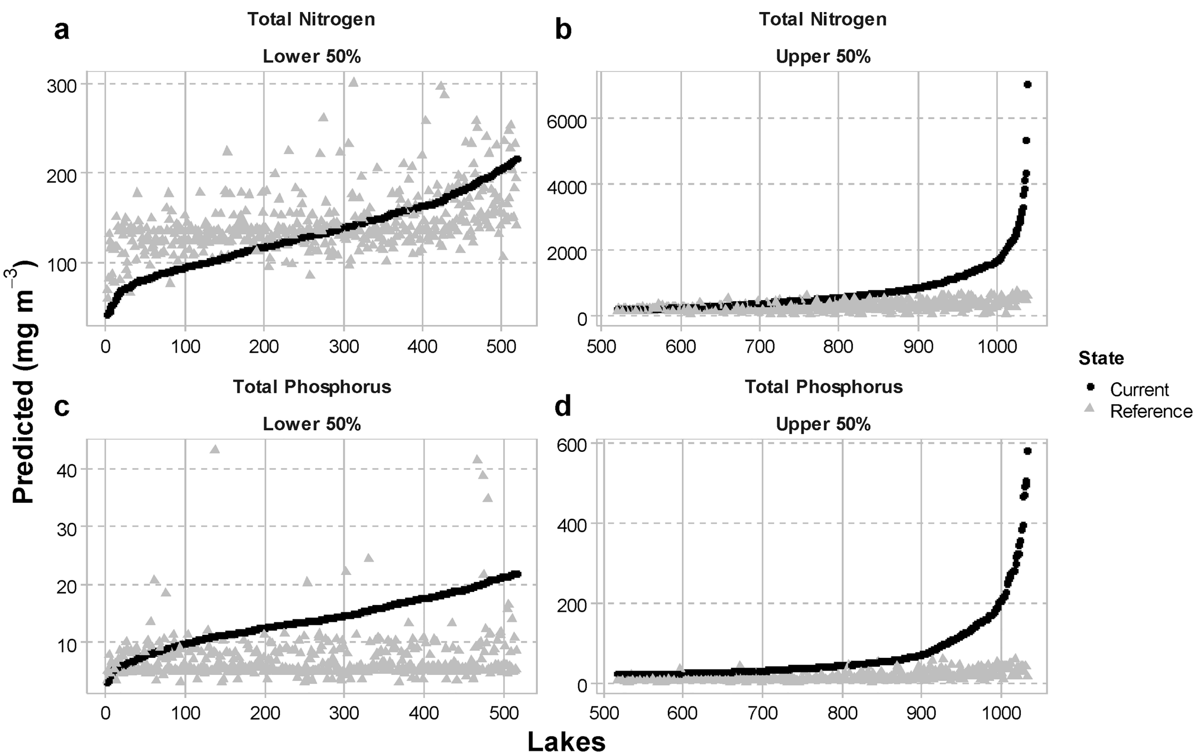

Figure 4. Predicted average current and reference total nitrogen (n = 1038; panels a and b) and total phosphorus (n = 1033; panels c and d) concentrations in lakes throughout NZ. For each variable, separate plots with different scales are presented to show predicted values less than (lower 50%) and greater than (upper 50%) the median value. Confidence intervals are omitted for clarity (see SI).

Table 3. Summary of Estimated Nutrient Concentrations in New Zealand Lakes Corresponding to Reference and Current States

		statistic			
variable	state	n	median	mean	standard deviation
total nitrogen (TN; mg m^{-3})	reference	1038	164	213	122
	current	1038	217	449	598
total phosphorus (TP; mg m^{-3})	reference	1033	7	10	9
	current	1033	22	43	64
TN:TP (mass)	reference	1033	24.0	24.3	9.1
	current	1033	10.4	13.9	13.9
change from reference state (TN)		1038	23%	90%	207%
change from reference state (TP)		1033	213%	292%	299%

sample because TP_{inflow} values for these lakes were identified as outliers (see SI).

To account for log-transformation bias, a correction factor (CF) was used to convert $\log_{10}$-transformed dependent variables back to a linear scale:[48]

$$CF = 10^{\log_e(10)\hat{\sigma}^2/2} \tag{13}$$

where σ is the standard error of the estimate.

Confidence intervals were estimated for model predictions using a two-stage parametric bootstrap with 1000 iterations. In stage 1, the model was refitted to new data sampled from a normal distribution centered around the fitted values with standard error of the estimate σ.[49] In stage 2, predictions were made by resampling inflow nutrient concentrations (TN_{inflow} or TP_{inflow}) from a normal distribution centered around the predicted inflow concentration and the standard error (SE_{inflow}). Standard error of the inflow nutrient concentration was 33% of the untransformed estimate, which was deemed reasonable based on a review of CLUES performance.[33,50] As the model was fit to Log_{10}Inflow nutrient concentration (Log_{10}Inflow), we used the Delta Method[51] to calculate the standard error:

$$SE\_Log_{10}Inflow = \sqrt{(Log_{10}Inflow \times \log_e 10)^{-2} SE_{inflow}^2} \tag{14}$$

where SE_Log_{10}Inflow is the standard error of Log_{10}Inflow.

Thus, for each iteration of the bootstrap, the prediction varied based on error in the model fit (assumed to include structural error that reflects simplifications of in-lake processes) and error in inflow concentrations (assumed to comprise the majority of the "errors in variables"). The 2.5% and 97.5% quantiles of the bootstrapped values were then calculated to estimate 95% confidence intervals. For modeled reference concentrations, confidence intervals were estimated using consistent methods. Error in the REF_TN_{inflow} and REF_TP_{inflow} values was incorporated in the same way using the standard error equal to 33% of the estimated reference concentration, which was the average of the standard errors calculated by McDowell et al.[28]

The NZ−specific Trophic Level Index[52] (TLI; similar to the more widely used Trophic State Index[53] (TSI)) was used to

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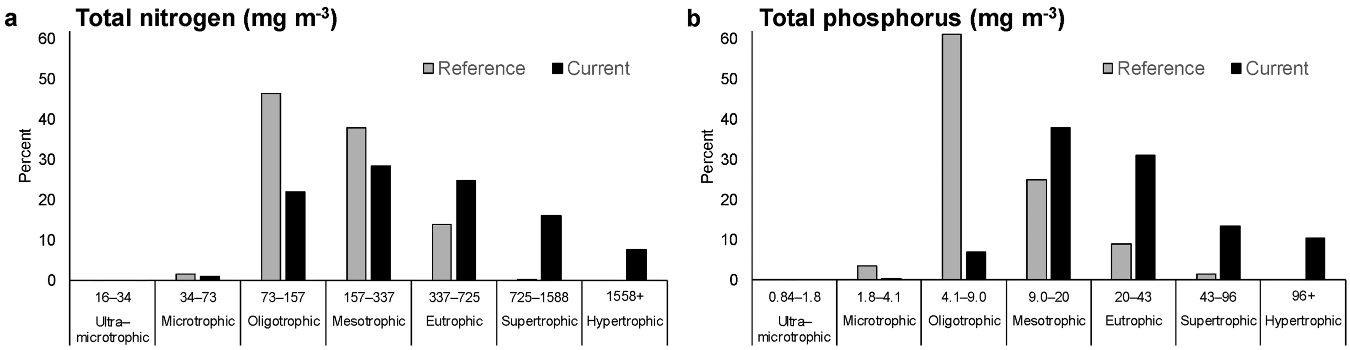

Figure 5. Comparison of the relative frequency (%) of lake trophic states (presented separately based on TN and TP concentrations) between a reference scenario and current conditions for 1033 (TP) to 1038 (TN) New Zealand lakes. Trophic state classifications and corresponding concentrations relate to the Trophic Level Index.[52]

relate REF_TN_{lake} and REF_TP_{lake} values to the TP- and TN-specific trophic state categories (note that the TLI includes TN whereas the TSI does not). Information about lake geomorphic type was obtained from the FENZ database.[35]

RESULTS

For our sample of ≥1033 NZ lakes, modeled average nutrient concentrations corresponding to current conditions were generally higher than concentrations corresponding to a reference state (Figure 4; Table 3). The mean of modeled TN_{lake} concentrations was 449 mg m^{-3} (median = 217 mg m^{-3}), which was approximately twice as high as the mean of REF_TN_{lake} concentrations (mean = 213 mg m^{-3}; median = 164 mg m^{-3}). The mean of modeled TP_{lake} concentrations (43 mg m^{-3}) was approximately 4-fold higher than the mean of REF_TP_{lake} concentrations (10 mg m^{-3}; Table 3), whereas there was an approximate 3-fold difference in the median values (22 mg m^{-3} vs 7 mg m^{-3}; Table 3).

For individual lakes, the mean difference between TN_{lake} and REF_TN_{lake} equated to an increase from a reference state to a current state of 90% (median = 23%; Table 3), although considerably larger increases were estimated for many lakes (see right-hand tail in Figure 4b), with average TN concentrations estimated to have at least doubled in approximately 25% of lakes. For TP concentrations, the relative change from a reference state was generally higher, with a mean increase in lake-specific average TP concentration of 292% (median = 213%) and an increase of ≥399% (approximately 5-fold or greater increase) in 25% of lakes (Figure 4c,d). In a minority of lakes, modeled average nutrient concentrations were lower for a current state than for a reference state (see left-hand tails in Figure 4a and c); this was the case for TN concentrations in 32% lakes and TP concentrations in 4% of lakes. However, the lower bound of the 95% confidence interval for the reference concentration did not exceed the upper bound of the 95% confidence interval for the current concentrations in any lake for either TN or TP (see SI). The higher relative change for TP concentrations meant that, on average, the estimated mass ratio of average in-lake TN concentrations to average in-lake TP concentrations (TN:TP) was lower under a current state than a reference state (average values of 13.9 and 24.3 respectively; Table 3).

For both TN and TP, there was a clear shift (Chi−square test, $p < 0.0001$) in the relative frequencies of lake trophic state toward higher trophic status under current conditions (Figure 5). Values of REF_TN_{lake} and REF_TP_{lake} corresponded to the microtrophic to supertrophic range of trophic state,[52] although only a small number of lakes were assigned to the supertrophic category (0.1% of lakes based on REF_TN_{lake} or 1.5% of lakes based on REF_TP_{lake}; Figure 5). Under a reference state, the most common trophic status was oligotrophic (46% of lakes based on REF_TN_{lake} or 61% of lakes based on REF_TP_{lake}; Figure 5). Values of TN_{lake} and TP_{lake} corresponded to the microtrophic to hypertrophic range of trophic state and the most common trophic status under current conditions was mesotrophic (38% of lakes based on REF_TN_{lake} or 25% of lakes based on REF_TP_{lake}; Figure 5). Under a reference state, approximately 10−15% of lakes (depending on the nutrient being considered) were estimated to have a trophic status of eutrophic or higher, whereas approximately 50−55% of lakes under current conditions were estimated to have a trophic status of eutrophic or higher.

Results varied among lake geomorphic classes (Figure 6). In general (e.g., based on median values for each class), reference concentrations were highest for dune, peat, riverine and shoreline lakes; intermediate for volcanic and dam-formed lakes, and; lowest for glacial and landslide lakes (Figure 6a,b). Variability in current concentrations was generally consistent with this pattern (Figure 6a,b), with median departure from reference state greatest in peat lakes and lowest in glacial and landslide lakes (Figure 6c).

DISCUSSION

Estimating reference (natural baseline) concentrations of N and P allowed us to quantify departure from reference conditions for a nationally representative sample of lakes. This showed that the trophic state of lakes in NZ has increased across a wide range of lake types. Such degradation in NZ lakes is symptomatic of the wider extent to which human activity has transformed nutrient cycling in catchments and receiving freshwater ecosystems generally.[46,54,55]

Our results are consistent with trend analysis for NZ rivers, which has highlighted general increasing trends in river nutrient concentrations in recent decades,[36,37] with agricultural intensification as a main cause.[27,56] However, our results contrast with results of Bachmann et al.,[19] who used a paleolimnological approach to examine departure from reference state for TN and TP concentrations in 240 lakes in the U.S. The authors found no evidence of a general increase in concentrations, instead concluding that TP concentrations had actually decreased overall by 14% since European settlement. Such a difference in results is somewhat surprising, given that extensive European settlement (and associated intensive land

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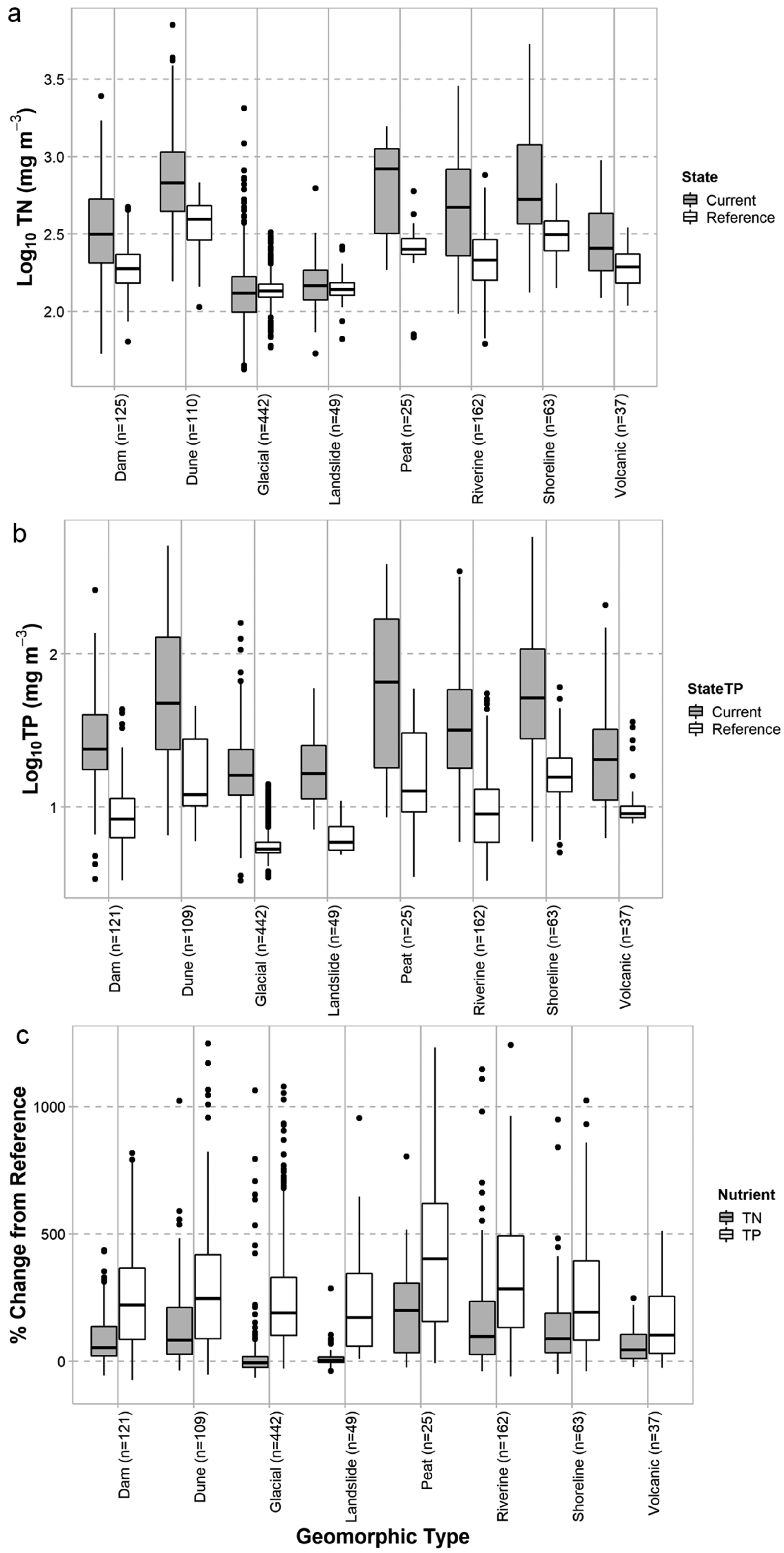

Figure 6. Standard box and whisker plots showing variability among lake geomorphic classes in (a) reference and current total nitrogen (TN) concentrations; (b) reference and current total phosphorus (TP) concentrations, and (c) lake-specific change in nutrient concentrations between reference and current states. Sample sizes are ∼1000; samples do not included lakes with an "undefined" class and there are small differences in sample sizes between nutrient variables.

management/use practices) occurred more recently in NZ than in the U.S.,[26] although the U.S. lakes that were studied may have been subject to less human disturbance than is representative of lakes there generally,[57,58] and general departure from reference state has been demonstrated in shallow lakes at a regional scale in the U.S.[22] The difference between the results of Bachmann et

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al.[19] for the U.S. and our results for NZ at least partly reflects the greater area of intensively managed pastoral land in NZ, which covers approximately one third of the country and is a strong determinant of elevated nutrient concentrations in lakes.[59] Overall, the general trend of anthropogenic eutrophication shown here for NZ lakes aligns with Europe, where there is consensus that nutrient concentrations in lakes are generally significantly higher compared to a reference state in the majority of lake types[6,60] (with boreal lakes in Sweden an exception[61]).

Care is necessary when considering the lake-specific reference concentration estimates developed here. When developing lake management policy, estimates need to be interpreted in the context of local expert knowledge about site-specific factors that may influence how natural and anthropogenic factors affect nutrient concentrations. Similarly, diligence is required if reference concentrations for groups of lakes (e.g., categories based on geomorphic type) are used as benchmarks for individual lakes, as lake-specific information first needs to be considered to determine whether a particular category is representative. Nonetheless, the systematic differences in reference concentrations among different geomorphic types of lakes (Figure 6) suggest that geomorphic type provides a useful variable to group lakes and derive benchmarks for similar lakes that were not included in our sample. The differences in reference concentrations among geomorphic types are intuitive: dune, shoreline and peat lakes had the highest median reference concentrations and are typically shallow and lie in relatively fertile low elevation areas. Further, lakes in these three classes exhibited the greatest median departure from reference state, reflecting that these lakes are frequently at the downstream end of catchments and are located in agricultural areas.[18,62] By contrast, glacial lakes (lowest reference concentrations) tend to be deep and have long residence times (thus high settling losses) and be situated in high-elevation catchments in the South Island (Figure 2) that have soils with low fertility. The general position of glacial lakes at the upstream ends of catchments in undeveloped areas also accounts for the low departure from reference state estimated for these lakes. Volcanic lakes, which are predominantly located in the central North Island (Figure 2), have reference concentrations that are intermediate relative to the other groups, although it is notable that reference TP concentrations (but not TN concentrations) were higher in volcanic lakes than in glacial lakes, which reflects the naturally high P concentrations in the underlying rhyolite-lithology and acidic volcanic soils (SI Table S1) that result in elevated baseline dissolved P concentrations in local streams.[28,63] The extent of departure was relatively low for TN concentrations in volcanic lakes, possibly reflecting that groundwater transit times are often long in volcanic lakes, meaning that measured lake TN concentrations do not fully reflect increases associated with land use intensification in recent decades.[64]

The finding that the TN:TP ratio was lower under a current state than a reference state (Table 3) is consistent with the general trend in eutrophication between reference and current states; TN:TP generally declines as trophic status increases,[65,66] as has been previously demonstrated for NZ lakes.[67] The causes of this shift are uncertain and could reflect changes in nutrient sources (e.g., increased inputs of P-rich domestic wastewater) and/or changes in nutrient cycling processes.[66] Interestingly, our finding that average increases between reference and current states were greater for in-lake TP concentrations than in-lake TN concentrations (SI Table S4) contrasted with those of Snelder et al.,[56] who showed that riverine N loads had increased to a greater extent than riverine P loads in most regions of NZ. This suggests that the stoichiometric shift with increasing eutrophication in NZ lakes reflects changes in in-lake biogeochemical processes, rather than changes in the nutrient composition of external loads. Key biogeochemical changes associated with eutrophication that might explain the shift to lower TN:TP are increased denitrification,[46] increased anoxia-driven sediment P release,[45] and, potentially, biotic interactions such as resuspension of P by invasive fish.[47] Despite this, we are cautious about using our results to make inferences about changes in nutrient cycling processes at the catchment scale as we recognize that precisely predicting nutrient stoichiometry in lakes is likely to be more challenging than predicting concentrations of individual nutrients,[68] whereas the predicted TN:TP ratios are highly sensitive to differences in the fitted parameters of the models used to predict each nutrient.

This study focused on the national scale and the methodology was predicated on numerous assumptions. Our analysis was conducted using a national sample (Figure 2) that included a diversity of lake types, although we acknowledge that the representativeness of some lake characteristics was biased in our sample (Figure 3, Table 2). In particular, our sample included lakes that were larger than the national average and therefore our results are most applicable to the status of larger lakes that typically have the greatest social and cultural values (and also receive the greatest monitoring effort). The approach considered only external nutrient loading from surface streams and therefore sources such as nutrient-enriched geothermal ground waters[63,64] or avifaunal subsidies[69] were not accounted for. The approach also assumed that the box models adequately reflect the range of in-lake processes; this assumption seems reasonable based on the good fit obtained for the best performing models, although the extent to which a lake can be well−conceptualised as a continuously−stirred mixed reactor will vary within the sample (see Higgins and Kim[70]). In addition, hydrology (modeled inflow discharge) was assumed to be unchanged between the reference and current scenarios, which is a simplification as, generally, land cover (and hence water yield) has been altered in the studied catchments since human colonisation. However, given that the box models depend on estimates of inflow concentration (not loads), significant error due to this assumption would not necessarily be introduced, providing any change in mean discharge was similar throughout a catchment. Finally, nutrient concentrations were estimated for approximately 120 reservoirs, yet it is debatable whether the concept of a "reference state" applies to man-made systems. We justify the inclusion of these waterbodies in the study as protective nutrient limits are necessary for reservoirs as well as natural lakes[22,23] and reference concentration estimates can therefore provide context to inform management of these systems, which have values beyond water storage.[71,72] Future studies could examine uncertainty in our reference concentration estimates by contrasting them with estimates derived using other methods, for example, paleolimnological studies in a subset of lakes.

Our approach ensured that the lake−specific reference concentrations reflect natural variations in factors that include hydrology, geology, elevation, climate, and, lake and catchment morphology. This study therefore addresses the recognized need for scientifically defensible and site-specific nutrient criteria to guide water resources management.[11,16] The lake−specific reference concentrations (Figure 4) provide a baseline that can help to set water quality restoration, ideally informed by

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other lines of evidence to account for uncertainties (sensu Schallenberg et al.[62]). These concentrations can guide management priorities based on the likely cost-effectiveness and speed of remediation.[73] In particular, the non−linearity (hysteresis) in relationships between nutrient loading and change in trophic state[10,74] means it is more efficient to identify existing high quality lakes and take prudent actions to safeguard current water quality, rather than reactively applying costly interventions at a later date.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b03120.

Additional methodological details, tables, and figures (PDF)

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Notes

The authors declare no competing financial interest.

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