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Is Sustainability Achievable? Exploring the Limits of Sustainability with Model Systems

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Successful implementation of sustainability ideas in ecosystem management requires a basic understanding of the often nonlinear and nonintuitive relationships among different dimensions of sustainability, particularly the system-wide implications of human actions. This basic understanding further includes a sense of the time scale of possible future events and the limits of what is and is not likely to be possible. With this understanding, systematic approaches can then be used to develop policy guidelines for the system. This article presents an illustration of these ideas by analyzing an integrated ecological-economic-social model, which comprises various ecological (natural) and domesticated compartments representing species along with a macroeconomic price setting model. The stable and qualitatively realistic model is used to analyze different relevant scenarios. Apart from highlighting complex relationships within the system, it identifies potentially unsustainable future developments such as increased human per capita consumption rates. Dynamic optimization is then used to develop time-dependent policy guidelines for the unsustainable scenarios using objective functions that aim to minimize fluctuations in the system's Fisher information. The results can help to identify effective policy parameters and highlight the tradeoff between natural and domesticated compartments while managing such integrated systems. The results should also qualitatively guide further investigations in the area of system level studies and policy development.

Introduction

The increasing intensity and scale of human activity across the globe, particularly after the industrial revolution, has led to severe depletion and deterioration of the Earth's natural resources (*1*). The realization that the existence of the human race is strongly dependent on the survival of these natural systems has mobilized efforts at the scientific, political and social levels, and sustainability has emerged as a new paradigm of analysis. Consequently, recent literature correlating the causes and their effects, predicting future consequences, and suggesting remedial actions is substantial (*2–7*).

Sustainability, conceptually defined by the Brundtland commission (*8*), exhibits many dimensions related to ecology, society, economics, technology, and other system aspects (*9–11*). The goal of a sustainable policy, then, is to promote the structure and operation of the human component of a system (society, economy, technology, etc.) in such a manner as to reinforce the persistence of the structures and operation of the natural component (i.e., the ecosystem) (*9*). This requires at least a basic understanding of the relationships between these different dimensions, often nonlinear and nonintuitive, with particular emphasis on understanding the implications of various human actions. This, allied with the focus on long-term effects, makes an integrated analysis based on systems theory critical for sustainable policy development, and this is the focus of this article.

Starting with an integrated ecological-economic model presented in Whitmore et al. (*12*), a stable and qualitatively realistic model representing Earth's ecosystem and some of its critical aspects is developed. The model is then used to conduct scenario studies, primarily to understand the complex relationships and to identify potential catastrophes. An approach based on dynamic optimization is then used to develop time dependent management policies for the previously identified unsustainable scenarios, e.g., increasing human population and per capita resource consumption, using information theory to formulate the appropriate optimization objectives.

Integrated Ecological-Economic Model. Several ecological-economic models with varying degree of emphasis on the economics have been proposed in literature. Environmental economists have often developed decision making models that ascribe a value to various environmental services, referred to as externalities (*13, 14*). Primarily ecological models with simplified economic considerations include Ludwig et al. (*15*), Brock and Xepapadeas (*16*), Costanza et al. (*17*), and Carpenter et al. (*18*). van den Bergh (*19*) presents a macroeconomic model that considers aspects of environment relevant for the economic processes such as resource extraction, waste assimilation, recycling, and pollution.

The model considered in this work, based on the integrated model proposed by Whitmore et al. (*12*), differs from earlier models in primarily three aspects: it addresses resource limits (the system is closed to mass and open to energy); it lays a legal foundation through the definition of property types; and it implements an explicit market system through a price-setting model for decision making (*12*). The integrated model, diagram shown in Figure 1, comprises 12 compartments, including two resource pools (RP and IRP, respectively, representing all biologically necessary resources and resources which are biologically inaccessible except through recycling by P2 and P3), three primary producers (Plants P1, P2, and, P3), three herbivores (H1, H2, and H3), two carnivores (C1 and C2), an industrial sector (IS) and humans (HH). It represents a compromise of allowing sufficient complexity to express some of the emergent behavior of a similar real system without being mathematically overly complicated. The system mass flows throughout are specified by the model, and individual compartments observe conservation of mass. The macroeconomic price setting model assumes a single period planning horizon and governs the dynamics (decisions) of the human households (HH), the industrial sector (IS) and the two private firms: a producer of plants (P1) and a producer of herbivores (H1).

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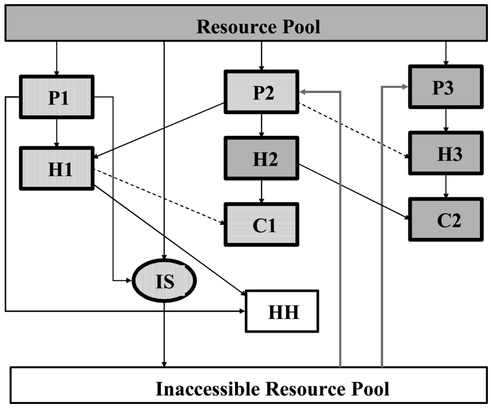

FIGURE 1. Integrated ecological-economic-social model: The compartments represent a resource pool (RP), three plants (P1, P2, and P3), three herbivores (H1, H2, and H3), two carnivores (C1 and C2), human population (HH), an inaccessible resource pool (IRP), and an industrial sector IS.

The mass in the nondomesticated (wild) compartments is a function of the natural growth, death, and predation (consumption). An exhaustive description of the model, including details of the macroeconomic model, can be found in Whitmore et al. (*12*).

Model Modification. In order to conduct scenario and policy guideline studies, it is essential to have a dynamically (relatively) stable and realistic model. However, model simulations, for the parameter values reported in Whitmore et al. (*12*), exhibit instability such as total loss of mass in some or all of the ecological compartments (*20*). Moreover, some model parameter values are based on logic and mathematical considerations rather than actual data on a real system. This caused some variables to take values that are not realistic. To formulate a stable and realistic model, various modifications to the basic model are proposed in this work. These modifications are

• Waste discharge fee: A waste discharge fee (on per unit mass basis) is charged to the industrial sector IS for contributing waste to IRP compartment. The discharge fee increases the IS product price and decreases the production targets, thereby shifting the focus from pure profit maximization. When the impact of discharge fee inclusion on the economic variables of the model is analyzed, it is observed that the demand functions are inelastic in nature. Thus, as product prices rise due to the imposition of a waste discharge fee on IS, humans (consumers) cut back their consumption by a proportion that is smaller than the proportional price increase.

• Human compartment: The parameters of the human compartment such as birth and mortality rates are modified to correspond with real data from published literature (*21*).

• Cyclic variations: Cyclic variations in the ecological compartments of the model are included to depict the natural low growth and high growth seasons (see Cabezas et al. (*22*)) of the plants. This also gives the model a measure of absolute time as one low plus one high growth season constitutes one year.

• New model parameter values: With the aforementioned modifications, a new model parameter value set was identified using a combination of sampling and partial rank correlation coefficient studies (*23, 24*), starting with the base case values reported in Whitmore et al. (*12*), and this led to a significant improvement in model stability.

These model modifications have led to the formulation of a stable and qualitatively more realistic model. This is one that we feel can provide qualitatively realistic and valuable insights into the behavior of real systems. Some of these insights are

• The demand functions for P1, H1, and IS are critical for model stability.

• Increasing the impact of economic functions leads to sudden nonlinear shifts in model dynamics.

• Maximum permitted grazing of P2 by H1 must be carefully regulated to ensure the stability of natural compartments.

• Consumption of RP (natural resources) by domesticated compartments must be reduced.

• The flow of mass from domesticated to wild (natural) compartments should be increased to maintain balance.

Scenario Analysis. Scenarios are plausible, challenging, and relevant stories about how the future might unfold under certain assumptions, which can be told in both words and numbers (*25*). While predicting the future is impossible, scenarios address real-world questions by offering insights into uncertainties and consequences of current and possible future actions, and hence support more informed and rational decision making. It is important to understand that scenarios are not forecasts, projections, or predictions. The actual future development can be a combination of multiple scenarios, and different scenarios might be realized for different systems in the world. The consideration of scenarios consistent with the model is, therefore, critical. Assumptions within the scenario must be plausible and logical (*3*) while being consistent with model scope and assumptions. It is for this reason that various scenario analysis studies differ in the details of scenarios, depending on the model capabilities and objectives. Some important studies include Bossel (*3*), Janssen (*26*), Meadows et al. (*2*), Millennium Ecosystem Assessment (*25*), and Global Environment Outlook-4 (*27*) reports. The integrated ecological-economic model considered here is a fairly abstract representation of the world. The primary focus is to track mass flow between important compartments while ignoring intricate details. Given this scope, the following three different scenarios are identified:

• Human population explosion.

• Increase in per capita consumption by humans.

• Human population explosion along with increased per capita consumption by humans (combination of the first two scenarios).

It must be noted that these scenarios focus on how mass flows will be affected and consider only those possibilities that are general and fairly well-known. To understand the impact of each scenario, the results are compared with a base case scenario. This base case scenario predicts the future development if the current population and consumption patterns are maintained. Although such a scenario is not possible in reality, it is very useful in isolating the impacts of the considered alternative scenarios. A simulation horizon of 200 years is considered. This highlights the long-term trends while ignoring short-term disturbances that might be inconsequential in the broader picture. The important results from the scenario simulations follow (a comprehensive discussion and plots are provided in the Supporting Information).

Human Population Explosion. Human population explosion represents a primary concern for environmentalists regarding resources and ecosystem services. Owing to the predicted drops in mortality rate (due to better health care) and birth rate (due to better education for women and increased awareness of birth control measures), the human population is expected to peak and settle to about twice the current value in 50−100 years (*25, 28*). Beyond this point, aging population and a further drop in human fertility rates is expected to lead to a steady drop in human population (*21*). Although a universal consensus on the exact numbers may not exist, this general trend is accepted to be the most likely scenario.

In the model, the human mortality and birth rates are varied together to model the desired population variations.

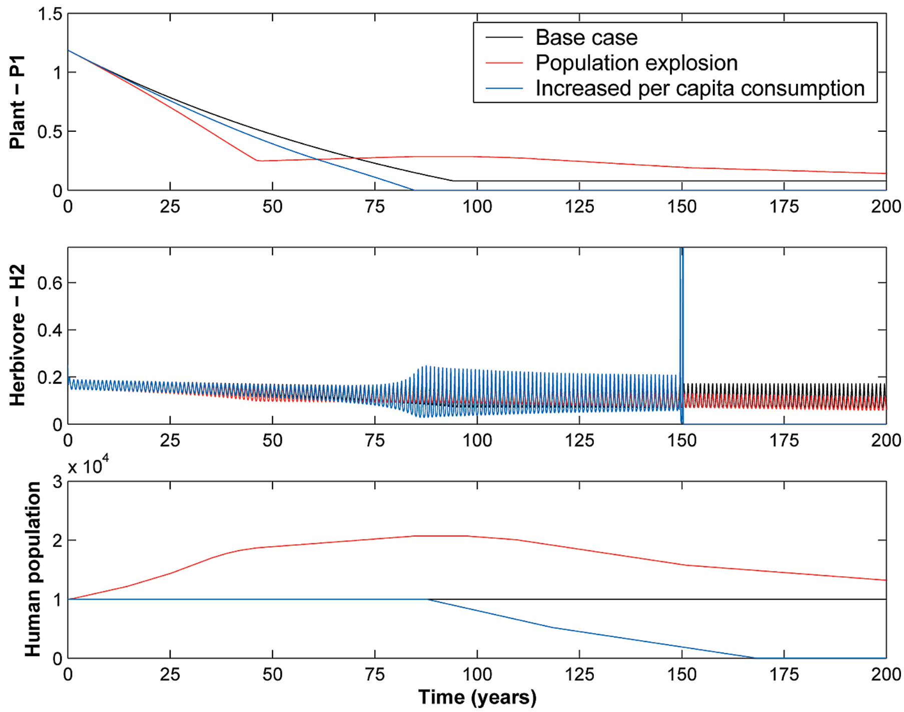

FIGURE 2. Scenario study results: Compartments P1, H2, and human population.

The mass fluctuations for some important compartments, along with those for the base case, are shown in Figure 2 in red (detailed plots in Supporting Information). The most important observation is the lack of any catastrophe in the system. The significant drop in the total mass of P1 has a cascading effect on compartments H2 and C2 due to the interconnected flows. In long-term, the mass of P1 is decreasing at a very small rate. Since P1 is a producer of plants, it can be correlated with the agricultural sector. The scenario, therefore, predicts a steady drop in the total mass of the agricultural sector due to increasing consumption by the rising population. During the second half of the horizon (years 100−200), the drop in the human population is not accompanied by a corresponding drop in the total human compartment mass, meaning that the per capita mass is increasing substantially. This "fattening" effect has been supported by recent studies by health economists and has been attributed to economic growth (*29*).

In terms of the economic parameters, fluctuations in human population have an inverse correlation with wage rate, which has implications on the prices of IS, P1, and H1. These fluctuations cause the demand for P1 by humans to reduce (due to the increasing scarcity) while the demand for IS and H1 rises to fulfill the overall human food demand. Although human population decreases during the second half of the simulation horizon, interestingly, the demand trends do not reverse. This is out of the necessity to satisfy the human demand for resources through a combination of P1, H1, and IS. Since the availability of P1 is continuously decreasing, the demand on IS and H1 increases even in the presence of increasing product price for IS and H1. From a social perspective, this alludes to the prediction that the per capita human consumption of resources will increase in the future, even at a higher cost.

To summarize, the scenario suggests a shift in the consumption patterns by humans due to price changes caused by the availability of cheaper labor. The changes in human population have cascading effects on the product prices, consumption patterns and ultimately the mass in various compartments. Although the scenario does not predict any true catastrophic changes, the mass in compartments P1, P2, and C2 is distinctly lower than at the start and is continuously decreasing. Under such conditions, the model is susceptible to breakdown due to sudden fluctuations such as those caused by natural disasters.

Increased Per Capita Consumption. With the ever increasing quality of life throughout the globe, the per capita consumption of mass and energy is continuously increasing. This has led to the fear that continued rise in consumption will ultimately cause a breakdown in ecosystem services (*30*), and this scenario attempts to shed light on this issue.

The future rise in the rate of resource consumption is more difficult to quantify than population rise because different resources have different trends (e.g., water and energy in terms of per capita). Some studies, however, predict that the average per capita consumption of many resources will increase on average by about 50% over the next 50 years (*2*). Taking this as the basis, the scenario models a 200% rise (linear) in per capita consumption rate in 200 years over the current rate. A recent Global Environment Outlook study by the United Nations reports that the present level of human resource consumption is 40% more than the sustainable threshold (*27*). Therefore, the assumptions in the scenario are reasonable. The coefficients of per capita demand functions in the price setting model are scaled linearly to vary the per capita consumption. The population is considered to remain constant. Although this is unrealistic, it allows one to isolate the effect of increasing consumption rates.

The mass fluctuations for some important compartments are illustrated in Figure 2 in blue (detailed plots in Supporting Information). The increasing demand causes the mass of P1 and IS to reduce sharply. Unlike the case of population explosion though, the mass of P1 does not settle at a small nonzero value, rather it reduces to zero. Since P1 is not available to humans as well as IS, IS is not able to continue production. This leads to a lack of resources for the humans, and the human population starts to drop suddenly in the later part of the simulation horizon (after about 80 years). This illustrates a catastrophe where scarcity of resources causes loss of human life. Once the human population reaches a low enough value making the availability of labor difficult (around 150th year) transformation of P2 into H1 is affected causing a disturbance in various other natural compartments (extinction of H1, H2, and C1).

In terms of the economic parameters, a monotonic rise in all demands and product prices is observed causing a continuous drop in the mass of IS and P1. In the population explosion scenario, price, and demand fluctuations in one product (P1) are balanced by fluctuations in the other products (IS and H1), which ensures the long-term stability of the system. In this scenario, however, no such mechanism is operational.

Human Population Explosion and Increased Per Capita Consumption. This scenario is a combination of the first two scenarios. The rates of change in population and consumption are the same as those for the first two cases. The results illustrate that the model becomes unstable due to lack of resources for humans, as for the increased per capita consumption scenario. The model dynamics leading to instability are, however, somewhat different than those for the increased consumption scenario. One might expect that increased consumption when aided by population rise should hasten model instability. This, however, is not observed. This is because the human population fluctuations have repercussions on the wage rate. This modulates the various product prices and demands and delays the extinction of compartment P1, which has strong implications on the model stability. Effectively, it is observed that the model becomes unstable at a later date (about 25 years) than that for increased consumption at constant population.

Since the human population is more than likely to follow the prediction mentioned in this scenario, the combined scenario is of more significance than increased consumption only scenario. The comparison of these two scenarios though emphasizes the nonlinear and nonintuitive nature of these interactions. Simulations show that up to a 50% increase in per capita consumption does not lead to instability in 200 years. Any larger increase, however, causes extinction in P1 and IS, and the extinction time is hastened with larger increases in consumption.

Time-Dependent Policy Development. The aim of the work is to explore and develop time dependent management policies for sustainability of the integrated model. Although such an approach is computationally more difficult than developing time-independent policies, the dynamic nature of all natural systems, such as the one considered here, makes it more appropriate. Control theory, particularly optimal control, has been at the forefront of these applications primarily due to its ability to effectively handle nonlinear systems (*31, 15, 32–36*). The model considered in this work, however, is quite complex and has model discontinuities (such as minimum or maximum functions), making the application of established control theory approaches infeasible. Instead, a dynamic optimization approach, where a policy variable at each time step is considered as an independent decision variable, is used. Since a gradient based optimization approach cannot be used due to previously mentioned model complications, pattern search technique, a popular direct search technique, is used in this work. This technique uses only the objective function value (and not the derivative information) to decide the optimization search direction, and therefore, is computationally simple, effective, and widely applicable (*37*).

To solve the dynamic optimization problem, it is essential to formulate a mathematical measure of sustainability or a sustainability index as the objective of the problem. Cabezas and Fath (*38*) have proposed Fisher information (FI) (*39*), a quantity from information theory, as a sustainability measure for dynamic systems. One of its interpretations, relevant for this work, is as a measure of the dynamic order or organization of a system or phenomenon (*40*). The FI based sustainable regime hypothesis states that the time-averaged Fisher information of a system in a persistent regime does not change with time. Any change in the regime will manifest itself through a corresponding change in Fisher information (*38, 41*).

Based on the hypothesis, this work uses minimization of the time averaged Fisher information as the objective in the pattern search problem, successfully implemented in previous studies (*35, 36*). The objective function is defined as

$$J = \min \int_0^T (I(\mathrm{t}) - I_\mathrm{c}(\mathrm{t}))^2 \mathrm{d}t \qquad (1)$$

where, $I(\mathrm{t})$ is the current FI profile, $I_\mathrm{c}(t)$ is the targeted FI profile for a stable system, and T is the total time horizon under consideration. The functional form of the equation for a discretized model, such as the integrated model analyzed here, is given as

$$J = \mathrm{Min} \sum_{i=1}^{N} (I(i) - I_\mathrm{c}(i))^2 \qquad (2)$$

where, N is the total number of model cycles considered, and $I(i)$ and $I_\mathrm{c}(i)$ are the average FI values for the ith model cycle for the current and targeted profile, respectively. In this work, one model cycle corresponds to one year and hence N is equal to 200, and $I(i)$ and $I_\mathrm{c}(i)$ are the average values for one year. The model simulations as well as control problem solutions explained later are done in FORTRAN.

Fisher Information Computation. Since Fisher information (FI) can be computed using different sets of model variables, it is important to determine the set that reliably reflects the model dynamics. This is important not only to make FI profile based prediction of future instability or model regime shifts more reliable, but also to formulate the right FI based objective function. Hence, the model variables can broadly be classified as either ecological (including all the compartmental masses) or economic (including human demands and per unit product prices of IS, P1, and H1, wage rate, GDP (gross domestic product), and mass of P1 consumed by H2). The FI profiles for these two variable sets are computed for the three scenarios mentioned before. The FI profiles are plotted in Figure 3. The FI profile comprises of a stepwise fluctuation in FI, where each step represents the average FI for one year. It is observed that, in general, FI computed from the economic parameters is a more reliable predictor of the model dynamics and instability than FI computed from the ecological parameters. For example, FI profile for economic parameters shows a steady decline for the scenarios with increased consumption as a predictor of the oncoming instability. It, therefore, is able to predict the instability much earlier (25 years for the second scenario). When FI profiles are computed using ecological as well as economic variables, the profile is very similar to that for the ecological variables only. This is possibly due to numerical reasons as the compartmental mass values are numerically much larger than the economic variables. Since computation of FI involves the calculation of first and second order derivatives of the system states with respect to time, rapid changes in compartmental values lead to large spikes in FI profiles, as observed in Figure 3. Such spikes are often indicators of system regime shifts and should not be ignored as numerical glitches.

Problem Formulation. For the objective of developing time-dependent policies, it is important to select parameters that can in reality be expected to be manipulable by humans as control variables. The possible control variables in the model, therefore, are

- discharge fee charged to the industrial sector (DF).
- mass of plant $P1$ necessary to produce one unit of the IS product (θ).

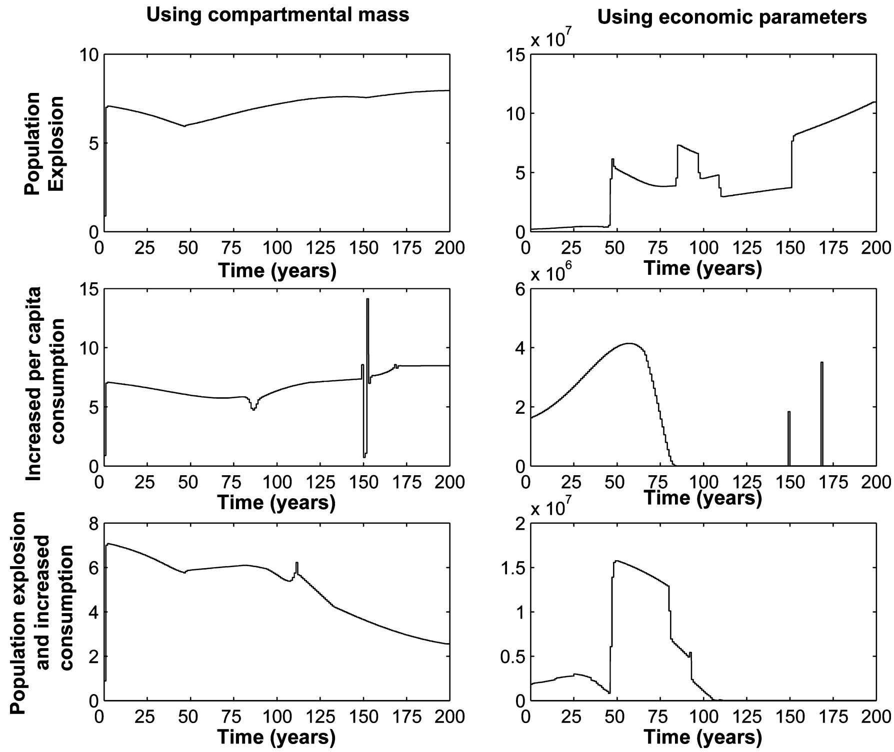

FIGURE 3. Fisher information profiles for various scenarios using different parameter sets.

• amount of P2 consumed by H1 through grazing (P2H1). The dynamic optimization scheme aims to stabilize the effects of population explosion allied by increased per capita consumption scenario which is unstable. Since population explosion is fairly well defined and certain, and it does not lead to an unstable system, it is considered as the target scenario. Therefore, $I_c(t)$ in the objective function (eq 1) is the FI profile of the population explosion scenario. It was observed that although economic variables are better predictors of model instability, use of FI profiles for ecological parameters in the objective leads to better results. This might be because (1) the compartment masses directly characterize the biological system collapse that the control system is trying to mitigate, and (2) changes in the economic variables are quite small, and their effect on the compartmental masses is often indirect. Therefore, the results reported here use compartmental mass values to compute the FI profiles in the objective function.

Policy Development Results. Initially, the dynamic optimization problem is solved using DF (discharge fee), θ (mass of plant P1 necessary to produce one unit of the IS product) and P2H1 (amount of P2 consumed by H1 through grazing) as control variables independently. Results using θ and P2H1 as control variables show that these parameters are ineffective as policy variables since there is insignificant improvement in the model dynamics. The use of DF, however, shows interesting results. For DF, the base case value is1×10^{-8}, while the lower and upper limit (bound) is 1×10^{-9} and 1×10^{-7}, respectively. The DF profile for the solution indicates that the discharge fee needs to be increased on an average by about 3.5 times the base case value. The effect of this increase on model instability is mixed. The mass profiles for some of the important compartments for the controlled model with DF as the control variable are illustrated in Figure 4. On the positive side, model instability in the form of human population drop is delayed by about 30 years. Higher DF raises the IS product price, which reduces the demand for IS by humans, and consequently the demand for P1 by IS. This delays the extinction of compartment P1 and, consequently, the model instability. There are some points of concern though. Compartment H2 goes to extinction, while compartment C1 shows reduction in the average mass in long-term. This is because maintenance of P1 reduces the availability of RP to P2. This leads to a reduction in the mass of P2 and subsequently in the mass of H2 and C1. This cascading effect of higher DF on P2 becomes very important when DF is increased to a value higher than that suggested by the optimal solution. Although higher DF further delays the extinction of P1, the mass of P2 decreases gradually and ultimately becomes zero, triggering instability in various wild compartments. Thus, excessive rise in DF is also not desirable. The use of discharge fee as a control variable, therefore, is able to achieve some success at the cost of the extinction of a wild compartment. An average rise of 3.5 times the base case value, though, is not unreasonable. It, however, cannot be a long-term solution to the basic problem of increased consumption.

An extension of the work is to manipulate multiple variables simultaneously, which corresponds to multivariable control. Since multivariable control is complicated due to the interactive effects (coupling) of control variables on model dynamics, a judicious combination of the control variables is important. This work solves the problem using two different control variable combinations: DF with P2H1, and DF with θ. The pattern search algorithm is modified to simultaneously search for the favorable values of two control variables instead of one, ignoring the coupling effects. Figure 4 shows the results using both combinations (detailed plots in Supporting Information).

The use of DF and P2H1 combination delays model instability by about 75 years, which is a significant improvement as compared to the uncontrolled and single variable controlled system. Moreover, the extinction of H2, which is observed when DF alone is used as the control variable, is averted. The control variable profiles show that on an average the discharge fee needs to be increased by 4.5 times and P2H1 needs to be decreased by about 75% over the respective base case values. The analysis of this result is quite interesting. The mechanism of the positive effect of increased DF on model stability is the same as explained before. It has been

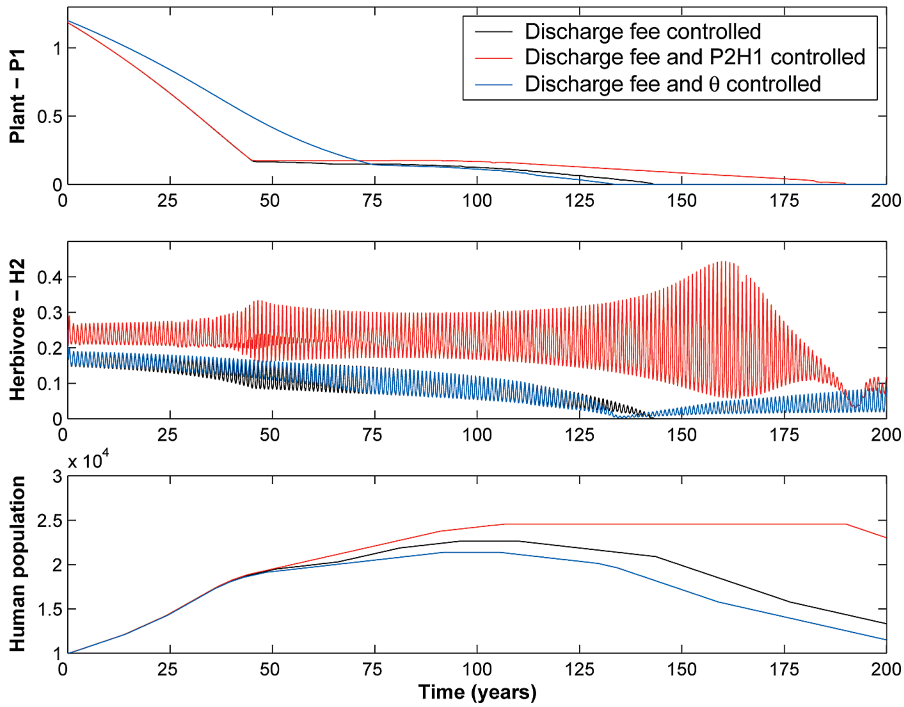

FIGURE 4. Control problem results: Compartments P1, H2, and human population.

mentioned that excessive rise in DF (more than 3.5 times the base value) causes P2 extinction, leading to overall model instability. A 4.5 times rise in DF in this case, therefore, should have caused instability. However, P2H1 limits the amount of P2 consumed by H1 (through grazing), and hence has an impact on the mass of P2. Reducing the value of P2H1 reduces the stress on compartment P2 and permits a larger rise in DF without causing model instability. This result highlights the typical trade-offs that are observed while managing such complex systems.

For DF and θ combination, although the overall performance is very similar to that using DF as a single control variables, the extinction of H2 is averted. The control profiles explain the reason for this observation. In the first half of the simulation horizon, the value of DF is high, whereas the value of θ is very low. This reduces the consumption rate of P1 by industries causing more P1 to consume RP, thereby affecting the P2 mass (as explained before). In the later half of the simulation horizon though, the Df is reduced and θ is simultaneously increased. This causes, due to various cascading effects already mentioned, the mass of P2 to rise and, hence, H2 to survive. There are some positive aspects for the human compartment also. When DF and θ are simultaneously manipulated, the rise in human mass and population (beyond that for the population explosion scenario) is avoided. Such a rise is observed for the case when DF alone is used as the control variable. It can, therefore, be concluded that the use of DF and θ together gives results that are better when considering the whole system.

Discussion

With sustainability emerging as the central theme in policy making, a systematic analysis of complex systems is essential. The aim of the work is to perform such an analysis on an integrated ecological-economic model system and to present qualitative results that will aid policy making within the scope of the model and its assumptions. Scenario studies on the integrated model show that increasing per capita consumption of resources is more critical than population explosion. Policy makers, therefore, should focus more on reducing the resource consumption rates. It is also observed that undesirable developments in the nondomesticated compartments are often overlooked by typical economic observations and, hence, might lead to sudden disasters if ignored. When Fisher information is used as an indicator of sustainability, it is found that economic parameters are better predictors of instability. Dynamic optimization is used to develop time dependent policies for the unstable scenarios, and Fisher information is used to formulate the objective. The results show that waste discharge fee, individually and in combination with other control variables, is effective in delaying but not eliminating instability. The results, though, highlight the trade-offs that will typically be observed in managing these systems where temporary solutions with different success levels can be achieved with associated costs and penalties. While extending conclusions drawn from a simple model system to reality can only be done in the most general sense, we note that none of the control schemes were successful in achieving sustainability over the long-term. It should be noted that our model studies suggest that the scale for observing system instability is of the order of one hundred years or less. Lastly, our model studies suggest that an effective control or management strategy, while not preventing catastrophes, may still delay them by enough time for new technologies or new paradigms to evolve.

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Supporting Information Available

Details of the integrated ecological-economic-social model, including the operational equations, input and output variable list and computation procedure; Exhaustive discussion of the scenarios; Exhaustive discussion of the dynamic optimization problem solutions including plots for control variable profiles. This material is available free of charge via the Internet at http://pubs.acs.org.

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