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# Current Challenges in Experimental Watershed Hydrology

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Additional information is available at the end of the chapter

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## 1. Introduction

The river basin, watershed or catchment is central to many of concepts in hydrology [1] including contaminant hydrology. In fact, two different studies in the Seine River basin during the end of seventeenth century, independently made by Pierre Perraut and Edme Mariotte have been identified by A. K. Biswas as the beginning of quantitative hydrology [2], Along with Perraut and Mariotte, Biswas also suggested Edmond Halley as co-founder of experimental hydrology. It was later accepted that scientific hydrology was founded on these two basin studies [3].

However, basin studies developed slowly until the end of nineteenth century when public demands accelerated. The first modern basin studies commenced in Emmental in Switzerland during the 1890s and were focused on hydrological differences between two small, ca 60 ha, basins [1]. A multitude of basin studies have appeared since the early twentieth century from many parts of the world. These include: (1) Wagon Wheel Gap experiment of USA begun in 1910 in two forested basins [4]; (2) Valday Hydrological Laboratory of USSR begun in 1933 and focused on field investigations of multiple hydrological parameters in watersheds with different scales [5]; (3) Coweeta Hydrologic Laboratory of USA set up since 1934 for forest hydrology and, ecological research in two main basins with 32 sub-watersheds under various treatments [6]; (4) Harz Mountains experiment of Germany with a pair of catchments, Wintertal and Large Bramke, begun in 1948 and focusing on hydrological effects of land use [7]; (5) Alrance experiment of France started in 1950 and focused on streamflow patterns[8]; and (6) Bluebrook Runoff Experiment of China established on 1953 and focused on drainage problems of the vast agricultural plain of the North Huai He River [9].

A period of rapid worldwide development of hydrological basin studies resulted from the International Hydrology Decade (IHD) Representative and Experimental Basin Programme 1965- 1974, with an estimated 3000 basin studies conducted during the Decade [1]. “Representative basins (RBs), which are selected as representative of a region of presumed hydrological similarity, are used for intensive investigations of specific problems of the hydrological cycle (or part thereof) under relatively stable, natural conditions” [10]. Experimental basins (EBs) are relatively homogeneous in soil and vegetation, have uniform physical characteristics, and are deliberately modified for study [10]. However, many EBs are just well-instrumented basins of small area [11]. Sometimes both RB and EB are included, as in the fruitful Plynlimon experiment in the UK [12].

If we designate the first phase of basin study (until ca the middle twentieth century) as foundational and the second phase (during/after the IHD) as developmental, it would appear that experimental watershed hydrology is inevitably going into a third phase of transition and innovation. Most field experiments in watershed science to date remain largely descriptive, with results that are difficult to generalize [13]. In effect, “... catchment hydrology is trapped in a dead-end track, a theoretical impasse” [14]. Experimental watershed studies, the core of watershed hydrology, are now confronting tremendous and even more complicated challenges, which are raised mainly from changing environment conditions and partly from the weakness of current watershed studies. This chapter provides an overview of the main challenges facing experimental watershed hydrology. Addressing these challenges will hopefully lead to substantial innovation in the field.

An important and instructive paper addressing a new vision of watershed hydrology [13] asked “What’s wrong with the status quo?” If we posed this same question to the experimental hydrologist, the limitations of most existing RBs/EBs might be characterized as mainly twofold. First, study basin designs have been limited by the black box concept and many misconceptions (e.g., the linearity, non-heterogeneity, additivity of hydrologic systems etc.). Second, operation has been substantially bounded by the hydraulic conception of these watersheds as isolated hydrological systems. All of these watershed studies monitor only total runoff at the stream-outlet and the subsurface responses of the watershed are only estimated by hydrograph separation, etc. These characteristics undermine the formulation of a unified theory of watershed hydrology [14] and the development of watershed models [13,15].

There is a clear need to move beyond the status quo and expand from this narrow hydrological perspective to generate hypotheses governing general behavior across places and scales [13], with the ultimate aim “to advance the science of hydrology” [15]. For the third phase, the stage of transition and innovation, instead of classic RBs as described above, another kind of experimental watershed, the Critical Zone Experimental Block (CZEB) is suggested, with its concept and infrastructures very different from that of current EB. Following the concept of Critical Zone defined by the National Research Council [16], the CZEB will have practical boundaries for different geomorphologic regions.

An ongoing trial application of the CZEB approach is presented in this chapter.

## 2. Challenges

Numerous challenges face researchers as they move forward to advance hydrology science using experimental watershed studies.

### 2.1. Demands of advancing hydrology science

More than two decades ago, V. Klemes proclaimed that “hydrology is as yet lacking a solid scientific foundation needed for its development as a natural science” [17] and this sentiment has been reiterated by others. Bras and Eagleson said “in the modern science establishment, this niche is vacant” in their paper titled “Hydrology, the forgotten earth science” [18]. Jim Dooge asked “Is hydrology now an established science? Is hydrologic practice now firmly based on scientific principles?” [19]. McDonnell et al concluded that “the critical ideas and positive vision presented in that (Dooge) paper remain just as fresh, relevant and, unfortunately, very much unfulfilled” [13]. Sivapalan also suggested several steps toward a new unified hydrologic theory [14]. Substantial progress in hydrologic science “ultimately depends on new experimental work, new field observations, and new data collection networks” as summarized by Kirchner [15]. But, what shape will these new experiments and networks take?

*Coupling processes.* A watershed is an inherently dynamic ecological system composed of a variety of biotic and abiotic processes. For such systems, it is important not only to focus on hydrological process but also on linked ecological, biogeochemical, pedological and geological processes. Misunderstandings of the interrelatedness of these processes are commonplace (e.g., summarizing the net effect of all biological processes as evapotranspiration). The vegetation root system forms a dynamically complex net for preferential flowpaths. It changes runoff generation mechanisms not only for surface runoff but the subsurface flows, and the root system itself is effected via feedback from the various, changing flow patterns. Vegetation growth also adapts according to climate, infiltration, soil water and even evapotranspiration while feedback mechanisms reinforce the interconnectedness of the watershed system [20].

*Innovative measurements.* More than twenty years ago, JE Nash suggested that discovery in hydrology sciences has been limited by “a deficiency in our empiricism” and, “our tolerance of poor methods of observations” [21]. Since then, methods of observations have improved significantly. However, our observation techniques still inhibit hydrology and a fundamental change to systematic measurement programs is needed. The current observation programs are aimed mostly at classic watershed hydrological process. Instead we should focus on a holistic description of heterogeneity of not only the watershed hydrological process but all related processes as described above, as well as landscape properties and climate inputs.

*Supporting and testing hydrological model.* Physically based hydrological models are one of the most promising directions for advancing hydrologic science. Grayson et al concluded that “the models are enabling us to ask more questions, many of which are fundamental to our understanding of the natural systems that are the subject of our models” [22]. However, vital weaknesses in current watershed models exist. First, they are generally based on “well known small-scale theories such as Darcy’s law and the Richards equation for coupled balance of mass

and momentum" [13] and do not accurately capture the spatial heterogeneity, inherent non-linearity, and non-additive properties of natural watershed systems. Second, a large number of models are heavily over-parameterized, leading to equi-finality, wherein multiple combinations of parameters can yield equivalent results [13]. Grayson et al showed that comparable fits to a hydrograph could be achieved using a saturated overland flow model or Hortonian flow assumptions [22]. Kirchner argued that "parameter-rich models may succeed as mathematical marionettes dancing to match the calibration data even if their underlying premises are unrealistic" [15]. Kirchner concluded that if "the present trend away from physical processes and toward mathematistry ('blackboard hydrology') continues in hydrologic education and practice, hydrology will end up in a dead end as a science and become useless for applications" [17].

To achieve substantive improvements, models should be developed "in conjunction with, and as an integral part of, carefully planned and executed field studies established for the purpose of advancing our knowledge of the natural system"[22]. Key experimental basin requirements for model-generated hypothesis testing include: use of innovative basin designs and inclusion of sufficient variation in physiographic settings, hydroclimates, and watershed scales.

## 2.2. Hydrologic replumbing and natural climate oscillations

The National Research Council of the US identified anthropogenic perturbation and replumbing of the hydrologic cycle [23] as a fundamental challenge [16] in watershed science. Human influences dominate "the natural cycle of freshwater causing environmental changes that have been argued to move the planet to a new geologic era termed the 'Anthropocene'" [23,24].

Natural climate oscillations with observed large, abrupt events, the widespread millennial scale climate changes of the last glaciation (consisting of Dansgaard-Oeschger oscillations, Heinrich events etc.) are hypothesized to have been forced by North Atlantic atmosphere-ocean-ice interactions [25]. It has been argued that these oscillations are ultimately driven by variations in eccentricity, axial tilt and precession of the Earth's orbit as described in the Milankovitch theory. The decadal and multidecadal climate variations are generally linked with recognized dynamics, such as the El Niño Southern Oscillation (ENSO) [23].

Anthropogenic modification of the water cycle could "push the climate into new regimes" because "climate change has taken the climate system out of the repeated cycle of glacial-interglacial episodes" [23]. These alterations may accelerate the arrival of millennial scale events and trigger a "tipping point" transition resulting in "major climatic perturbations on time scales of decades to centuries"[26]. How might experimentalists, a small proportion of total hydrologists, address these challenges with progressive basin studies?

*Long term monitoring.* Well-established representative basins and benchmark experimental basins distributed across different physiographic and hydroclimatic conditions are critical for assessing hydrologic changes due to replumbing. Basins instrumented during or before the 1950's may also be used to determine patterns of rainfall redistribution, resultant stream flow and related biological issues.

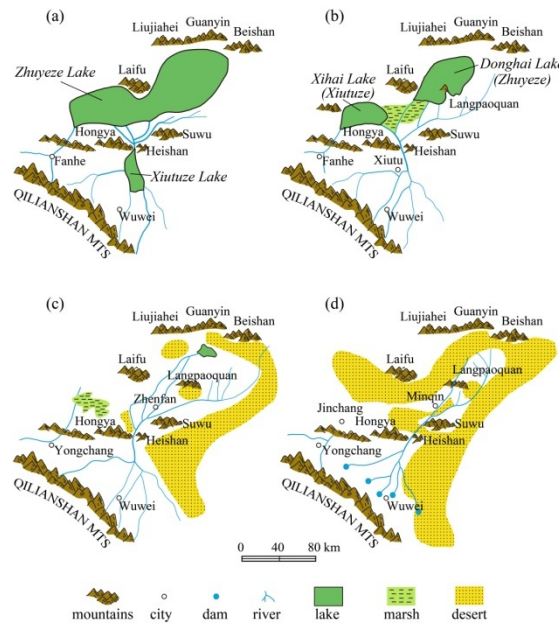
*Consequences of replumbed hydrology.* Monitoring and unraveling the hydrological and ecological responses and the feedbacks due to the replumbed hydrological cycle from water conservancy projects, especially those in arid and semi-arid regions, is needed. Dams are perhaps the most dramatic examples of human capacity to transform nature in the name of development [27], but development programs and projects create both winners and losers [28]. Dams in arid and semi-arid basins, especially in endorheic basins, break the natural cycle, looting downstream groundwater and accelerating desertification [29]. Long distance water diversion may reasonably be viewed as an ecological "planning disaster"[30]. There are concerns that development is outpacing scientific understanding, a circumstance that may prove disastrous for the environment for the generations to come.

*Fundamental studies.* Various hydrological and biogeochemical fluxes through catchments are important to understanding the effects of hydrologic replumbing, but many of these are poorly understood and rarely measured. For example, various subsurface ecological and hydrological components cannot be measured directly and must instead be estimated from a small number of point measurements. Particular emphasis has previously been placed on the downward flux of catchment processes components (i.e., rainfall and discharge) rather than upward flux- land evaporation, transpiration, and upward recharge of groundwater flux, etc – even though "these fluxes serve as important regulators of the dynamics of the cycle"[23]. There are various unknown or poorly understood mechanisms regarding natural catchment behavior that are affected by the hydrologic replumbing occurring at local, regional and global scales. These include: (1) the old water paradox [31]; (2) network-like preferential flow[13], natural symmetry between water, landscape, soil and vegetation and the underlying organizing principles between soil, vegetation and other biotic elements [14]; (3) the combined mechanisms of the geological water-cycle and hydrologic water cycle in the shallow hydrosphere, as well as the deep circulation of groundwater, which go far beyond current hydrogeological boundaries [32]; (4) the puzzle of the missing sink for the remaining 40% of watershed carbon balance, which after years effort a consensus for it still has yet to emerge [33]; (5) the mechanisms involved in low flow hydrology and its relation to shallow and deep groundwater, regolith, land use, water quality and the biodiversity of aquatic ecosystems [34]; and (6) the coupling of basin geomorphologic processes with hydrologic and ecologic processes, including the poorly understood processes and mechanisms of environmental release, transport, and biological transformation of various contaminants in the unsaturated zone, shallow aquifers and deep aquifers.

*Historical data on water replumbing.* Historical data from sources other than hydrometric stage records will be very helpful to understand large-scale environmental changes, serving as a reference for the natural variation and anthropogenic impact. Figure 1 illustrates desertification in an endorheic basin as the result of human impacts. Four stages of man-earth relationship are illustrated, from the 'stage of natural harmony' until the stage of the environmental disasters, the punishment of nature (see Figure 1 caption).

*Design systematic experimental facilities.* The dynamic systems currently facing experimental hydrologists exhibit a wide range of heterogeneity and process complexity across large spatiotemporal scales. There are multiple ways to overcome such scaling problems [13,14], but

hydrometric measurement systems in natural basins still face challenges (e.g., the ability of current rain gauges with conventional deployment techniques to obtain precise precipitation inputs for natural basins remains controversial). It is necessary to have systematic experimental facilities consisting of laboratory physical models, hillslope “catchment”, small-size natural watershed with uniformly dominant vegetation, small-size natural watershed with more complex vegetation, unchannelized catchments, nested sub-watersheds, and watersheds of large size. Such experimental facilities need to be situated in zones with different physiographic and hydroclimatic conditions. Obviously, this would require collaborative efforts at an international scale.



**Figure 1.** Desertification of the Rocksheep River (Shiyang River) Basin [35], an endorheic basin with total drainage area of 40,690 km<sup>2</sup>. (a) prior to 121 BC, the “stage of natural harmony”, the area of its terminal lake around 540 km<sup>2</sup>; (b) 420- 589 AD, the “stage of relying”; (c) ca 1900AD, the “stage of impact”, the cultivated area during Qing Dynasty was developed quickly, which reached to an area about 4/5 of that of modern time. (d) ca 1990’s, the “stage of damage”, there are 4 dams since 1959, 22 dams until 1972 [35].

### 3. The Critical Zone Experimental Block (CZEB)

For the future of hydrologic basin study, another kind of experimental basin is suggested: the Critical Zone Experimental *Block* (CZEB) customarily known as Critical Zone Experimental *Basin*.

### 3.1. Deficiencies in the design and operation of current experimental basins

*Watershed surface laterality.* Ignoring the inherent interconnectedness of surface and subsurface watershed components reflects the fact that watershed study is driven by practical rather than scientific requirements. Hydrologists still lack a comprehensive understanding of surface responses to nonvisible and difficult to measure subsurface constraints. A physiological analogy might be studying the skin while ignoring the functions beneath. Focusing on surface responses using surface monitoring will quickly trap the observer with misconceptions. For instance, hydrologists delineate watersheds with surface topography, though actual watershed boundaries may cross topographically determined boundaries depending on subsurface features. Even our most fundamental measurements of the watershed – its shape and size – are strongly influenced by our inability to peer beneath the surface of the catchment.

*Hydrologic process laterality.* The hydrologic cycle is tightly coupled with other cycles [36]. The hydrologic cycle is also dynamic, not static, in nature. Studying the hydrologic cycle while ignoring the interconnectedness of other cycles is equivalent to studying blood circulation while only looking at blood vessels.

*Downward components laterality.* There is a tendency to emphasize measurements of downward components (e.g. rainfall, discharge and infiltration) rather than upward components (e.g. evaporation and transpiration aforementioned), even though upward components can be the dominant terms in water balance [37].

### 3.2. Lessons learned from the Chinese experimental basin studies over the last fifty years

The Chinese experimental watershed studies experienced a saddle-backed fluctuation with two peaks [9] during their fifty-year history. The lessons learned were summarized in cooperation with J.J. McDonnell, C. Kendall and N.E. Peters, as following [9]:

1. The research facilities need not only the RB and EB of natural conditions, but also those with controlled boundary conditions. All EBs of recent decades referred only to the natural boundary of the surface watershed, ignoring that of the bedrock. This frequently led to controversial results.
2. The EB should be treated as an integrated system, from surface to bedrock.
3. The runoff response of an EB, including surface and subsurface components, should be monitored hydrometrically. For all EBs in past decades, these components were monitored incompletely or incorrectly, insufficient even for 'black box' evaluations. As a consequence, most of the results can't be explained physically.
4. It is necessary to use multiple tracers to look inside the physical processes of the hydrological cycle and the anthropogenic impacts at the catchment scale.
5. Comparative EBs should be located in different climate and morphological regions and should have comparable monitoring strategies.

It was also suggested that hydrological experimentation should not address "only to the hydrosphere but the interactions between atmosphere, lithosphere, biosphere and the intelligence-sphere." To this end, there is a need to "develop approaches using physical,



chemical, isotopic, biogeochemical and hydrometeorological methods for basin studies as a 'hybrid' basin research"[38].

### 3.3. What is CZEB?

The term "critical zone" has been used in many publications to refer to wide-ranging things in areas of geosciences and mineral, etc (e.g., the geological formation, the rhizosphere, the transitional zones in alluvial coastal plain rivers, etc)[36]. In 2001, the National Research Council of US recommended the integrated study of the "Critical Zone" (CZ) [36], defined as "the heterogeneous, near surface environment in which complex interactions involving rock, soil, water, air and living organisms regulate the natural habitat and determine availability of life sustaining resources" [16]. The CZ is one of the most compelling research areas in Earth sciences in the 21<sup>st</sup> century [36]. This zone was further defined as extending from "the vegetation canopy to the zone of groundwater" in 2005 [39], or "the bedrock to the atmosphere boundary layer" [40] and, "top of the vegetation down to the bottom of the aquifers" [36] by 2010.

The Critical Zone Experimental Block (CZEB) geologically is a monolith-block with its surface, the watershed, bounded by topographical water divides, which define the surface boundary, and which lacks defined subsurface boundaries. A drainage basin conceptually is only its surface part, the visible face (Figure 2). The CZEB is actually an Experimental "Block" within the Critical Zone with a surface drainage basin (the watershed). It is a dynamic ecosystem coupled with various supporting systems but using hydrological processes as the unifying theme. It is a living, breathing, evolving boundary layer where rock, soil, water, air and living organisms interact [41]. The CZEB is the experimental hydrological watershed study in the CZ framework. It follows that the components within the CZEB (including monolith, hillslope, sub-watershed, etc) will be monitored according to the "downward and upward approaches to theory development in catchment hydrology" [14, 42]. "The Critical Zone Observatory network is the only type to integrate biological and geological sciences so tightly"[40], and following this, the CZEB network will perhaps be the only network to integrate the hydrological, biological and geological sciences so tightly as well.

### 3.4. Boundaries of CZEB

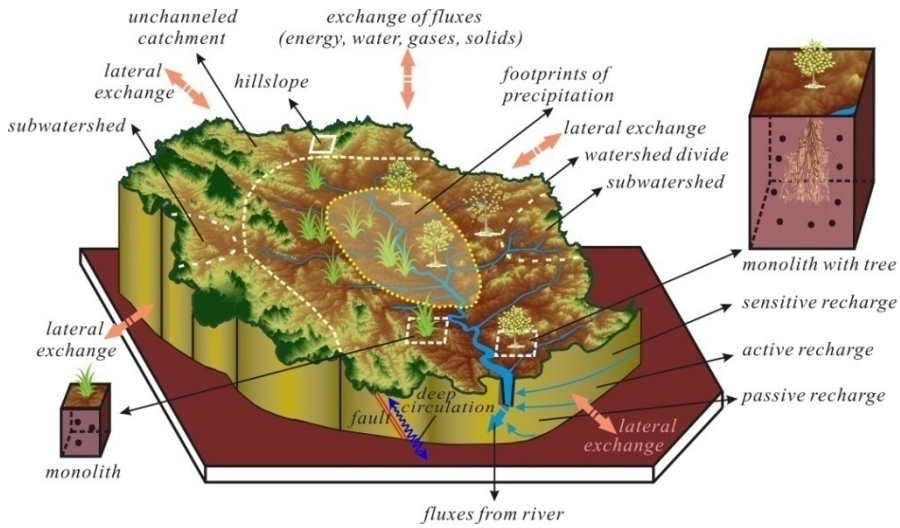
*Top.* The evaporation surface of the canopy in general is not the mean surface of the canopy but slightly lower. If the mean evaporation surface of the canopy is  $h$  (m) above the mean ground surface, then the top boundary of a CZEB is defined as  $H(m)$ , where  $H=(1.5 \text{ to } 2.0) \times h$  with coefficients of  $h$  varying according to the vegetation. This is mainly for the purpose of energy budget and eddy covariance flux observations.  $H$  is just the lower part of the atmosphere boundary layer.

*Bottom.* There are three cases:

Case I: Bedrock is situated at a relatively shallow depth from the ground surface while the regolith is shallow, too. The bottom boundary is defined as the geological boundary (Figure 3a).

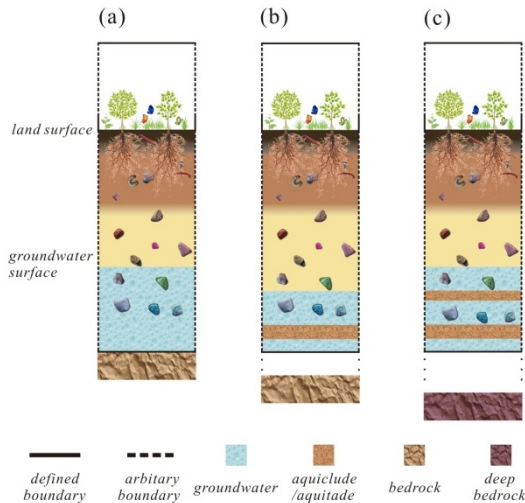
Case II: Bedrock is deep. The bottom boundary is defined as the plane where the tritium content of groundwater approaches zero or the detection limit of  $\pm 0.7$  TU (the "tritium naught line" (TNL)[43]), which is same as that of Case III (Figure 3b).





**Figure 2.** A conceptualized Critical Zone Experimental Block (CZEB)

Case III: There are stratified alluvia, potentially with multiple aquifers with aquicludes and aquitards, common in flat areas with thick deposit and deep bedrock, up to hundreds of meters or more. Groundwater recharge to the river can be separated into sensitive, active and passive zones. In this case, the bottom boundary of CZEB is defined as the bottom of the active recharge zone, the plane of TNL (Figure 3c).



**Figure 3.** Boundaries of CZEB

### *Lateral sides*

Part I: Above the ground surface up to height  $H$  as described above, delineated according to the surface topographic watershed boundary (Figure 3).

Part II: Below the ground surface, it is in general defined arbitrarily, except in the case of existing geological boundaries.

### **3.5. Functioning of CZEB**

CZEB is an ecological dynamic and evolving system. It also is a natural open system, exchanging mass and energy with its surrounding environment across 'arbitrary' boundaries. It is a dissipative, complex system, however, with some degree of self-organization [36].

*Interfaces.* CZEB encompasses "the near-surface biosphere and atmosphere, the entire pedosphere, and the surface and near-surface portion of the hydrosphere and lithosphere" [36]. Within the CZEB, various processes including hydrologic, atmospheric, lithospheric, geomorphic and geochemical processes are coupled and dynamically interrelated. To simplify the organization of various interfacing processes throughout the CZEB, compartment zones can be broadly separated as follows:

1. The zone above ground surface, the "aboveground vegetation zone" [36].
2. The unsaturated zone, "the belowground root zone and the deeper vadose zone" [36].
3. The saturated zone, the "saturated aquifer zone" [36].

This layering has a general trend of increasing density with depth, has a dampening effect on state variables with depth, and an increase in distance to energy input at the soil surface [36]. There is also "an overall trend of increasing response time" [36, 44].

*Mass – the material aspect of CZEB.* The material base of the CZEB is fixed and limited, involving rock, soil, water, air and living organisms [16]. On this point it happens to coincide with the ancient Chinese philosophy, the so-called "Five-xing", which holds that five fundamental elements form the universe and the Earth: "Jin" (metal), corresponding with the term "rock"; "Mu" (wood), with that of "living organisms"; "Shui" (water) with that of "water"; "Huo" (fire) with that of "air"; and "Tu" (soil).

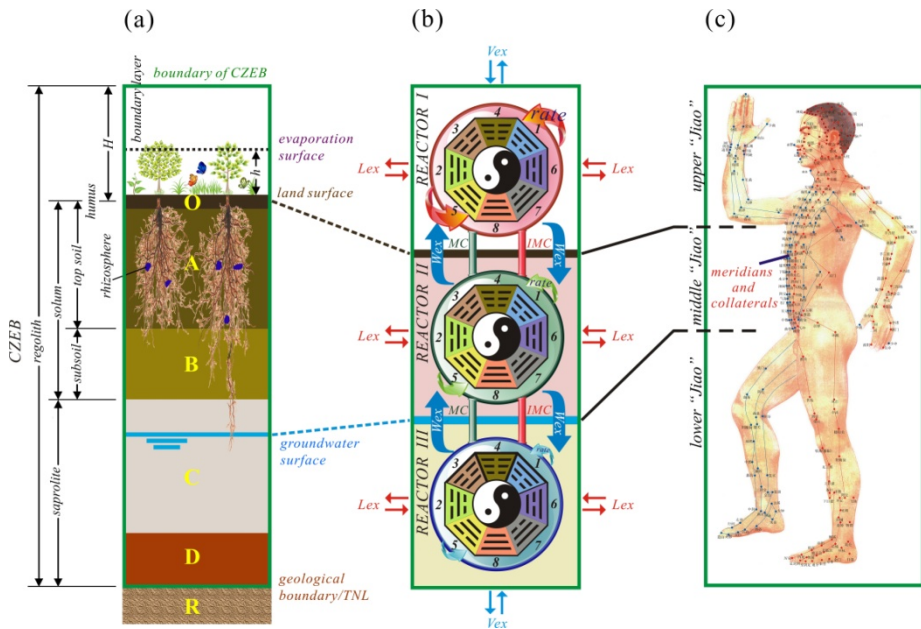
*Energy and force – the driven aspect of CZEB.* Continuous energy fluxes and inherent forces drive the CZEB system. External solar energy is certainly the vital component of external energy source. Various inherent forces include: gravity, surface tension, intermolecular forces, capillary force, etc which control its dynamic situation.

*Organization and entropy – the philosophical aspect of CZEB.* This open system of structural dissipative processes and irreversible evolution tends to increase its entropy spontaneously and go towards disorder [36]. Even the exchange of entropy fluxes across its boundaries is continuous. The second law of thermodynamics, from which the concept of entropy was derived, is one of the fundamental natural laws. However, the role of feedbacks of this nonlinear system will promote "self-organization" as energy dissipates, providing opportunities for dissipating energy to act again within the system towards the direction of order. "Conservation without evolution is death. Evolution without conservation is madness" [45,

46]. Conservation of energy appears in the CZEB at all scales, with observable ‘behaviors’ of organization, symmetry, genes, etc. Lin suggested including “information” as one of the four general factors (i.e., conservation of energy and mass and, the accumulation of entropy and information, which dictates the evolutionary outcome and the functioning of the CZ ) [36]. This is equally applicable to CZEB.

### 3.6. The reactors of CZEB

The CZEB can be separated into three interdependent zones as mentioned above with each zone consisting of the same base materials, the “five-xing” rock, soil, water, air and living organisms as well as non-material bases of energy, force and information. This coincides well with the ‘Bagua’ (‘eight-gua’) of the earliest Chinese philosophical work “I–Jing” (“The Book of Chang”). According to I–Jing, the origin of the universe was just a singularity of chaos (‘Tai–ji’), consisting of ‘Yang’ and ‘Yin’, the unity of opposites, which produced ‘eight-gua,’ the general principles governing development in the material world. Each “gua” is a combination of three whole lines (‘Yang’) and broken lines (‘Yin’). As in the four general characteristics of CZ elaborated by Lin [36]; in the CZEB, the material base refers to the five ‘gua’s (the “five-xing”) and the non-material base refers to another three ‘gua’s of energy, force and information, which together form a dynamic ‘Bagua’. Each zone has the same five material and three non-material components with different rates, Figure 4.



**Figure 4.** Schematic ‘reactors’ of the CZEB. The dynamic ‘Bagua’ in each ‘reactor’ operates with its own rate; 1- 5 : the material base (1- air, 2- water, 3- soil, 4- rock, 5- organisms); 6-8: the immaterial base (6- energy, 7- force, 8- information).

Each zone will behave as a ‘feed-through reactor’ according to Anderson et al [47]. It follows then that there are three feed-through reactors coupled together in the CZEB (Figure 4), with probably different rates and residence time of materials.

*Reactor I.* The first zone is the above ground surface zone, which also contains above ground vegetation up to its interface with atmosphere (Figure 4a). Evaporation and plant transpiration may account for 50% or more of the total local precipitation and use up to 50% of the total solar energy.

*Reactor II.* The second zone is the unsaturated zone, extending from the soil surface to the upper surface of the groundwater table. It consists of the whole soil profile: The O horizon (humus), A horizon (topsoil), B horizon (subsoil) and C, and potentially D, horizons (Figure 4a). The unsaturated soil zone has been recognized as “the most complicated biomaterials on the planet” [36].

*Reactor III.* The third zone is the saturated zone, extending from the groundwater table down to bedrock, including the capillary fringe (Figure 4a). There are two general cases for CZEB, including phreatic groundwater and confined aquifers.

#### *Functioning of reactors*

1. Reactor I includes hydrometeorological and ecohydrological processes, Reactor II includes hydrogeological and hydroecological processes, while the Reactor III is more exclusively for hydrogeological processes.
2. There is an overall trend of decreasing operation rates from Reactors I to III.
3. Each Reactor has its own lateral flux exchanges  $L_{ex}$  via the CZEB lateral boundaries (Figure 4b). The current hydrometric runoff data is only that from channel. Reactor I has vertical fluxes exchange  $V_{ex}$  with atmosphere via the upper boundary of CZEB while Reactor III has  $V_{ex}$  with deep aquifers and/or deep circulation via the bottom boundary of CZEB (Figure 4b). Fluxes include all material and immaterial components.
4. Reactors are closely coupled within the boundaries of CZEB. There are flux exchanges  $W_{ex}$  (Figure 5) between Reactor I and II and, and between II and III within the CZEB. There are flux channels  $MC$  (Figure 4) through I to III for materials, and flux channels  $IMC$  (Figure 4) through I to III for non-materials.
5. The  $MC$  appears similar to blood vessel system and  $IMC$  to meridians and collaterals of the human body from the Chinese art of acupuncture. The three “reactors” are similar to the so-called human “three Jiaos” (Figure 4), a central concept of Chinese medicine philosophy.

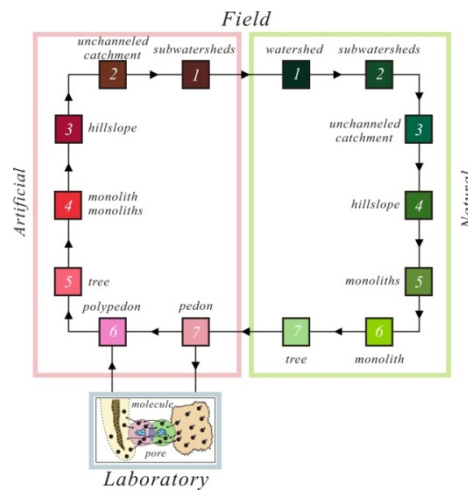
## **4. A trial of the CZEB: The Chuzhou CZEB experimental system**

### **4.1. The strategy: A two-way multi-scale approach**

*Isolation of the natural system.* In order to investigate the natural watershed system, which is filled with complexity and heterogeneity, simplification is incorporated as much as possible

(e.g., “isolation”) [9, 38]. Using detailed observation on ever-decreasing scales allows the investigator to focus and isolate processes from the naturally occurring heterogeneity (i.e., watershed, sub-watershed, unchanneled watershed, hillslope, monolith, a tree) (Figure 5). To some extent it is similar to dissection: splitting “problems into their smallest possible components”[48].

There are limitations in the field, however. Using a single tree as an example, the tree itself can be well instrumented, as well as the subsurface with pits up to meters deep, but still the fluxes from its bottom boundary would be unknown. In this sense, *isolation* refers to a system rather than the physical isolation of a natural unit, which can only be partially isolated in field. Strictly speaking, the field result from such natural systems can be explained only empirically, from a statistic or a stochastic view. Understanding causality mechanisms from observational studies alone is problematic. Thus, McDonnell et al (2007) noted that “most field experiments and observations in watershed science to date remain largely descriptive” [13]. They also discussed a shared ‘philosophical path’ which claims that “if we characterize enough hillslopes and watersheds around the world through detailed experimentations, some new understanding is bound to emerge eventually”, the authors concluded that “what this approach to experimental design has succeeded in doing is to help characterize the idiosyncrasies of more and more watersheds, in different places and at different scales, but with little progress toward realizing the Dooge vision” of “hydrologic laws”[13]. This does not mean, however, that field studies have no place in hydrology. On the contrary, the problem becomes how to “put the pieces back together again” [48]. In our case it is how to put together hydrologic mechanisms for a monolith, a slope, a sub-watershed and, a watershed.



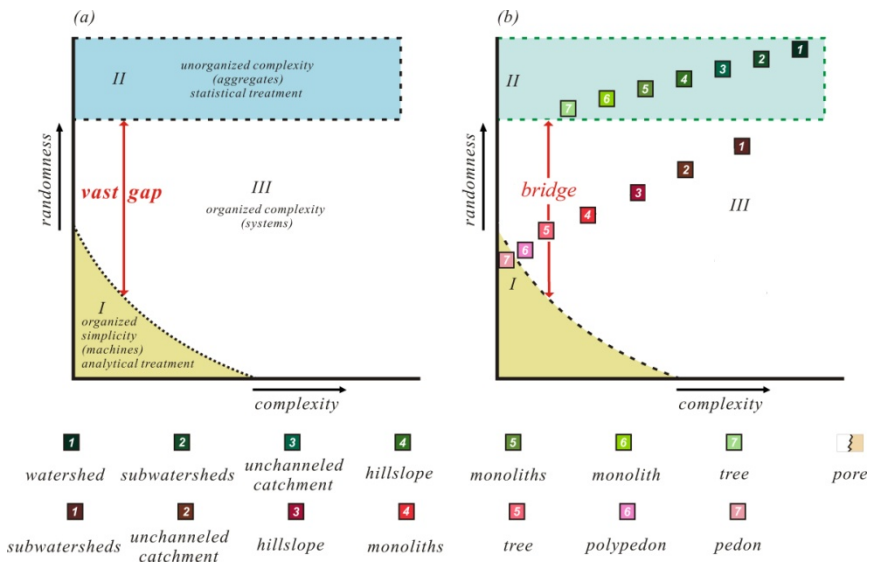
**Figure 5.** Concepts of the two-way multi-scale approach: natural and artificial

*Control and synthesis in the artificial system.* A series of artificial systems are suggested and defined for “control and synthesis” [9, 38]. They encompass the pedon, polypedon, monolith,

hillslope, catchment, until subwatershed (Figure 5). Artificial systems are necessary in order to move beyond problems of boundary and parameter control found in natural systems. It is in fact inspired by the aforementioned empirical impasse of natural systems; artificial systems overcome heterogeneity by controlling all variables and leaving only test variables to fluctuate.

*The laboratory system.* The two-way multi-scale approach can only be applied at the macroscopic level of watersheds or, maybe the mesoscopic level (e.g., catena monolith). In fact many problems (e.g., the old water paradox) can be trace back to mechanisms occurring on a microscopic level (e.g., the molecular and pore related mechanisms as that of microbial processes of some contaminants transformation). The laboratory system connects with and supplements the artificial system (Figure 5).

*Could this be a new vision for watershed hydrology and a unified theory of hydrology?* Lin summarized three systems of different levels of complexity and a vast gap between the two extremes [36] as shown in Figure 6. The coupled natural and artificial systems are analogous to ‘Yin’ and ‘Yang’, which is the successful philosophy of ‘Taiji’ (Figure 7). Klmes suggested “a rational search for meaningful conceptualization in hydrology can proceed along two routes: upwards and downwards” [42], or, the “upward or bottom-up approach” and the “downward or top-down approach” [14]. The two-way multi-scale approach perhaps could also be upwards and downwards. Hopefully, it could be a way for a new vision for watershed hydrology [13] and, for the unified theory of hydrology [14].



**Figure 6.** The gap between extremes of complex catchment systems. (a) Three types of complex catchment systems related to different treatment methods [14,19,36]; (b) The supposed field and artificial two-way multi-scale framework and the gap depending on the level of control. The gap becomes the bridge.



#### 4.2. The trial practice of the CZEB: The Chuzhou CZEB experimental system

The Chuzhou CZEB can be attributed to the artistic conception of the *West Brook*, described in a poem of Tang Dynasty where intellectual exiles of Song Dynasty found their Arcadia ca 900 years ago. On the same brook in 1962, a hydrological experimental field station with three watersheds was founded. About fifteen years later, construction of the experimental watershed hydrology had barely survived from the cultural revolution of the 'Road to Perdition'. The Chuzhou CZEB was an adventurous plan for a two-way experimental watershed system including natural catchments and artificial catchments with controlled boundaries. The experiment was, unfortunately, interrupted again during the 'market-economy tides' of China and only partially completed. A turning point emerged a few years ago, with more efforts towards an innovative CZEB system. In the meantime, the components of the natural and artificial systems are: (1) the natural system with main infrastructure of hydrological monitoring and isotope biomonitoring encompasses watersheds with drainage area of 82.1 km<sup>2</sup> (hill 34%, forest 5%, ponds 1.2%), 17.5 km<sup>2</sup> (56%, 10%, 0.6% *idem*), 4.5 km<sup>2</sup> (100%, 2%, 0% *idem*), 3.3 km<sup>2</sup> (100%, 100%, 0% *idem*), 2.0 km<sup>2</sup> (5%, 2%, 30% *idem*), 0.06 km<sup>2</sup> (80% paddy field), 4573 m<sup>2</sup> (Morning Glory Catchment, 100% weathered debris), and, a mini CZEB with directly observable surface and subsurface responses with a drainage area of 7897m<sup>2</sup>; (2) the artificial system with measurable surface and subsurface responses encompasses a catchment with surficial and bottom area (horizontal projection) of 490m<sup>2</sup>, a crop monolith of 150m<sup>2</sup>, two grass monoliths (L1, L2) of 32 m<sup>2</sup> each, a saturated monolith (LS) of 1 m<sup>2</sup>, and several weighing lysimeters. Here, the focus is on results from the two experimental watersheds - the mini CZEB watershed and the artificial catchment.



**Figure 7.** Reconciliation of two-way natural and artificial, 'Yin' and 'Yang' [14, 42].

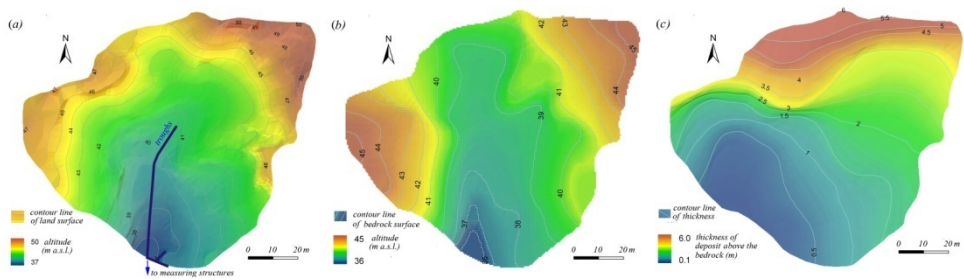
##### *The natural CZEB, Nandadish*

Nandadish has a surficial drainage area of 7897 m<sup>2</sup> and rests on the consolidated bedrock of the concordant body of andesitic and tuffaceous facies with a thin weathered layer. The boundary condition of this mini CZEB belongs to the type shown in Figure 3a. The Quaternary regolith overlies the bedrock, its depth ranges from of 1 to 7 m with an average of 2.46 m; its bed rock topography was surveyed via 69 drillings (Figure 8). The regolith consists of brunisolic soil of heavy loam, medium and clay loams; saprolite with prismatic and block structures. Horizontal and vertical fissures and cracks developed in the upper regolith. This watershed

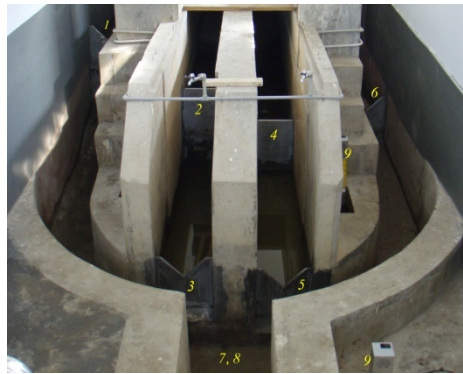


has an altitude difference of 12.9 m with a surface slope of 6.7% to 17.1% at different directions. The brook gradient is 6.7%. The coverage during the watershed's construction in 1979 was natural grasses with small shrubs and a few Masson pines aged 5 to 6 years. Since then, coverage has shifted to a dense forest with canopy height ca 8 m.

The main infrastructure includes: (1) measurement for energy flux, water flux, geochemical flux, gas flux; (2) changes of watershed storages of energy, water, gas and that of geochemical ions; (3) CZ tree experiment; and (4) variations of precipitation isotope fingerprints in flux and in storage. The surface and subsurface runoff are monitored directly via a trench, which extends upslope to capture subsurface and surface flow. This CZEB block has deep soils near the divide but only 1-meter depths near the outlet, making the block easy to close via a concrete wall installed to the bedrock at the outlet point. In this way, all surface and subsurface flow drain into discharge measuring structures (Figure 9).



**Figure 8.** The mini CZEB Nandadish. (a) Its surficial watershed; (b) Its bedrock; (c) The depths of deposit above its bedrock.



**Figure 9.** Discharge measurement structures for different runoff components from troughs with its location showing in Figure 8(a). 1- for rainfall; 2 and 3- for surface runoff; 4 and 5 – for interflow (30 cm below the soil surface); 6 – for interflow and groundwater flow (down to bedrock); 7 and 8 for total runoff (weirs are not shown). 1,3,5,6 and 8 are V-notch sharp-crested weir; 2, 4 and 7 are the full width rectangular sharp-crested weir; 9 is the tracking water head gauge. The picture design is the original concept, through renovations are now underway for gauge modernization.

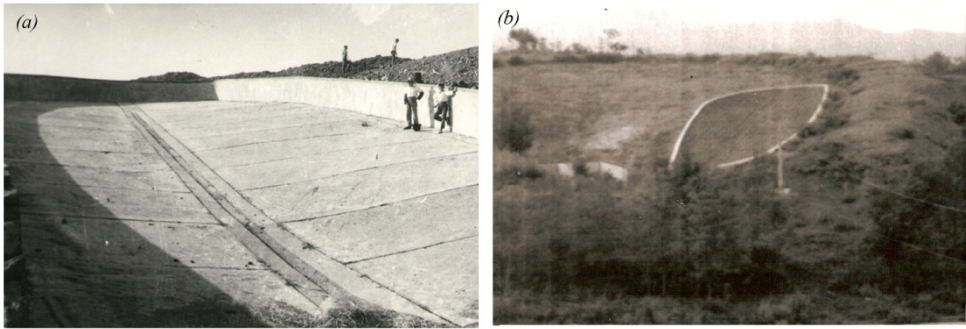
### *The artificial catchment, Hydrohill*

The Hydrohill catchment with a drainage area of 490 m<sup>2</sup> (horizontal projection by plane surveying) - 512 m<sup>2</sup> for its inclined surface [49, 50] - is sited on a small andesitic hill. The entire hillslope was first excavated to bedrock to create a bare catchment of ca 4700 m<sup>2</sup>. A concrete aquiclude was created above it and consisted of two intersecting slopes dipping towards each other at 10° with an overall downslope gradients of 16.9° (Figure 10a). Impermeable concrete walls enclose the catchment on all sides to prevent any flow of water across the catchment boundaries. Silt-loam soil was removed, layer by layer, from an agricultural field near the Hydrohill site and installed on the artificial aquiclude, layer-by-layer again. Also the bulk density of the soil was adjusted during filling to approximate the natural soil profile of the original agricultural field. Hence, the final 1-m soil 'profile' was identical, at least in composition, to the natural soil profile. Grass was then planted over the surface and soil allowed to settle for 3 years (Figure 10b), after which time a central drainage trench was then excavated at the intersection of the two slopes and the water-sampling instrumentation was installed.

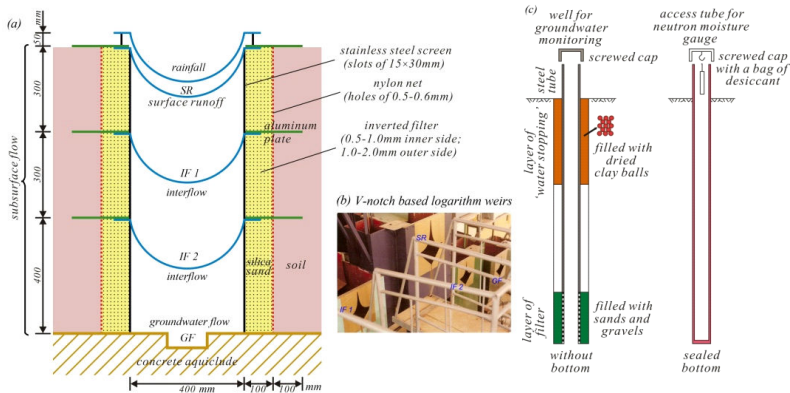
Five fiberglass troughs, each 40 cm wide and 40 m in length, were installed longitudinally in the trench. These troughs were stacked on top of each other to create a set of long zero-tension lysimeters (Figure 11a). Each trough has a 20 cm aluminium lip that extends horizontally into the soil layer to prevent leakage between layers. Water collected in troughs is routed through V-notch based logarithm weirs located in a gauging room (Figure 11b) under the hill where discharge is continuously monitored. Water samples are collected manually above the ponding at the weirs.

As illustrated in Figure 11a, the uppermost trough collects rain; the next lower trough collects surface runoff. The next three troughs collect subsurface flow from soil layers spanning the depths of 0-30, 30-60, and 60-100 cm. These troughs will be referred to as the 30 cm, 60 cm, and 100 cm troughs. The source of the water in these troughs (i.e., whether the water is derived from interflow or saturated flow) varies locally and during storms. The lowermost trough collects either saturated flow or interflow, depending on the height of the water table. When the water table is high, saturated flow may be collected in both of the lower two troughs.

A network of 21 aluminium alloy access tubes for neutron moisture gauges [51], a tensiometer scanner, and 22 wells for water-table measurements and groundwater sampling were installed (Figure 12). The wells were drilled to the aquiclude and are slotted along the lowermost 20 cm. After installation, the spaces around the slotted lengths were packed with sands to allow movement of groundwater to the well, space above the slotted lengths were packed with clay to prevent vertical drainage along the pipes (Figure 11c). The neutron probe access tubes (Figure 11c) and soil water potential tensiometers were positioned adjacent to the wells for soil water monitoring. Catchment evaporation was monitored by methods of water balance, energy budget and, soil water variations above zero flux potential. The plan view of the surface topography, the locations of wells and access tubes, and, the central stacked lysimeter troughs and that of energy budget set are showing in Figure 12.



**Figure 10.** The artificial catchment Hydrohill: (a) Its concrete aquiclude; (b) Three years waiting for the settling of filled soil to try to match close to the natural soil profile.



**Figure 11.** The artificial Hydrohill: (a) Schematic cross-section of rain, surface, and subsurface flow collectors at the catchment; (b) The V-notch based logarithm weirs for discharge measurement for these components; (c) The constructions of wells for groundwater monitoring and access tubes for neutron moisture gauge. (b) and (c) are the original design, now under renovation;

The renovation phase of the main infrastructures for this physically modeled CZEB includes: (1) measurement for energy flux, water flux, geochemical flux, gas flux; (2) changes of watershed storage of energy, water, gas and that of geochemical ions; (3) a small separate laboratory within the gauging room for high-frequency, real time measurement of isotope concentrations in all hydrologic components using of a laser spectrometer LWIA [52]; (4) a removable rainfall simulator on the tracks capable of covering an area of more than 500 m<sup>2</sup>, capable of simulating a range of rainfall intensities; and (5) a system for carbon balance of this block.

## 5. Selected findings

### 5.1. Runoff composition

The catchment response to precipitation consists of surface and subsurface components. These components were directly measured at Hydrohill with both isotopic and hydrochemical evidences (Figure 13) [53, 54]. Surface runoff was not always the dominant component during a rainfall-runoff event. The hydrograph composition of surface and subsurface flows was discriminated into four broad categories (Figure 14): 1) S type which is dominated by surface runoff; 2) SS type which is dominated by subsurface flows; 3) Intermediate type M, with largely equal contributions of surface and subsurface flows; and 4) Variation type V, with switching between components during an event.



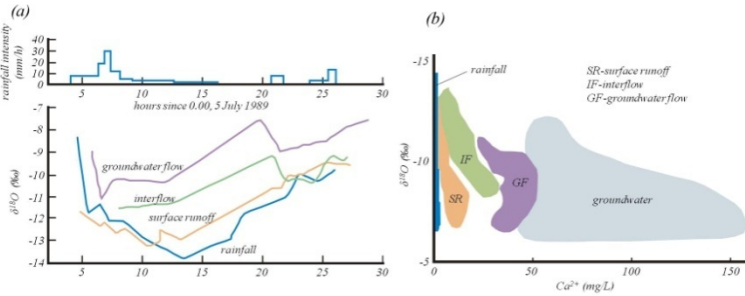
**Figure 12.** Hydrometric monitoring of Hydrohill. (a) Plan view of the surface topograph showing the locations of groundwater wells, access tubes for neutron moisture probe with potential scanner, the central stacked lysimeter troughs, the energy budget set. (b) Whole view of its surface watershed in operation during early years (1980's), under renovation now.

### 5.2. Spatial and temporal variability in hydrological parameters

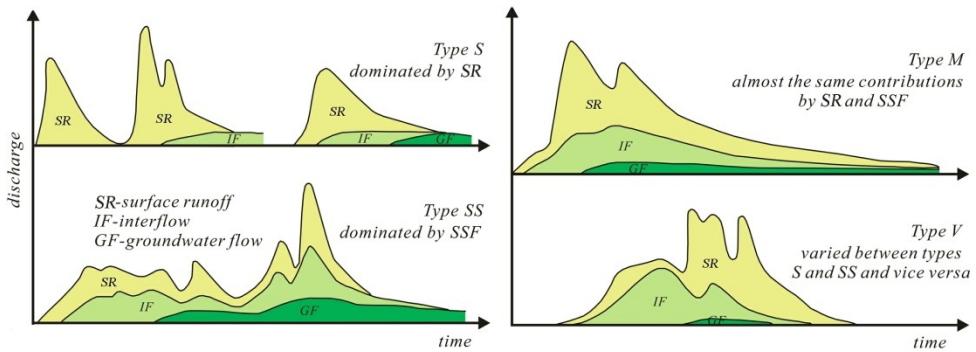
The physical based distributed-parameter modeling appears to be a promising direction for watershed hydrology. However the scale of model elements, and their parameters, is a lingering problem. This has led to investigations into the establishment of a 'representative elementary area' (REP), 1 km<sup>2</sup> [55]. In fact, because of the spatiotemporal heterogeneity of hydrological parameters, little is known regarding hydrologic parameterization in scales less than 1 km<sup>2</sup>. Previously unimagined heterogeneities were observed at Hydrohill:

*Intrastorm isotopic heterogeneity of shallow groundwater.* During a storm in July 1989, the newly developed groundwater showed considerable spatial and temporal variability in the  $\delta^{18}\text{O}$  (Figure 15a), with values ranging from -12‰ to -6‰ while the groundwater derived from pre-event soil water ranged from 0 to 100% at different times and locations in this catchment [56].

The groundwater samples for  $\delta^{18}\text{O}$  were collected at three times during the storm from the wells distributed in whole area as shown in Figure 15a. At the end of the storm, the groundwater  $\delta^{18}\text{O}$  values showed a 4‰ range in composition. This range of compositions during the storm appears to be caused by the combination effects of intrastorm variation in rain  $\delta^{18}\text{O}$  (Figure 13) and spatial and temporal variability in subsurface flowpaths [56] – including the downward displacement of pre-storm water versus delivery of new water to the aquiclude via macropores [54].



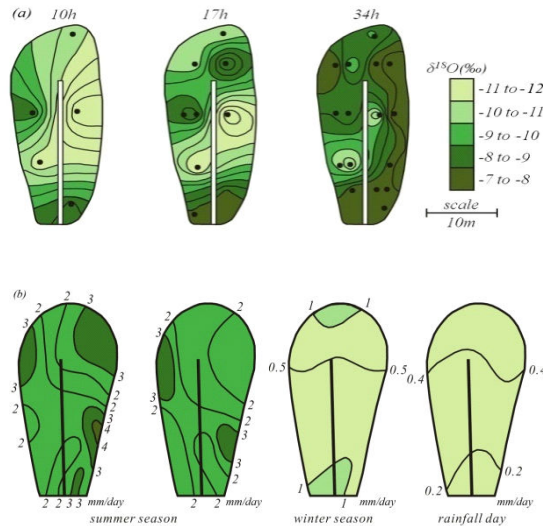
**Figure 13.** Isotopic and hydrochemical evidences of various runoff components. Samples for isotopes were analysed in Menlo Park laboratory of USGS by C. Kendall (same hereinafter)



**Figure 14.** Types of runoff composition summarized from observed data of Hydrohill during 1981- 1992.

*Annual heterogeneity of land evaporation rate* Land evaporation was measured via the variations of unsaturated soil water at different positions in the soil profile during its matrix potential distribution in that profile showing a zero flux plane. Intensive measurements of volumetric soil moisture by neutron probe at 4 points within each of the 21 access tubes (84 in total) were made during the wet and dry seasons. Representative examples of heterogeneity in the daily evaporation rate are shown in Figure 15b [51].





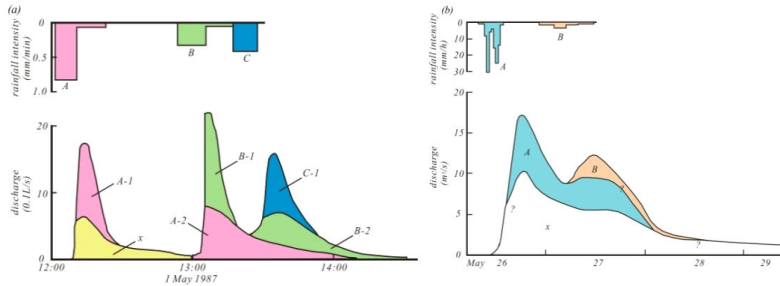
**Figure 15.** (a) Contour diagrams of  $\delta^{18}O$  in groundwater at three sampling times and (b) Typical spatial distribution of the daily land evaporation rate.

### 5.3. The double paradox

About ten years ago, Kirchner described a double paradox in catchment hydrology and geochemistry [31]. His objective was to raise questions rather than provide answers and a worthy challenge was then put forward (i.e., “need to take up the search for a unified theory”, so that the paradoxes “would no longer seem paradoxes”) [31]. For this challenge, perhaps catchments with directly observable surface and subsurface components could be useful in two ways: First, to identify how this double paradox will emerge in the subsurface and surface components individually, instead of in the integrated form at the stream outlet; and secondly, to trace surface and subsurface generation mechanisms using isotopic fingerprints.

*The delayed correspondency phenomena.* Three isolated rainfall events are presented from May 1987 at Hydrohill, and are ideal for establishing rainfall-runoff relationships as well as the unit hydrograph (Figure 16a). In short, the unit hydrography conceptualization is that rainfall event A produces the hydrograph peak A, rainfall events B and C produce the peaks B and C etc. It can be named as the concept of ‘one-to-one correspondency’. However, using isotope tracers, rainfall-runoff events at Hydrohill showed that rainfall event A corresponded to the hydrography peaks A1 and A2, though A2 only emerged later during event B. This pattern continued throughout subsequent storms with event B corresponding to hydrograph peaks B1 and B2, though B2 only emerged later during event C. This was termed as the phenomena of *delayed correspondency* [57]. In the case of Hydrohill, this delay appears very distinct. It is presumed that such a *delayed correspondency* will tend to diminish as drainage area increases. The diminishment of the *delayed correspondency* phenomena has been verified in a natural water-

shed with drainage area of 82.1 km<sup>2</sup> (Figure 16b). Rainfall event A produces a runoff hydrograph peak A, some of the constituents of rainfall event A were delayed to emerge during the rainfall event B. It is worth noting that the hydrograph produced by rainfall event A does not have the bell shape referred to in the unit hydrograph concept. In fact this *delayed correspondency* shows the formation of pre-event water (“old” water) during the runoff process. It follows that the current ‘*one-to-one correspondency*’, used to conceptualize rainfall-runoff relationships in applied hydrology, will be associated with large uncertainty.

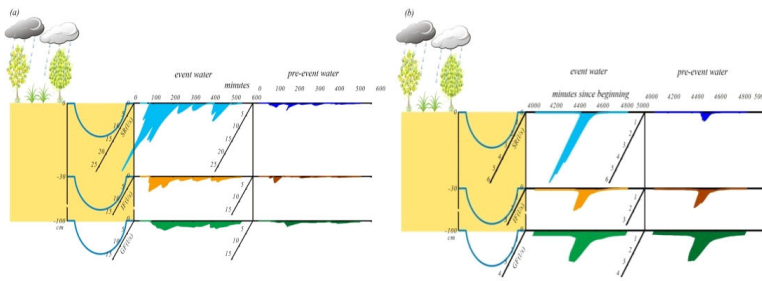


**Figure 16.** The delayed correspondency phenomena.

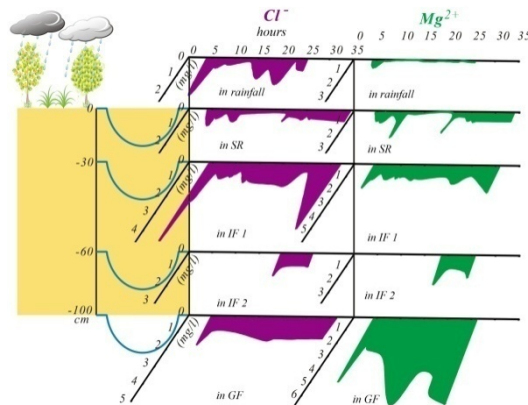
“Old” water paradox identified from both surface and subsurface flows. Data from the ‘mini CZEB’ watershed, Nandadish, suggest that pre-event water (“old water”) appeared in all runoff components including surface runoff, interflow and groundwater or saturated flow (Figure 17). Figure 17a and Figure 17b refer to the surface runoff dominated type (type S, from above), and subsurface flow dominated type (type SS) respectively. The surface runoff and subsurface runoff processes are shown separately with their corresponding proportions of pre-event water. For the type S, the pre-event water accounts for 9% and 24% of the total amount of surface runoff and of subsurface flow respectively while for the type SS, it becomes 11% and 89% respectively [58]. This reveals that even in a catchment with an average soil depth of 2.46 m, large volumes of pre-event water (“old” water) are stored and released promptly by event input.

*Hydrochemistry distribution paradox* This was termed by Kirchner as the “variable chemistry of old water” paradox: although baseflow and stormflow are both composed mostly of ‘old’ water, they often have very different chemical signatures” [31]. However, the artificial catchment at Hydrohill shows paradoxically the distribution of hydrochemical compositions in different runoff components, including event rainfall. This ‘hydrochemistry distribution paradox’ results largely from: (1) inorganic ions in event rainfall input emerge in all runoff components; (2) a strong similarity between rainfall and surface runoff but less similarity in subsurface components; and (3) the fact that the total amount of ions of event rainfall is sometimes much smaller than the sum of all runoff components [59]. Figure 18 shows the processes of Mg<sup>2+</sup> and Cl<sup>-</sup> in event rainfall, surface runoff, interflow and groundwater flow (saturated flow).





**Figure 17.** Processes of various runoff components, and that of their pre-event water of Nandadish. (a) for S type; (b) for SS type



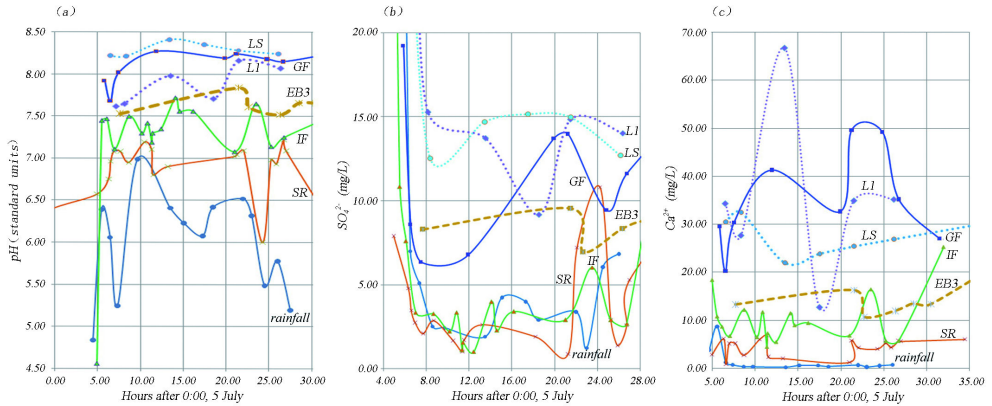
**Figure 18.** The  $Mg^{2+}$  and  $Cl^-$  processes in event rainfall and runoff components of Hydrohill. Samples were analysed in Atlanta laboratory of USGS by N.E. Peters (same hereinafter)

#### 5.4. Does diel signal of hydrochemical constituents emerge linkage among multi-processes?

For the better understanding of the links between contaminants and multi-processes, exploration of diel signals in natural waters may yield “insight into the intricate linkages among hydrological, biological, and geochemical processes [23]”. Diel variations of various hydrological constituents in surface and subsurface runoff responses during rainfall events were monitored in artificial catchments and monoliths with examples as following (Figure 19).

*Variations of pH.* The diel variations of pH in SR, IF, and GF of Hydrohill show their own individualiations (solid lines in Figure 19a). Diel variation curve of SR to a large extent appears similar to that of rainfall. However, pH variation curve of IF is contrary to that of rainfall after 10 a.m., while the GF curve appears as the flattened IF curve. The pH variation curve of the interflow of monolith L1 and that of the flow of monolith LS are reasonably similar to that of

IF and GF of Hydrohill respectively (dot dash lines in Figure 19a). This reveals that the variation of pH in interflow and saturated flow is not driven only by rainfall input but also by “the biological processes of photosynthesis and respiration [22, 60].” This is ascribed to the coupling of the three “reactors” mentioned above via their MC and IMC for transformation and exchange (Figure 4).



**Figure 19.** Diel variations of pH, and hydrochemical constituents  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$  in event rainfall and runoff responses of Hydrohill and monoliths. SR, IF, GF- runoff responses from Hydrohill; L1- monolith of 32 m<sup>2</sup> serves as an element slope of Hydrohill; LS - saturated monolith of 1m<sup>2</sup> serves as solum in reduction environment; EB3- Morning Glory Catchment of 4573 m<sup>2</sup> serves as a catchment without saturated zone formed by debris only.

*Variation of ionic species.* (1) For anion  $\text{SO}_4^{2-}$ : the diel variation curves of runoff responses of both Hydrohill and monoliths, with few exceptions, are contrary to that of rainfall. Mostly their peak concentrations happened during evening and midnight (Figure 19b). Different from the case of pH,  $\text{SO}_4^{2-}$  of SR has a strong inversion with the event rainfall curve at night time. (2) For cation  $\text{Ca}^{2+}$ : the diel curve of rainfall (Fig.19c) shows a small variation after sunrise. However, it triggers variations in runoff responses with their peak concentrations at both a.m. and before midnight. Highest peak happens to L1 at afternoon. These diel variations in runoff perhaps are metabolism related. (3) Nimick et al [60] discussed diel cycles of dissolved trace metal concentration in a Rocky Mountain stream. They found that the anionic species “have their highest dissolved concentrations in the late afternoon” while the cationic species “have their highest dissolved concentrations shortly before sunrise” [23,60].

*Role of soil.* Variations of pH,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$  in total runoff of Morning Glory Catchment, a special designed catchment without soil but debris (EB3 in Figure 19 with broken lines), are very similar to that of rainfall curves after 20:00 (EB3 data are not enough before this time). The role of soil in the formation of ionic species in runoff is apparent. Even the variations of pH,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$  in total runoff of EB3 are triggered by the event rainfall, but the resultant variations appear much simpler than various curves of Hydrohill and monoliths. This implies that only a simple process (i.e., mainly the hydrological process is involved in EB3). So, the complex diel varia-

tions of the runoff responses from Hydrohill and monoliths (Figure 19) can be reasoned as the results of multi-coupling among hydrological, biological, and geochemical processes. It shows that the reactor II (Figure 4) provides a key operator in contaminant hydrology.

### 5.5. Runoff generation

The measurements of various runoff components, within artificial systems containing controlled boundaries, provide the possibility to look inside the formation mechanisms of individual components. Isotopic and geochemical tracing can help to investigate these mechanisms but only if significant differences in isotopic compositions of components occur. The general mechanisms for these runoff components (i.e., the surface runoff, interflow and groundwater flow (saturated flow) ) are discussed in the following paragraphs [61,62].

*Surface runoff (SR).* Precipitation input is, of course, the essential condition for the generation of surface runoff. However, in order to actually generate runoff, there must be enough precipitation to form a thin, saturated soil layer ( $L_{sat}$ ) at the ground surface. Saturation is key because unsaturated water movement is thought to be too slow to generate runoff. The thickness of  $L_{sat}$  at catchments, monoliths and plots was found to vary between 5 to 50 mm throughout events, increasing downward during rainfall events and receding once rainfall stopped, although these findings were complicated by the irregularities of the soil surface. SR was not generated until a  $L_{sat}$  was established at the surface. Once a  $L_{sat}$  was developed, regardless of its thickness, SR was generated immediately. *Overland flow* was only observed on impermeable surface (*DO*) at artificial plot and, on saturated surface (*SO*) from a special lysimeter designed for  $L_{sat}$  simulation. Intrastorm variation of isotopic composition of *DO* and *SO* can match that of event rainfall. In most cases however, SR is generated within the  $L_{sat}$ , with turbulent mixing of event and pre-event water stored in  $L_{sat}$ , i.e., the *saturated mixing surface flow (MS)*; Alternatively, small amounts of event water act on the surface of  $L_{sat}$ , to force out pre-event water in the  $L_{sat}$ , termed here as the *saturated expelled surface flow (ES)*. The isotopic composition of *MS* shows a mixing of event rainfall and pre-event water in  $L_{sat}$ . The isotopic composition of *ES* is similar to that of  $L_{sat}$ .

*Interflow (IF) in the unsaturated zone.* Three generation mechanisms are observed. (1) In cases there are soil layers with distinct bulk density and/or hydraulic conductivities, *IF* can be generated at the interface of soil layers. This was only observed in the 'mini CZEB', Nandadish, where *IF* occurred at the interface of the layers A (topsoil) and B (subsoil). This is termed as *layered interflow (LI)*. (2) During percolation, soil water moves downward and laterally towards the drainage interface and accumulated until saturation. The saturation will expand if the soil matrix potential  $\psi < 0$ . Once  $\psi$  reaches zero, the accumulated saturated soil water will be released immediately at drainage interface. A small amount of infiltration water can trigger this process, to expel the accumulated soil water out as interflow. This process was observed for most *IF* events in Hydrohill and the 32 m<sup>2</sup> monolith. This is termed as *expelled interflow (EI)*. (3) *Macropore interflow (MI)* was also observed in a special designed catchment without saturated zone with area of 4573 m<sup>2</sup>.

*Groundwater flow (GF) from saturated zone.* In most cases, groundwater flow was due to a rising groundwater table resulted from event water recharge via the capillary fringe zone, and was

termed *recharged groundwater flow (RG)*. RG was observed in most cases of both artificial and natural catchments. In addition, the *macropore-induced groundwater flow (MG)* is the only component not directly observed in this work but inferred from the isotope data in catchment GF during events.

## 5.6. Unreasonableness of current two-component isotope hydrograph separations

Hydrograph separation using conservative, two-component mixing models has been done in various natural basins. However, data from these two catchments, including both the natural and artificial systems indicate that this technique is unacceptable for natural basins and artificial catchments, yielding misleading results due to the unreasonableness of most of the assumptions involved and of the unrealistic operation procedures [63].

*Violation of assumptions.* In studying natural processes it is inevitable to make simplifying assumptions. However, as Kennedy et al [64] indicated, when simplifying assumptions are made, one runs the risk of drawing misleading conclusions. That may have been the case for the five assumptions [65] summarized for the use of isotopic techniques in precipitation-runoff studies.

The first assumption, that “Groundwater and baseflow are characterized by a single isotopic content” [65], is problematic. Baseflow in natural basins can be recharged by the active zone, passive zone, each with differing flowpaths for recharge resulting in differences in the isotopic composition of baseflow itself, which changes with time and discharge. It is also unreasonable to expect a single isotopic content for groundwater which is subject to both temporal and spatial variations (Figure 16).

The second assumption, that “Rain or snowmelt can be characterized by a single constant isotopic composition or, the variations are documented” [65], is problematic, because, as shown in Figure 14a the intra-storm variability of isotopic composition of rainfall is very significant. High isotopic variability has also been observed in experimental watersheds at Maimai, Panola, etc [56]. Additionally, the spatial variability of isotopic composition of rainfall in a watershed exists and shouldn't be ignored. Isotopic data from a 82.1 km<sup>2</sup> watershed showed that the largest difference in rainfall  $\delta^{18}\text{O}$  reached 8.2‰.

The third assumption, that “The isotopic composition of rain water is significantly different from that of groundwater/baseflow” [65], can be true. However it must be demonstrated by appropriate sampling [64].

The fourth assumption, that “Contributions from soil water are negligible, or the isotopic composition is identical to that of groundwater” [65] is problematic, because it is a misconception. In a natural watershed, this can only occur if there is no unsaturated zone.

The fifth assumption, that “Contributions from surface water-bodies (such as ponds) are negligible” [65], is reasonable for most upland watersheds [64]. However, in catchments of hilly area with land use including series of ponds and paddy fields, the contributions are large enough to significantly influence results [62].

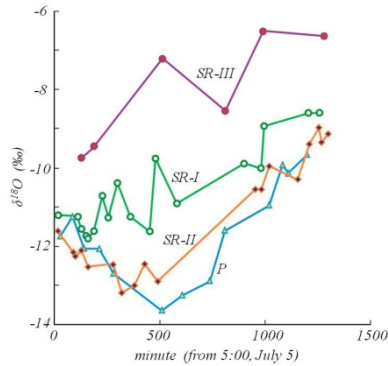
Based on such problematic assumptions, one of the basic conclusions that “most stormflow is old water” [66], which resulted from two-component hydrograph separations in multiple catchments with different drainage areas, appears suspect.

*The physically ambiguous ‘old’ and ‘new’ water.* This two-component separation model in fact is based on the classic Horton infiltration theory with a very simple runoff generation concept that the soil surface acts as a sieve capable of separating rainfall into two basic components [67]. This ‘old’ and ‘new’ water separation model has led to many usages with a variety of defined components in addition to ‘old’ and ‘new’ water component of flow, e.g., ‘pre-event and event component’, ‘pre-storm and storm component’, ‘pre-storm water and rain in storm runoff’, ‘pre-event and rainfall water’, ‘groundwater component and event water’, ‘groundwater and rainwater’, ‘groundwater discharge and surface runoff’ [68], ‘surface and subsurface runoff’ [69]. The classic two-component method has also been extrapolated to incorporate three components (e.g. that of channel precipitation, soil water and groundwater [70]). Thus, it seems that success can be assured because there are no calibration constraints. As Kirchner [15] noted some models are “often good mathematical marionettes, they can dance to the tune of the calibration data”.

The ‘old’(O) and ‘new’(N) water actually correspond to different generation patterns of runoff components. In fact, multiple runoff mechanisms can result in the same proportions of old versus new water, a problem known as equi-finality. As seen in Hydrohill, surface and subsurface runoff patterns emerge from multiple runoff mechanisms (e.g., macropore flow can result in a large proportion of event water or a large proportion of pre-event water depending on antecedent wetness in the catchment). The runoff components corresponding to old water and new water can be very ambiguous. Applied to surface and subsurface runoff [68, 69], surface runoff is labeled new water [62]. This is a misconception as shown in Figure 20.

*The mass balance equations for this model are untenable.* The two-component tracer mixing model is stated by two mass balance equations for the composition of stormflow at any time which is used for hydrograph separation:  $Q_s = Q_n + Q_o$  and  $Q_s \delta_s = Q_n \delta_n + Q_o \delta_o$  where  $Q$  is streamflow,  $\delta$  (Delta) is the D or  $^{18}\text{O}$  content, the subscripts s, n and o represent the stream, new and old respectively [56]. At any time  $t$ , the first equation is correct (i.e., discharge  $Q_{s(t)}$  at the outlet of a watershed is equal to the sum of the unknown  $Q_{n(t)}$  and  $Q_{o(t)}$  right at the same outlet at the time  $t$ ). During time  $t$ , the  $Q_{s(t)}$  at the outlet and its isotopic concentration  $\delta_s$  are measurable and are the value of known without problem, however both the  $Q_{n(t)}$  and  $Q_{o(t)}$  at the outlet of time  $t$ , are the results of confluence from somewhere within the watershed before time  $t$ , i.e., the results of a convolution integration. The  $Q_{n(t)}$  and  $Q_{o(t)}$  at the outlet of time  $t$  are concentrated from the separated isochrone area somewhere within the watershed with different time of concentration. So, the second equation with algebraic sum is physically unrealistic, hinging on the assumptions outlined above. For this linear algebraic sum to be physically realistic, the isotopic concentrations in the reservoirs of new and old water must be constant, without mixing or fractionating during the time interval of event hydrograph. To be physically realistic and representative of the mixing and non-conservative nature of water in natural catchments, it would likely take on the form of an integral or a finite-difference isochrone [63]. No affluxion

happens to both the new and old water in the watershed. This can only happen in a small pond, operationally for a watershed, it's "the Procrustean bed [17]".



**Figure 20.** Intrastorm isotopic variations of an event rainfall, and isotopic variations of observed surface runoff in different watersheds. P–rainfall, SRI–surface runoff of the 'mini CZEB', Nandadish, with drainage area of 7897 m<sup>2</sup>, SRII–surface runoff of Hydrohill of 490 m<sup>2</sup>, SRIII– runoff of Morning Glory Catchment of 4573 m<sup>2</sup>.

## 6. Conclusion

The river basin, watershed or catchment is a central concept in hydrology. Basin studies to assess watershed hydrology are approaching a period of transition and innovation. In fact, experimental watershed studies, the core of watershed hydrology, have tremendous and complicated challenges ahead.

The current challenges in basin experimental hydrology are mainly twofold. Advancing hydrologic science creates a fundamental challenge. Because the watershed system is a dynamic ecological system composed of a variety of biotic and abiotic processes driven by water and climatic processes, experimental watersheds should multi-couple these processes, organizing innovative measurements and approaches while continuing to support and test hydrological models. Anthropogenic hydrologic replumbing and natural climate oscillations are equally challenging. Field studies are the key to understanding and modeling the effect of hydrologic replumbing and climate change. There is a need for long term monitoring, systematic experimental facilities as well as data mining to reclaim historic data useful for determining baseline watershed metrics.

The main problems of the historical experimental basin approach are threefold: watershed surface laterality, hydrologic process laterality and downward components laterality. Lessons from fifty years of Chinese experimental basin studies are: (1) research facilities require natural conditions and also artificial controlled boundaries; (2) address not only the surface watershed, but also downward to the bedrock; (3) surface and subsurface runoff components should be

directly monitored hydrometrically; (4) isotopic and hydrochemical tracers are key to understanding runoff generation mechanisms; and (5) account for interactions between the hydrosphere and multiple watershed processes.

Another kind of experimental basin is suggested going forward, namely the Critical Zone Experimental *Block* (CZEB), geologically a monolith-block within the Critical Zone. CZEB is a dynamic ecological and evolving system, coupled with various systems and united by hydrological process. The CZEB is a natural open system; both the energy and mass exchanges exist across its boundaries. It is a dissipative complex system with some degree of self-organization. The function of CZEB is threefold: mass, its material aspect; energy and force, its driven aspect and, organization and entropy, its thermodynamic/philosophical aspect.

To advance watershed hydrology and support development of a unified theory of hydrology, a two-way multi-scale experimental watershed system is suggested, including the natural system and the artificial system. Both have multi-scale subsystems from monolith, slope, sub-watershed to watershed and follow the research idea of upwards and downwards routes. A trial for such a strategy is partly completed at the Chuzhou CZEB Experimental System.

To advance contaminant hydrology, the suggested two-way multi-scale experimental watershed system may provide a key to unravel the complex mechanisms coupling hydrological, biological, and geochemical processes. Contaminant transformation and fate and their effects on regional degradation of groundwater basin involve highly complex mechanisms. This two-way multi-scale system calls for new models using both natural and artificial experimental basin results and using upwards and downwards approaches to multidisciplinary techniques, including isotope tracing.

All hydrological knowledge ultimately comes from observations, experiments, and measurements [15]. Progress in hydrology results mainly from challenges to prevailing approaches and concepts [71]. Hydrological experimentation, including CZEB experimental watershed studies, is the building block for development of a unified theory of hydrology including contaminant hydrology. However, it is important to remember Werner Heisenberg's warning that, "what we observe is not nature herself, but nature exposed to our method of questioning".

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