Tracers in Hydrology

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To

Fritz Gygax, University of Berne, Switzerland
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Ingeborg Vonderstrass
A personal dedication to my wife for giving me the scope for research and for the substantial support of my professional work
(Christian Leibundgut)

Contents

Preface

'Tracers in Hydrology' has a long history. In the 1950s the use of tracer techniques in hydrology began to be developed broadly. This development was possible, in particular because of the progress made in measurement techniques and by the digitalization of data processing. Simultaneously, the computer era began and opened up new possibilities for environmental modelling. During this fascinating phase of development of natural science and, in the case of Tracerhydrology, came the evolution of a holistic approach towards the use of tracers in hydrology.

Besides many other factors, three fortunate milestones marked this development. A powerful framework for realizing numerous ideas was created through the founding of the Association of Tracerhydrology (ATH). The ATH promoted the use of tracer techniques in Europe between the 1960s and the end of the twentieth century in many ways.

The second milestone was the establishment of the Isotope Hydrology Laboratory by the International Atomic Energy Agency (IAEA) in Vienna in 1961. It pushed the rapid development of the isotope techniques, beginning with environmental tritium, as a research tool for investigating the hydrological cycle worldwide.

The XXth General Assembly of the International Union of Geodesy and Geophysics in 1991 in Vienna can be considered to be the third milestone. The International Commission on Tracers (ICT) within the International Association of Hydrological Sciences (IAHS) was established at this assembly. Its aim was, amongst others, to bring together the experimental hydrologists with modellers for the integrated investigation of the hydrological system. This event is significant since at the zenith of the modelling phase in hydrology the establishment of a clearly experimentally oriented commission within the IAHS was not without opposition. The following years showed an increasing integration of the tracer methods into hydrological research and applied hydrology by the international community, which validated this structural development. Experimental hydrology, in particular the strongly emerging catchment hydrology, used tracer methods increasingly in order to assess hydrological processes and system functions. In particular the calibration and validation of mathematical models was based increasingly on tracerhydrological research.

The authors are looking forward to the further development of new applications of tracer techniques and, in particular, to an increasing combination of tracer techniques with other hydrometric and hydrological methods. In order to provide independent, experimentally based hydrological data for the reliable modelling of hydrological processes and systems, the further methodological development of tracer methods is expected.

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1
Introduction

'Tracers in Hydrology' defines the scientific field that aims at *understanding the hydrologic system* by making use of environmental and artificial tracers and modelling. Tracing of water provides unique methods for a direct insight into the dynamics of surface and subsurface water bodies. The relevance of tracer techniques in hydrological investigations and in applied hydrology ensues from the astounding complexity of water flow in natural systems. How much runoff in rivers really stems from rainstorms? How does water flow through a hill-slope or a glacier? How large is the storage of water resources in aquifers? Where, how and when was the water found in an aquifer formed? Tracer techniques are a useful tool in understanding the transport processes and quantifying their parameters. Tracers help to identify and quantify the phase changes (evaporation, condensation, sublimation), shed light on the origin of pollution and assist in the respective remediation processes. The natural tracers constitute a tool of prime importance in the reconstruction of the climate during the Holocene period when studying ice cores, old groundwater and the unsaturated zone in arid and semiarid regions. Tracer methods are also a major tool for calibration and validation both of strategies in modelling catchment hydrology and hydrological models of groundwater systems. Furthermore, tracer approaches are commonly used to address issues like surface water–groundwater interactions, paleohydrology, water movement in very low permeable rocks, calibrating and validating numerical flow and transport models and evaluating vulnerability of water resources. Finally for Integrated Water Resource Management, tracer techniques have great potential as tracers that provide integrated information and can be very efficient in characterising complex systems in remote areas.

The empirical observation of flow and transport processes with tracers and the theoretical formulation of flow and transport processes depend on each other and have resulted in a beneficial coevolution of both approaches if adequately combined. Tracers provide empirical data of real and often unexpected flow patterns – models provide tools for flow and transport predictions.

The term '*Tracerhydrology*' is used as a short expression for the use of tracers in hydrology understood as an advanced method that allows for an integrative investigation

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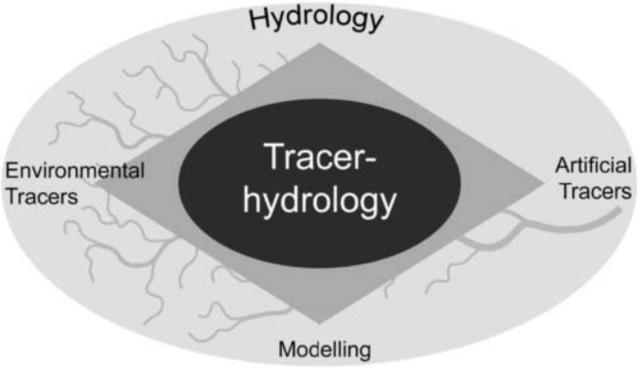

Figure 1.1 Tracerhydrology as a method of application tracers in water sciences understood as a holistic approach of hydrology and water research.

of the hydrologic system. It is not regarded merely as an isolated technique for solving particular problems of applied hydrology, although it certainly can be useful in those fields.

This book originated from the idea of interweaving knowledge from the fields of artificial and environmental tracers and of modelling, in order to present the options, opportunities and limits of tracers in hydrology to students, scientists, engineers and other users. In the following chapters the explanation of tracer and modelling basics builds a foundation for students and users to be able to understand the techniques, which are then applied to case studies of both specific applications and integrated studies. Herein, tracer techniques are described with regard to their relevance for advancing hydrological science and to their role in solving problems in applied hydrology. Students, scientists and consultants will find a wealth of information on tracers and modelling in order to introduce them to the field of tracerhydrology. A methodological chapter provides specific techniques such as the calculation of injection mass and the chloride method and also case studies dealing with the different approaches and problems of applied tracerhydrology (groundwater recharge).

Scientists can see the range of opportunities that tracer techniques offer through the variety of comprehensive case studies that are presented. Engineers and other users will find a large collection of work examples and may apply the methods described, for example tasks in integrated water resources management or the allocation of water supply protection zones, as well as many others. In this book the application of tracers in hydrology is understood basically as the integrated use of tracers in hydrology and therefore as a part of an integrated hydrological approach (Figure 1.1).

In Chapter 2 a detailed concept of tracerhydrology will be presented. The role of modelling in integrated tracerhydrology will be defined in a separate chapter. The combined application of tracerhydrology and modelling is presented by means of selected examples of applications in various hydrological compartments (glaciers, rivers, lakes, groundwater). The authors wish to present a textbook that starts from a simple and general overview and moves on to the more complex topics of tracerhydrology in order to facilitate an easy understanding by the readers, be they students, water research scientists, engineers or applied hydrologists.

The application of tracers in hydrology has a long tradition among the geo- and water sciences. After what were at first somewhat 'trial and error' – based experiments about 150 years ago a fascinating development began. Artificial salt and fluorescence tracers have been used for decades. In the 1950s a wide variety of new artificial tracers were included in tests designed to trace water, mainly in karst aquifers. At the same time a compelling new direction in tracerhydrology based on the use of natural, mainly isotopic tracers began to develop. Most of the fundamental principles had been developed during this phase. Stable isotopes have provided a major input into the study of hydrological processes such as runoff generation and runoff component separation as well as recharge and groundwater flow and are still at the centre of defining the conceptual models of hydrological processes. The role of isotopes in the validation of circulation models and response of ecosystems to climate change is not yet fully explored.

In addition to an increasing number of papers on tracerhydrology published in international hydrological journals, there are many publications on the use of tracers for water research issued by international organizations, such as (i) the IAHS (International Association on Hydrological Sciences), (ii) the symposia proceedings of the IAEA (International Atomic Energy Agency), and (iii) the proceedings and project volumes of the ATH (International Association of Tracers). These publications are an excellent resource for all matters concerning the methodological aspects and application of tracers.

Comprehensive presentations of large combined tracerhydrological studies are given in the reports of the ATH (International Association of Tracers). The focus of these investigations was on groundwater systems but the approach was holistic within the respective river basins. Increasingly, investigations on runoff generation and catchment modelling have adopted an integrated tracerhydrological approach.

Innovations in analytical techniques will provide new tools for tracerhydrology. There are trends towards reducing sample volumes, increasing the number of samples analysed, reducing detection limits and identifying new natural and industrial substances that can be used for tracer studies. Certainly, natural remediation and reactive transport processes will be explored increasingly with tracers. For the advance of hydrological science, empirical data provided by tracer methods have and will continue to play an important role. Further integration of experimental and theoretical approaches leading to an integration of tracers into soil water atmosphere transfer schemes and catchment and groundwater models, will provide additional means of validating the hydrological concepts.

2

The Integrated Concept of Tracers in Hydrology

2.1 System approach

The system analysis of watersheds and aquifers draws key insights from artificial and environmental tracer data. Artificial tracers help to understand flow processes, to estimate main hydrological system parameters and to visualize the movement and mixing of otherwise indiscernible distinct water volumes. Hence, they provide a tool for understanding and characterizing complex flow through the soil, on surfaces, in channels, through and along hill-slopes, in aquifers or in artificial systems. Environmental tracers have become key tools for estimating water resources in the catchment areas, for the reconstruction of hydrological processes from the past, in ungauged basins or for the integration of hydrological processes that otherwise would be far beyond observation. Both environmental and artificial tracers have their own theoretical basis. This textbook will provide an introduction to both environmental and artificial tracer techniques along with their respective theoretical background and will demonstrate how both techniques can be modelled, combined and integrated into hydrological applications that work.

When trying to analyse hydrological systems' hydrometric data, hydro-chemical information and system characteristics need to be reconciled within a common system model (Dyck and Peschke, 1995). The aim of tracerhydrology is to develop, test and validate those representations of the hydrological system that best agree with the available data by making use of environmental and artificial tracers and modelling.

The general approach in system hydrology is based on the determination of:

- a known or measured input (volume, concentration, energy) as a function of time and space,

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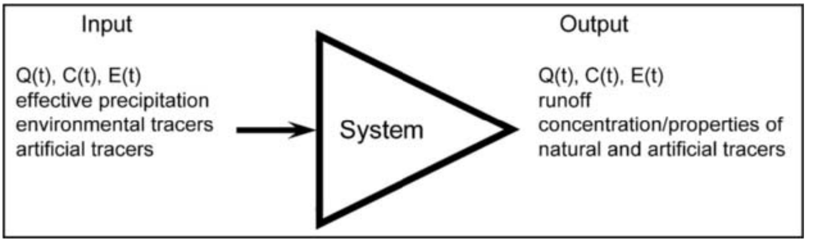

Figure 2.1 Hydrological system approach adapted to tracerhydrology by the convergence approach. Q: volume of water; C: concentration; E: energy.

- a function characterizing the system (e.g. catchment, spring. . .) by a set of equations describing the flow and/or transport processes at the atmosphere-surface boundary, in surface water or in subsurface-water,
- a known or measured output of the same parameters as a function of space and time (Figure 2.1).

Linking this to tracer techniques, the input will be the concentration of tracer in infiltrating water and effective precipitation (for environmental tracers) or the injection mass of artificial tracers. After flowing through the system the output will be characterized by the runoff volume, the environmental tracer concentrations (for isotopes and geochemical compounds) and/or artificial tracer concentrations. This concept has been described by Leibundgut (1987) and named the convergence approach. In other words tracerhydrology is based on decoding of information contained in the output parameters of a system. The simplest example is the system of a spring.

Both input and output parameters will be measured in order to understand the processes in a natural hydrological system be it a catchment, an aquifer or surface water. Besides the hydrological water balance parameters in particular, data of environmental and artificial tracers are measured. Models are simplified abstractions of nature that are used to obtain information from measured data about the system. The transfer function between input and output is identified from tracer data and can be used for predictions or system characterization. The modelling of both environmental tracers and artificial tracer experiments is a necessary tool for evaluating the application of tracers.

The application of the convergence approach in tracerhydrology can be used to derive concepts of hydrological systems (Figure 2.2). These conceptual models can be simple or more structured. They represent the principal functioning of the investigated hydrological system (Leibundgut, 1987; Attinger, 1988). Predictions derived from an existing, conceptual system model allow for an improved design of the experimental planning and the observation network.

The combined and simultaneous use of several independent methods and techniques in investigating a hydrological issue is considered as an axiom of tracerhydrology. This principle is applied using the different tracer techniques (natural, artificial tracers)

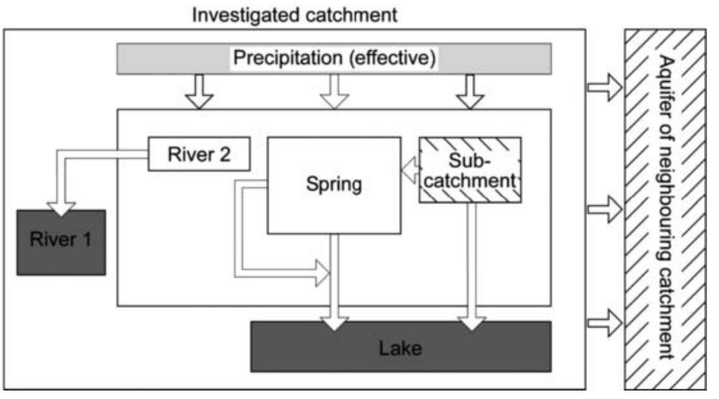

Figure 2.2 Conceptual model (Structure model) of a complex system (catchment) evaluated by tracer techniques serving as a base for further research and mathematical modelling (see colour plate section P3b-d.

in combination with independent hydrological methods. First, this means that several techniques should be combined in multi-tracer experiments, if possible. The combination of different tracers ensures that the specific limitations of single tracers or methods do not bias our understanding of the hydrological system. While it has become almost common practice to combine different artificial tracers, the combination of environmental *and* artificial tracers is the most promising approach. Furthermore, tracer methods should also be combined and integrated with other hydrological and scientific methods (hydrometry, geophysics, hydrochemistry, remote sensing, etc.).

The fascination of an integrated approach is the reconciliation of results obtained by different independent methods. If different methods provide consistent or concordant data, the scientific conclusion is more soundly based and validated. The fuzziness of individual methods can be overcome if different methods point in the same direction. Finally, contradictions between different methods can be very instructive and push for new experiments or research aimed at resolving the problem.

2.2 Definition of tracers

Environmental tracers are defined as inherent components of the water cycle. Sometimes, accidental injections can be used for hydrological studies. Global input functions have been created as the side effects of industrial or military activities (CFCs, ^{85}Krypton or bomb-tritium). Artificial tracers are defined by their active injection into the hydrologic system in the context of an experiment.

In nature tracers are widely used as markers, such as wildlife marking their territory or ants using pheromones for marking itineraries. Such markers are effective at extremely low concentrations (10^{-15}). All tracers carry discernable and preferably unique information. These two properties – carrying information that can be identified most effectively at low concentrations – categorize substances as trace elements.

Table 2.1 Systematic tracer classification, distinguished by their application. Pollution tracers originate from anthropogenic activities, however their input in the hydrological system can be similar to that of natural tracers

Environmental tracers	Artificial tracers
Utilization	*Application*
Environmental isotopes	Chemicals
Hydrochemical substances	Biological substances
Pollution tracers	Drift substances
Characteristics:	*Characteristics:*
Spatial input via precipitation, geogenic sources	punctual input (injection), defined by time, place, hydrological situation
Pollution tracers (e.g. Cl^- SF_6 CFCs)	

Hydrological tracers are dissolved, suspended or floating substances according to their purpose and field of application. Some natural and artificial substances which are suitable for scientific studies or can be applied for the investigation of hydrological systems and subsystems are given in Table 2.1 (Leibundgut, 1982). In principle, hydrological trace elements have to be detectable in solutions with mass ratios of water:tracer of $>10^9$.

Environmental tracers are inherent components of the water cycle, thus we speak of their *utilization*, while artificial tracers are brought actively into the hydrologic system, so that we refer to their *application*. Not belonging completely to either of the two groups, pollution tracers are substances introduced into the water cycle by anthropogenic activity, coming either from punctual contaminations such as waste deposits or brought in by accidents, or originating from the production of pollution gases released into the atmosphere. Consequently, they are not natural but feature the same input channels as environmental tracers.

The input of environmental tracers into the hydrologic system of surface, soil and ground water takes place by diffuse and continued processes via precipitation or the solution from minerals. An investigation with large scales of time and space is possible, and thus environmental tracers serve, in particular, to follow an integrated approach, for example not only catchment studies and water balances but also as a base for solving various applied problems. Often, the variability in time and space of the input function is hard to acquire, and the input 'signal' might be weak.

Artificial tracers are added to the system in well-defined hydrologic situations of time and space, by punctual injections; it is possible to label a specific component of the water cycle or investigated system, for example an inflow to the lake. The scales in time and space for application are limited, and it is only possible to gain insights into a part of the system during the time of the experiment. In general, artificial tracers are used in systems which have a residence time smaller than one year. Owing to this limitation it might happen that the hydrologic situation chosen for the experiment is not representative for the system. A list of available tracers in three groups is provided in Table 2.2.

Table 2.2 Currently available hydrological tracers.

	Artificial tracers	
Environmental tracers	Solute tracers	Dissolved gas tracers
Stable isotopes	**Fluorescence tracers**	Helium
Deuterium (^{2}H)	Naphtionate	Neon
Oxygen-18 (^{18}O)	Pyranine	Krypton
Carbon-13 (^{13}C)	Uranine	Sulfur hexafluoride (SF_6)
Nitrogen-15 (^{15}N)	Eosine	
Sulphur-34 (^{34}S)	Rhodamines	
Radioactive isotopes	**Non fluorescent dyes**	
Tritium (^{3}H) (and Helium-3 (^{3}He))	e.g. Brilliant Blue	**Particulate tracers**
Carbon-14 (^{14}C)	**Salts**	Lycopodium spores
Argon-39 (^{39}Ar)	Sodium/potassium chloride	Bacteria
Krypton-85 (^{85}Kr)	Sodium/potassium bromide	Viruses
Radon-222 (^{222}Rn)	Lithium chloride	Phages
Radium-226 (^{226}Ra)	Potassium iodide	DNA
Silicium-32 (^{32}Si)	Sodium borate (borax)	Synthetic microspheres
Chlorine-36 (^{36}Cl)	**Fluorobenzoic acids**	Phytoplankton
Noble gases	**Deuterated Water (^{2}H)**	
Geochemical compounds	**Radionuclides**	
e.g. Silicate, chloride, heavy metals	e.g. Tritium	
Physio-chemical parameters	Bromide-82 (^{82}Br)	
e.g. Electrical conductivity, Temperature		
Pollution Tracers		
e.g. CFCs, SF_6, phosphate, boron, nitrate, radioactive compounds		

2.3 Modelling in the context of integrated tracerhydrology

Finding the parameters from the tracer experiment is only possible if an adequate mathematical model is used, meaning that the model is based on the proper concept of tracer transport and its behaviour in the system. In order to understand this, some definitions will be given below and the application of the mathematical models will be discussed (Maloszewski and Zuber, 1992a, b; 1993).

A *Conceptual model* is a qualitative description of a system and its representative factors (e.g. geometry, hydraulic connections, parameters, initial and boundary conditions) related to the intended use of the model. In practice, the conceptual model demonstrates the principal idea of water circulation in the system (Figure 2.2).

A *Mathematical model* is a mathematical description of a conceptual model, representing a hydrological, physical and/or hydro-chemical system, using functions designed to help in understanding and predicting the behaviour of the system under specified conditions. In tracerhydrology the mathematical model represents the solution to the mathematical equation(s) describing water and tracer transport for given boundary conditions.

Model calibration is a process in which the mathematical model assumptions and parameters are varied to fit the model to the experimental data. Calibration can be carried out by a trial-and-error procedure, or by an automatic fit based on an objective function. The calibration of the model to experimental data solves the inverse problem by finding the right values for system parameters.

Model validation is a process of obtaining assurance that a model is a correct representation of the process or system for which it is intended. Ideally, validation is obtained when the parameters derived from the model agree with independently measured parameters (e.g. porosity) as described above.

The tracer method is usually applied to a system that is poorly known. As a consequence the mathematical model required to determine the system parameters must be as simple as possible. As mentioned earlier, mathematical modelling of experimental data in tracer hydrology can be separated into two different approaches, depending on the considered tracer method. The two approaches are i) deconvolution or inverse modelling of information provided by tracers and ii) mathematical modelling based on the transport equation.

In general, the tracer injection for artificial tracer experiments is reduced to a single point only (well, sinkhole, karst doline) or to a line (trench, river cross-section). Some pollution tracers also rather represent point sources (e.g. pollutants release by accident or from point sources). In this case mathematical models are used that are usually based on dispersion theory. Analytical solutions for advection-dispersion processes in one, two or three dimensions and different boundary conditions are available and described in detail in Chapter 5. For heterogeneous systems and complex boundary conditions, transport equations can be solved by numerical schemes.

For the modelling of environmental tracer data, a quite different approach is needed. In general, the 'injection' of tracer occurs naturally over an area and during a longer time either by precipitation or by solution of minerals from earth substrate. For instance, in the case of stable isotopes of water the tracer enters the hydrological system by precipitation that infiltrates. The environmental tracer concentration is observed in places where water discharges (e.g. in a river, at springs, at a pumping well).

Knowing both input and output concentrations as a function of time, one can consider the aquifer as a 'black-box'-system. Often this system can be described by mean parameters (volume of water, transit time and flow rate through the system). In this case, tracer transport between input and output (Figure 2.1) can be described by a lumped-parameter approach. Transport of tracer between input and output is characterized by the transit time distribution function, which needs to be defined for the investigated system (Maloszewski and Zuber, 1982, 1985). In mono-porous media, where the portion of stagnant water can be neglected, this type of modelling yields the mean transit time of water in the system as the main parameter. This parameter can be further used to estimate the volume of water in the system and thus the available water resources. In strongly heterogeneous (so called double-porous) media, for example fissured aquifers, which consist of mobile and stagnant water, application of the lumped-parameter approach to environmental tracer data yields the mean transit time of tracer instead of the mean transit time of water. The transit time of tracer describes both the transport of tracer by a mobile water component and the diffusive exchange of tracer between mobile and stagnant water. A detailed description of the lumped-parameter approach for mono- and double-porous media is given in Section 5.2. The transit time

Table 2.3 Main fields of applications in tracer hydrology

	Atmospheric water	Surface water	Soil and unsaturated zone	Ground-water	Glacier and Snow	Catchment hydrology	Special application
Global circulation	+					+	
Discharge measurement		+		+	+	+	
Delination of hydrological units and protection zones			+	+	+	+	
Hydrologic/hydraulic connections			+	+	+	+	
Evaluation of flow paths			+	+	+	+	
Altitude of source areas				+	+	+	
Age dating		+	+	+	+	+	
Experimental hydrograph separation				+	+	+	
Runoff generation processes			+	+	+	+	
Residence times		+	+	+	+	+	
Flow and transport parameters	+	+	+	+	+	+	
Dispersion and diffusion processes		+	+	+	+	+	+
Mixing processes	+	+		+		+	+
Permeabilities			+	+	+	+	
Infiltration processes			+			+	+
Infiltration/exfiltration processes		+	+	+	+	+	+
Groundwater recharge		+	+	+	+	+	
Interaction between surface and subsurface water		+	+	+		+	
Hyporheic exchange		+	+	+		+	+
Filtration processes		+	+	+			+
Stratification of lakes		+					
Circulation currents		+					
Contaminant transport	+	+	+	+		+	+
Engineering hydrology		+		+			+

of water in double-porous systems can be derived if the porosity of the mobile and stagnant compartment are known. Both porosities can be only obtained by performing an artificial tracer experiment. The estimation of mean transit time of water can then be made based on these data (Maloszewski and Zuber, 1985, 1991).

2.4 Fields of application

Tracer methods provide direct insight into the dynamics of water in all compartments of the hydrologic cycle. The dynamics include the processes of motion, distribution and dissemination. Tracer techniques are experimental and independent and can thereby be applied to calibrate models. The fact that tracer methods allow for measurements of process- and system-parameters turns them into an effective tool for consultancies and legal authorities.

Regarding the general fields in hydrology where tracer techniques are applicable, experience was gained in all of the components of the water cycle. The possibilities of tracer techniques are vast and comprise, among many others, the investigation of processes such as groundwater recharge, runoff generation, water and solute transport and pollution assessment. A list of important questions that can be tackled by tracer techniques is given in Table 2.3. This list represents the most important components and is not conclusive or necessarily complete.