

lundi 22 octobre 2012

Sediment quality – Geochemistry / environmental chemistry

- > Catchments and weathering: foundations of geochemistry,
- > basic knowledge of lsotopes,
- > The geochemistry of sediments at the catchment scale: the Loire basin as example,
- > Suspended matter and sediments,
- > The carbonate factory and the erosion quantification,
- From the sediment to The labile fraction : how to characterize the anthopogenic environment,
- Small catchments of the basin : primary characterization stage of sediment,
- > To the estuary,
- > The labile fraction and the extension to the basin,
- > From the present to the past,
- > And the future...



CATCHMENTS AND WEATHERING: FOUNDATIONS OF GEOCHEMISTRY







The foundations

> fluvial system and catchment

- water course is a continuum from rainwater towards the ocean
- through runoff, evapotranspiration, infiltration, flow in rivers, unsaturated zone and aquifers.
- > therefore, the way to study hydrologic functioning of fluvial systems must be global.





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The foundations

- > knowledge of the different inputs to the dissolved and particulate loads carried by streams and rivers:
 - study of the different natural and anthropogenic sources.
- > Identify and quantify the different inputs to the dissolved and particulate loads:
 - to describes the spatial evolution of weathering and mechanical erosion rates
- > Identify particle sources and weathering mechanisms:
 - to determine the temporal variations of chemical species bound to the suspended matter and sediments



The foundations: dissolved load

- > Weathering processes initiate the dissolved and suspended loads of most of the world's major rivers.
- > Chemical weathering of rocks and soils is one of the essential processes in the geochemical cycling of elements in rivers





The foundations: suspended load

- > Residual products from chemical and mechanical weathering are carried by rivers and streams to the ocean:
 - as suspended load, typically smaller than a few microns in diameter,
 - and as bed load representing the coarser fraction





indicated by arrows. Sediment yield (tons/km²/yr) for various drainage basins is also shown by appropriate pattern (see legend). Open pattern indicates essentially no sediment discharges to the oceans. (After J. D. Milliman and R. H. Meade. "World-wide Delivery of River Sediment to the Oceans," *Journal of Geology*, 91(1), p. 16. Copyright © 1983 by The University of Chicago Press, reprinted by permission of the publisher.)











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The foundations



TABLE 1.3 Common soil minerals

Name	Chemical formula	Importance				
Quartz	SiO ₂	Abundant in sand and silt				
Feldspar	$(Na,K)AlO_2[SiO_2]_3$ CaAl ₂ O ₄ [SiO ₂] ₂	Abundant in soil that is not leached extensively				
Mica	$K_2Al_2O_5[Si_2O_5]_3Al_4(OH)_4$ $K_2Al_2O_4[Si_2O_5]_3(Mg,Fe)_6(OH)_4$	Source of K in most temperate-zone soils				
Amphibole	(Ca,Na,K) _{2,3} (Mg,Fe,Al) ₅ (OH) ₂ [Si,Al) ₄ O ₁₁] ₂	Easily weathered to clay minerals and oxides				
Pvroxene	(Ca,Mg,Fe,Ti,Al)(Si,Al)O3	Easily weathered				
Olivine	(Mg,Fe),SiO4	Easily weathered				
Epidote	Ca ₂ (Al,Fe) ₃ (OH)Si ₃ O ₁₂	Highly resistant to chemical				
Tourmaline	NaMg3Al6B3Si6O27(OH,F)4	weathering; used as "index mineral"				
Zircon	ZrSiO ₄	in pedologic studies				
Rutile	TiO ₂					
Kaolinite	Si ₄ Al ₄ O ₁₀ (OH) ₈	Abundant in clay as products of				
Smectite	M _x (Si,Al) ₈ (Al,Fe,	weathering; source of exchangeable				
Vermiculite >	$Mg_{4}O_{20}(OH)_{4}$, where	cations in soils				
Chlorite J	M = interlayer cation					
Allophane	Si ₃ Al ₄ O ₁₂ · nH ₂ O	Abundant in soils derived from				
Imogolite	Si ₂ Al ₄ O ₁₀ · 5H ₂ O	volcanic ash deposits				
Gibbsite	Al(OH) ₃	Abundant in leached soils				
Goethite	FeO(OH)	Most abundant Fe oxide				
Hematite	Fe ₂ O ₃	Abundant in warm regions				
Ferrihydrite	Fe10015.9H2O	Abundant in organic horizons				
Birnessite	(Na,Ca)Mn7O14 · 2.8H2O	Most abundant Mn oxide				
Calcite	CaCO ₃	Most abundant carbonate				
Gypsum	$CaSO_4 \cdot 2H_2O$	Abundant in arid regions				

FIG. 1.2 Average concentrations (filled circles) and ranges (vertical lines) of trace metal concentrations in topsoils compared with average crustal concentrations. (Reprinted with permission from *Metal Ions in Biological Systems*, Vol. 20, *Concepts on Metal Ion Toxicity*, H. Sigel (ed.). Marcel Dekker, New York, 1986.)







BASIC KNOWLEDGE OF

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					232.00	231.00	238.00	237.00	239.10	(243)	(247)	(247)	(251)	(254)	(257)	(256)	(254)	(258)	J









The stable isotopes

Table 9.1. Isotope Ratios of Stable Isotopes

Element	Notation	Ratio	Standard	Absolute Ratio
Hydrogen	δD	$D/H (^{2}H/^{1}H)$	SMOW	1.557×10^{-4}
Lithium	δ ⁷ Li	⁷ Li/ ⁶ Li NI	IST 8545 (L-SVE	C) 12.285
Boron	$\delta^{11}B$	${}^{11}\mathrm{B}/{}^{10}\mathrm{B}$	NIST 951	4.044
Carbon	δ ¹³ C	¹³ C/ ¹² C	PDB	1.122×10^{-2}
Nitrogen	$\delta^{15}N$	$^{15}N/^{14}N$	atmosphere	3.613×10^{-3}
Oxygen	$\delta^{18}O$	¹⁸ O/ ¹⁶ O	SMOW, PDB	2.0052×10^{-3}
	$\delta^{17}O$	¹⁷ O/ ¹⁶ O	SMOW	3.76×10^{-4}
Sulfur	$\delta^{34}S$	$^{34}S/^{32}S$	CDT	4.43×10^{-2}



$$\delta^{18}O = \left[\frac{({}^{18}O / {}^{16}O)_{sam} - ({}^{18}O / {}^{16}O)_{SMOW}}{({}^{18}O / {}^{16}O)_{SMOW}}\right] \times 10^3$$

The *fractionation factor*, α , is the ratio of isotope ratios in two phases:

$$\alpha_{A-B} \equiv \frac{R_A}{R_B}$$



THE GEOCHEMISTRY OF SEDIMENTS AT THE CATCHMENT SCALE: THE LOIRE BASIN AS EXAMPLE



Loire

- > The surface waters of the Loire drainage basin offer unusual opportunities for selected geochemical studies because:
 - the Loire drains areas with two main types of bedrock the silicate basement of the Massif Central and the sedimentary area of the southern Paris Basin
 - the watershed is an inland basin in which atmospheric input can be characterized by local rains,
 - parts of the watershed are industrialized and parts are agricultural, where anthropogenic activities may contribute in varying degrees to the dissolved load.





of the total basin surface.







SUSPENDED MATTER AND SEDIMENTS

Evolution of SPM concentration with discharge for the daily (open circles) and monthly (filled circles) samples

weak correlation of increasing SPM concentration with increasing river discharge identified (no cyclical relationship with river flow is observed). The existence of dams

The existence of dams along the river implies that, whereas the SPM originates from the natural erosion of the watershed, the suspended load could be controlled by non-natural processes.

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> Evolution of calcite and Kfeldspar abundances with river discharge

• The quartz and K-feldspar contents increase, calcite contents decrease with increasing discharge

K feldpars (%)

Evolution of major-and trace-element concentrations with discharge in suspended matter

- > concentrations of chemical species can be related to fluctuations in the mineralogical assemblages:
 - abundance of illite and Kfeldspar for K, Si, Fe, Zr, Rb and Ti,
 - abundance of calcite and plagioclase for Ca and Sr

suspended matter: characterisation of natural and anthropogenic fluxes

Fluctuations in ⁸⁷Sr/⁸⁶Sr in the suspended load (★) discharge of the Loire river (●) as a function of months

➢ low flow, lowest ⁸⁷Sr/⁸⁶Sr, agree with the geochemical signature of weathered carbonate bedrock, groundwaters and fertilizers inputs

Increase of the ⁸⁷Sr/⁸⁶Sr ratio with increasing K-feldspar abundance and, conversely, a decrease in the ⁸⁷Sr/⁸⁶Sr ratio with increasing calcite abundance

Sr isotope systematics

The ⁸⁷Sr/⁸⁶Sr ratio fluctuates accordingly to the inverse fluctuation of calcite and Kfeldspar with river discharge. The relationship between the ⁸⁷Sr/⁸⁶Sr and Rb/Sr ratios clearly indicates binary mixing between the calcite and Kfeldspar endmembers.

Suspended matter: characterisation of natural and anthropogenic fluxes

Similarities between the ⁸⁷Sr/⁸⁶Sr ratios near the 1:1 line confirm the existence of authigenic calcite, imply the considerable abundance of this phase primarily during low river flow.

The increase in ⁸⁷Sr/⁸⁶Sr ratios in the SPM is linked to the corresponding increase in K-feldspar abundance.

Relationship between the ⁸⁷Sr/⁸⁶Sr ratios of the suspended particulate matter and of the dissolved load.

THE CARBONATE FACTORY AND THE EROSION QUANTIFICATION



The erosion rates



The detrital and authigenic exportation fluxes were calculated at the two sampling sites.

- At Orleans, the total annual flux is calculated to be 370 103 T/y including 16% of authigenic carbonates.
- At Brehemont, the flux is calculated to be 525 103 T/y with a percentage of authigenic carbonate ranging between 10% and 25%.

> An homogeneous specific erosion rate of 8 t/y/km² has been determined at Orleans and Brehemont



FROM THE SEDIMENT TO THE LABILE FRACTION : HOW TO CHARACTERIZE THE ANTHOPOGENIC ENVIRONMENT





Figure 2.16. Common groups of layer silicate clay structures found in soils, pictured in terms of their tetrahedral () and octahedral () sheets. The usual locations of structural charge and exchange cations are indicated by – and + signs.



The labile fraction

Relationship between the abundance of acid extracted matter (AEM) in the suspended particulate matter and the discharge of the Loire river



REE in the labile fraction

Upper Continental Crust (UCC) normalized patterns for REE in acid extracted matter (AEM) in the suspended particulate matter of the Loire river

REE patterns reveal the nature of the solid phase carrying trace elements: Fe and Mn hydroxide coatings on particles are the most probable candidate.





SR and REE in the labile fraction

Plot of Sr and REE elemental concentration (in ppm) vs. the percentage of acid extracted matter (AEM) in the suspended particulate matter of the Loire river.



dissolved Sr is removed both by oxides and carbonates

fluctuations of Σ REE and AEM concentrations are due to a "dilution" of a terrigenous component (i.e. Fe-Mn coatings) by calcite precipitation during low flow



Sr isotopes in the labile fraction

⁸⁷Sr/⁸⁶Sr ratios plotted against the abundance (%) of acid extracted matter (AEM) in the suspended particulate matter of the Loire river



- Sr integrated into neoformed calcites is directly derived from the local dissolved load.
- Fe and Mn hydroxides may have precipitated in waters similar to those found in the upstream part of the basin (crystalline basement of the Massif Central).

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SMALL CATCHMENTS OF THE BASIN : PRIMARY CHARACTERIZATION STAGE OF SEDIMENT





Location Location map of the Loire watershed and location of the small investigated watersheds. DW ALW refers to the Allanche watershed; Ć DW refers to the Desges watershed. ζ ζ Orléans ξ ξ D5 γ alkaline basalt bedrock <u>ALW</u> D11 γ Centra γ γ **D**8 γ A5 γ 5 km γ ξ Micaschists, γ Granit ζ Gneiss leptyno-amphibolite group Geoscience for a sustainable Earth Allagnon river brqm 5 km



Allanche catchment The investigated area in the Massif Central consists in the Brioude-Massiac ore District where the metalliferous mineralizations contain mainly Pb-bearing minerals together with Cu and Sb as main associated trace elements and Sb-bearing minerals with Zn, Cu and Pb as trace elements.

The Allanche catchment is underlain by alkaline basalt bedrock.

No evidence of ore deposits on the Allanche river, covered by basalt bedrocks.

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5 km

Desges catchment



The investigated area in the Massif Central consists in the Brioude-Massiac ore District The bedrock of the DW consists of three main units; granite, gneiss and micaschist. At higher altitude, the DW is underlain by Margeride granite and gneiss. The lower part of the DW is composed of micaschist and rocks of the leptyno-amphibolite group (massive gneiss, amphibolite, etc.). The bed sediments and residual soil samples were collected along the two watersheds.

The Desges River drains a site of known ore mineralizations deposits and some mining activities are well known during more than thirty years.









Mn and Th are generally linked by a linear relationship but correlation coefficient is better in ALW (r = 0.99) than in DW (r = 0.55).

Slope is greater in DW samples than in ALW ones and Th tends to be more enriched in ALW soils and bed sediments than in DW ones.

Slope for Fe-oxides mimics that of ALW leach with a lower range of content.

Bulk residues show lower Mnhigher Th contents that plead in favour of Th concentration in

¹₁₀ [Th] resistant detrital minerals in sediments.





Sr isotopes



basalt watershed: the Srisotope compositions of AEM fall in the same range as that of water and residues.

granite watershed: AEM shows ⁸⁷Sr/⁸⁶Sr ratios similar to those of the water in the upstream part, whereas a divergence between the two ratios appears downstream.

This implies that oxides have precipitated upstream and are not re-equilibrated during their transport downstream.





ALW Leach ALW residu DW Leach DW residu Allier Rainwaters (Roy & Négrel, in prep) Leach RW solid matter Waters draining silicate, Seine river (Roy, 1996) Stream sediments HBr 0.5M, Seine river (Roy, 1996) Stream sediments, silicate residu, Seine river (Roy, 1996) Fe-oxides (Cézallier, this study)

 \cap

Lead in DW reflects a natural origin for lead, coming from processes of weathering and erosion of bedrock and associated ore mineralisations. The second origin for lead can be related to anthropogenic inputs by fertilisers. On the ALW, lead cannot be linked with the presence of ore mineralisations. Lead isotopes point out an origin from weathering and erosion of basaltic bedrock and anthropogenic inputs from gasoline.







TO THE ESTUARY



Figure 7.5 Concentration of suspended matter versus chlorinity in York River, Virginia, showing a turbidity maximum at the landward limit of sea salt penetration into the estuary; e.g., where the chloride concentration drops to nearly zero. (After R. H. Meade, 1972. "Transport and Deposition of Sediment in Estuaries," Geol. Soc. Amer. Memoir 133, p. 100, and B. N. Nelson, 1960. "Recent Sediment Studies in 1960," Va. Polytech. Inst. J., 7(4), pp. 1-4.)



Figure 3 - Reconstitution expérimentale de vasières dans l'estuaire de la Seine. En haut : plan de situation. En bas : vue aérienne de la vasière (document mission pont de Normandie).



Figure 7.6 Suspended matter transport (arrows) and sediment accumulation near landward limit of seawater mixing in an estuary. Dashed line is fresh-saline water boundary. (After R. H. Meade, 1972. "Transport and Deposition of Sediment in Estuaries," Geol. Soc. America Memoir 133, p. 112.)



As starters

The sediments and the associated contaminants carried through the estuaries have a noticeable impact on the coastal water quality and on the shoreline evolution.

Estuaries, however, experienced in the last decades large changes:

- The efficiency and the presence of large swamps, that constituted a complex system of water-sediments exchange between rivers, land and ocean decrease.
- The turbidity maximum extends deeply inland



Figure 1 - Évolution des estrans entre 1834 (gauche) et 1979 (droite) dans l'estuaire de la Seine (Avoine, 1981).

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As starters

Streams and rivers transport the result of erosion processes to the ocean as suspended and dissolved forms,

The natural balance of chemical species is disturbed by anthropogenic additions deriving from domestic and industrial activities,

Estuaries and Coastal catchments represent:

- \rightarrow a transfer box for sediments between land and open ocean
- \rightarrow consist as very reactive areas









Simplified geological map of the Loire basin. Geographical situation of the estuary and location of the turbidity maximum in 1992 (greyed area).







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Mixing in the estuary

Assuming two end-member mixing, the following mixing equation can be applied (Faure, 1986):

$$Xm/Ym = aXa/Ya + (1 - a)Xb/Yb$$
(1)

where X/Y is the La/Fe or Ce/Fe ratio, *m* is mixing, *a* and *b* are the two components involved in the mixing, *a* is the proportion of *a* in *a*+*b*, and 1-a is the proportion of *b* in *a*+*b*.



Mixing % The relationship between REE (La and Ce) vs. Fe ratios can be used as a tool for quantifying the amount of the two particle sources in the estuarine area. In addition, these results illustrate the important contri-Estuarine, Coastal and Shelf Science (1997) 44, 395-410 Æ bution that sediments of a fluviatile origin make to **Multi-element Chemistry of Loire Estuary Sediments:** Anthropogenic vs. Natural Sources the bed sediments within the Loire estuary system. Ph. Negrel Approximately 50% of the sediments transported seaward is of terrestrial origin. 100 %particules origine continentale 90 80 70 60 50 40 30 10 50 70 80 20 60 90 0 30 40 Distance/Océan (km) Geoscience for a sustainable Earth Fluviatile Ocean

Lead and its isotopes in the estuary



The increase in lead and Mn contents along the estuary accompanies variations in the ²⁰⁷Pb/²⁰⁶Pb ratios, which first increase and then decrease down to the outlet.

The sampling point in the fluvial part, differs by its ²⁰⁷Pb/²⁰⁶Pb ratio due to the decrease in leaded-gasoline; the sampling point near the estuary outlet differs mainly by its content due the decrease of lead emissions in the estuary. The large increase of the ²⁰⁷Pb/²⁰⁶Pb ratio corresponds to a lead input from the large industrial areas of harbour and ship-building activity

Range in the middle Loire

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THE LABILE FRACTION AND THE EXTENSION TO THE BASIN







Fe-Mn oxides act as the main carrier phase of the elements in the AEM extracted by cold HBr. Thorium displays mostly insoluble behaviour in hydrosystems, but the small amount of dissolved Th shows a strong tendency to be adsorbed onto oxyhydroxides Mn and Th (as well as other trace elements) correlate well in AEM, the correlations of Mn, and Pb with Th as a typical indicator of crustal weathering points to their derivation from the silicate basement of the upstream part of the catchment.





The different Pb sources are natural Pb from weathering processes, Pb derived from industrial activities, Pb from gasoline, Pb from agricultural activities through the use of fertilizers and amendments, and Pb from past mining activities.




The calculation of the mixing lines shows that mixture а EM1 between and EM3 is found in the Desges catchment. Chatillon and Villerest samples. Any lead derived from natural sources can be identified this from mixing line. as it would fall on the line. Thus we can plead in favour of at least mixing between lead derived from mineral deposits and agricultural activities,

Mixing between EM1 and EM2 encompasses samples from the and natural derived Allanche watershed and those of the Loire River during the lowest lead. discharge period. The Maine River sample appears to be largely marked by lead derived from gasoline plus an influence of mineral lead. The other samples plot between the mixing lines and thus reflect a multi-component mixing.



Samples from the Loire-Allier junction up to the estuary all define a strong linear trend between the higher Pb-isotopic composition represented by the gasoline field and the lower Pb-isotopic composition represented by inputs from the upstream catchments

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FROM THE PRESENT TO THE PAST



Site location



Schematic cross section and relationships between main bodies

The sampling sites are situated in the middle part of the Loire River course (Avaray Valley). In the Avaray Valley, the Loire is incised into Aquitanian (e.g. Lower Miocene) lacustrine limestone and numerous incisions, oxbows and channels related to meander migration have been identified. Five alluvial bodies were investigated (A to E). A core for geochemical analysis (M25B) was extracted from 6.3 m of Holocene sediments in an oxbow infilling











There are two important differences between the Sr isotope ratios of AEM in the sediment column and those of modern suspended matter in the Loire River





- The small amounts of carbonate show lower and more variable ⁸⁷Sr/⁸⁶Sr ratios in the alluvial sediments than in the modern suspended matter.
 - This may be due to different proportions of the sources, granites, metamorphic and basaltic rocks.
- Compared with the modern carbonate endmember, which is in isotopic equilibrium with dissolved Sr in the Loire river, that in the sedimentary column has lower ⁸⁷Sr/⁸⁶Sr ratios, similar to those of both the lacustrine limestone and gastropods.
 - Since the latter reflect the isotopic composition of dissolved Sr, it is possible that the AEM from carbonates in the sediments is authigenic rather than detrital in origin.





The hypothesis that AEM contributions result from crustal weathering is confirmed by the Pb isotope ratios in the late Holocene Loire sediments.

Lead content and Pb isotope data provide useful markers in riverine sedimentary records.

Lead originated from the granite and basalts of the Massif Central, through erosion processes and mining activities but fluvially transported material should have been the mean for lead dispersion, as found with Sr isotopes.





Fluctuations in Pb content and isotopic composition were probably related either to natural inputs (erosion processes, volcanic events) or human impacts (burning and of forest clearance for agriculture, mining and smelting of Pb ore) during the Holocene. Volcanic events such as the Vasset/Killian tephra event (VKT) are recorded in the AEM of the sediment core.

The duration of increases in tons/year) Pb contents and the ²⁰⁷Pb/²⁰⁶Pb ratio variations were sometimes greater than explained be by can weathering of recent volcanic deposits alone.).





Increased erosion as the result of soil cultivation the mid late in to Holocene have may prolonged the contribution of volcanic material to the alluvial deposits. In the late Sub-Boreal and the Sub-Atlantic periods the Pb originated from weathering of granites and basalts (natural plus human induced) and from ore mining (e.g. production curve).





Granulometry, mineralogy of the channel infill



The semi-quantitative XRD analysis showed that:

clay minerals representing between 25 and 90% of the sediment,

quartz, feldspar and calcite are the main mineral components (less than 5% to 40%).

The clay mineralogy in the < 4μ m fraction is dominated by the interstratified smectite-illite (40 to 90%), illite and kaolinite make up the rest of the clay minerals. The positive correlation between calcite and AEM (r = 0.86) indicates that the control of AEM content is mainly carbonate



Stable O and C isotope of the channel infill 60 · AEM AEM Present day suspended matter of the Loire catchment (Négrel et al., 2000) Present day suspended matter of the Loire catchment (Négrel et al., 2000) 5139 8315 30 -5139-2950-ര 10 --12 -8 -16 -4 -2 -10 -8 -6 $\delta^{13}C$ δ¹⁸Ο Geoscience for a sustainable Earth brqm

Carbonate and stable O and C isotope of the channel infill The large decrease of the



The large decrease of the $\delta^{13}C$ in the upper part of the record has been ascribed to the progressive closure of the meander and thus the control of the C isotope signature by a local environment with more negative $\delta^{13}C$ values due to oxydation of organic matter. can postulate that We the variations in the δ^{18} O are related to the origin of the water conferring its signature to the carbonate and being less dependent of the meander evolution.

This functioning also agrees with the variation in the clay-mineralogy.



As summary





Summary

The historical record of stable O and C isotope ratio variations together with the distribution of particle sizes allow to constraint the evolution of the river dynamics. The relationship between the decrease in particle size and associated changes in C isotopic records showed the gradual closure of the meander. In this context, the closure is the image of the progressive disconnection of the meander with the bed alive and changes in the C origin.



TO CONCLUDE

Whatever the size of the river

- Think about tools to be used
- Bulk includes mainly geogenic
- Leach concerns anthropic
- Use multi tools approach

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Dra

AND THE FUTURE



ISOTOPES PRINCIPLES AND APPLICATIONS

feresa M. Mensing







