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Pesticide Residues in Food and Human Health.

Pesticide Residues in Fruit and Vegetables in European Countries and in Greece, and Long-term Risk to Consumer Health

Athanasios Valavanidis, Thomais Vlachogianni
Department of Chemistry, University of Athens, University Campus Zografou, 15784 Athens, Greece

Abstract. Most of the agricultural crops from the beginning of agriculture suffered from pests and diseases causing large losses in yield. The growth in the use of agrochemicals and organic synthetic pesticides accelerated in the 1940s and progressed through the last decades increasing substantially the yields of basic crops. The world population in the last century increased rapidly as a result of adequate food supply and better health conditions. Adequate food supply remains a serious problem for modern farming techniques. In the last decades, developed countries approved new regulations restraining the use of agrochemicals for the protection of the environment and consumers' health through scientific toxicological testing of chemical substances and enforcement of lower concentration limits for the pesticide residues tolerated in food and water. The International Maximum Residue Limits (IMRLs) of Codex Alimentarius (1963), established by the Food and Agriculture Organization (FAO) and World Health Organization (WHO) regulate food standards, codes of practices and food safety.

Pesticides in modern agriculture were needed to protect crops from pests, improve yields, lower food prices and increase the availability of vegetables, fruits, beans and meat. Additionally, insecticides protect livestock from diseases and agricultural populations from malaria vectors, yellow fever, typhus and other parasitic diseases. The perception that pesticide residues are dangerous to health, especially to children, is overestimated and scientific toxicological results are misrepresented in popular media. Codex Alimentarius of FAO/WHO was established as an international system for control of pesticide misuses and setting acceptable levels. This review describes research and epidemiological studies of risk assessment of residues in food products. Also, presents statistical data of the last decade in the world, European countries and in Greece for the annual surveys of food commodities with pesticide residues from sampling and analysis by sensitive techniques of GC-MS of very low residue concentrations. The European Food Safety Authority (EFSA) plays an important role in assessing the safety for consumers based on the toxicity of the pesticide residues and working with regulatory bodies, scientists and policy makers worldwide. Monitoring programmes showed that only 2-3% of foods analyzed (in Europe and in Greece) have higher concentrations than the MRL levels and more than 60% have no detectable residues at all. At the same time research on total diet intake (TDI) showed that daily consumption of fruit and vegetables have many advantages (vitamins and antioxidants) for a healthy diet, whereas perceived health risks are very low. Organic food commodities contain lower concentrations of residues but their much higher prices might prevent low income consumers to have the necessary five-seven servings per day. Developed countries (USA, Western Europe, etc) have for years an active health campaign promoting the consumption of fruit and vegetables. The review presents selected papers of the last years on pesticide residues in various countries and total diet studies estimating health risk to the general population and young people.

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Corresponding authors : Prof. A.Valavanidis, **www.valavanidis@chem.uoa.gr**

Υπολείμματα Φυτοπροστατευτικών Δραστικών Ουσιών στα Τρόφιμα και Ανθρώπινη Υγεία

Υπολείμματα Φυτοπροστατευτικών σε Φρούτα και Λαχανικά σε Ευρωπαϊκές Χώρες και στην Ελλάδα και Επιπτώσεις στην Υγεία των Καταναλωτών

Αθανάσιος Βαλαβανίδης, Θωμαΐς Βλαχογιάννη

Τμήμα Χημείας, Πανεπιστήμιο Αθηνών, Πανεπιστημιούπολη Ζωγράφου, 15784 Αθήνα

Περίληψη: Από την εποχή της αγροτικής επανάστασης οι καλλιέργειες καταστρέφονταν από παράσιτα και ασθένειες οι οποίες μείωναν σημαντικά τις γεωργικές αποδόσεις. Η ανάπτυξη και χρήση των φυτοπροστατευτικών δραστικών χημικών ουσιών (βιοκτόνα) για καταπολέμηση των παρασίτων (έντομα, μύκητες βακτήρια, ζιζάνια, κλπ) βελτίωσε σημαντικά τις αποδόσεις των βασικών καλλιεργειών (δημητριακά, ρύζι, καλαμπόκι, σόγια, κλπ) από το 1940 και μετά. Τις τελευταίες δεκαετίες η χρήση λιπασμάτων και φυτοφαρμάκων αυξήθηκε δραματικά για να ανταποκριθεί τις διατροφικές ανάγκες του πλανήτη Γη με τον σημερινό πληθυσμό των 7 δισεκατομμυρίων. Αν και η ποσότητα τροφίμων αυξήθηκε σημαντικά οι γεωργικές καλλιέργειες απειλούνται από κλιματολογικούς και γεωλογικούς παράγοντες, επάρκεια νερού, διάβρωση-ερημοποίηση εδαφών και φυτοπαθολογικές ασθένειες και ζιζάνια. Η χρήση φυτοπροστατευτικών δραστικών ουσιών για τους επιστήμονες θεωρείται βασικός παράγοντας προστασίας των γεωργικών καλλιεργειών, αλλά η περιβαλλοντική προστασία απαιτεί νέες μεθόδους εφαρμοσμένων γεωργικών πρακτικών και φυτοπροστατευτικά προϊόντα με μειωμένη τοξικολογική δράση. Τα υπολείμματα γεωργικών φυτοπροστατευτικών ουσιών στα τρόφιμα εδώ και πολλές δεκαετίες ρυθμίζονται με διεθνή και εθνική νομοθεσία και αυστηρούς τοξικολογικούς περιορισμούς που λαμβάνουν υπόψη τις διατροφικές κλίμακες της υγιεινής διατροφής. Το σύστημα για τα Ανώτατα Επίπεδα Υπολειμματικών Φυτοφαρμάκων (MRLs) με το Codex Alimentarius της Διεθνής Οργάνωση Τροφίμων και Γεωργίας (1963, FAO) και της Παγκόσμιας Οργάνωσης Υγείας (WHO) ρυθμίζει τα επίπεδα των ανώτατων επιτρεπόμενων υπολειμμάτων φυτοφαρμάκων (με τοξικολογικά δεδομένα) στα διάφορα τρόφιμα και τις πρακτικές για την ασφάλεια των τροφίμων.

Τα φυτοπροστατευτικά προϊόντα καλύπτουν τις γεωργικές καλλιέργειες και τις κτηνοτροφικές δραστηριότητες από παράσιτα, ζιζάνια, μύκητες και βακτήρια και αυξάνουν την απόδοση φρούτων, λαχανικών, όσπρια, κρέατος κ.α. Συγχρόνως, οι φυτοπροστατευτικές και βιοκτόνες δραστικές χημικές ουσίες προστατεύουν γεωργικούς πληθυσμούς από ελονοσία, κίτρινο πυρετό, τύφο και άλλες παρασιτικές και λοιμώδεις ασθένειες. Η αντίληψη ότι τα υπολείμματα φυτοφαρμάκων στα τρόφιμα είναι υπεύθυνα για αλλεργίες, δερματικές ασθένειες, γαστροεντερικά προβλήματα, κακοήθεις νεοπλασίες και άλλες ασθένειες, είναι εξαιρετικά διαδεδομένες και παρουσιάζονται έντονα σε μεγάλης κυκλοφορίας δημοσιογραφικά έντυπα και μέσα μαζικής επικοινωνίας. Αντίθετα, οι επιστημονικές έρευνες που διεξάγονται ετησίως για την ανίχνευση φυτοφαρμάκων στα τρόφιμα και οι επιστημονικές εκτιμήσεις για τις επιπτώσεις στην υγεία των καταναλωτών είναι σχετικά άγνωστές εκτός από στενό κύκλο ειδικών. Επίσης, τα ανώτατα επιτρεπόμενα όρια φυτοπροστατευτικών ουσιών (MRLs) και τα τοξικολογικά δεδομένα στα οποία στηρίζονται, βελτιώνονται με νεώτερα δεδομένα και εφαρμόζονται σε διεθνές και εθνικό επίπεδο ε είναι γενικώς άγνωστα σε μεγάλα ποσοστά καταναλωτών. Η ανασκόπηση αυτή φιλοδοξεί να παρουσιάσει μία σφαιρική ενημέρωση για τα στατιστικά στοιχεία προσδιορισμού φυτοπροστατευτικών ουσιών στα τρόφιμα στις χώρες της Ευρωπαϊκής Ένωσης και στην Ελλάδα. Μέσα από την πληθώρα επιστημονικών εργασιών καταγράφονται τα αποτελέσματα επιστημονικών αναλύσεων για τις συγκεντρώσεις υπολειμμάτων στα διάφορα τρόφιμα, και μελέτες για τον κίνδυνο στην υγεία των καταναλωτών. Η ανασκόπηση καταγράφει τα στατιστικά στοιχεία των τελευταίων ετών για τις 70-80 χιλιάδες αναλύσεις τροφίμων στη ΕΕ (Ευρωπαϊκή Υπηρεσία για την Ασφάλεια των Τροφίμων) και το μικρό ποσοστό τροφίμων (2-3%) με υψηλές συγκεντρώσεις. Επίσης στην ανασκόπηση αναλύονται τα αποτελέσματα των πλέον σημαντικών και πρόσφατων διεθνών ερευνών για τις επιπτώσεις στην υγεία των καταναλωτών, και ιδιαίτερα παιδιών, υπολειμμάτων φυτοφαρμάκων στα διάφορα τρόφιμα. Οι ποσοτικές αναλύσεις τροφίμων βιολογικής και συμβατικής καλλιέργειας έχουν δείξει ότι τα βιολογικά έχουν χαμηλότερες συγκεντρώσεις υπολειμμάτων. Η ανασκόπηση περιλαμβάνει τις εκστρατείες σε εθνικό επίπεδο πολλών αναπτυγμένων χωρών για την κατανάλωση φρούτων και λαχανικών με σκοπό την προστασία της υγείας του ανθρώπου λόγω της υψηλής περιεκτικότητας σε βιταμίνες, ιχνοστοιχεία και αντιοξειδωτικές ουσίες. Τέλος, η ανασκόπηση περιλαμβάνει μεγάλο αριθμό επιστημονικών εργασιών για τις μελέτες ολικής διατροφικής σε διάφορες αναπτυγμένες χώρες και τις δυνητικές επιπτώσεις στην υγεία των καταναλωτών από υπολείμματα φυτοπροστατευτικών δραστικών ουσιών.

Υπεύθυνος του άρθρου. Καθηγητής Αθανάσιος Βαλαβανίδης www.valavanidis@chem.uoa.gr

Introduction: Agriculture and the Cradle of Civilization

Agriculture first began about 12,000-10,000 years ago in the Fertile Crescent of Mesopotamia where edible seeds were initially gathered by a population of hunter gatherers. Historians agree that human civilization began in the neolithic period due to the agricultural revolution. Out of agriculture, cities and civilizations grew, and because crops and animals could now be farmed to meet food demand, the global population rocketed, from five million people 10,000 years ago, to more than seven billion today. The Fertile Crescent was a wide expanse of land along the Tigris and Euphrates rivers, stretching from the Persian Gulf to the Mediterranean Sea, and even into the Nile River. These river valleys, known as the "cradle of civilization," had rich soils in which crops flourished. Civilization first emerged in Mesopotamian cities like Babylon, Sumer, Ur, and Uruk in modern-day Iraq and in Jericho to the west. Egyptian civilization emerged in the Nile River Valley later. The first crops were wheat, barley, peas, lentils, chickpeas, etc. Similarly, in China rice and millet were domesticated 7,500 years ago rice and sorghum were farmed in the Sahel region of Africa.[1,2]

Most of the farmed crops from the beginning of agriculture suffered from pests and diseases causing a large loss in yield. Destruction of agricultural products resulted in great famines, reduction of the population and in some cases civilizations were destroyed. Despite the advances in agricultural sciences, the use of modern crops, pesticides and fertilizers, losses due to pests and diseases range from average 35% to 40%, for all potential food crops. Inevitably farmers and agricultural scientists have a great incentive to find ways of overcoming the problems caused by pests and diseases. Historical records have references for the use of insecticides at about 4,500 years ago by Sumerians [sulphur (S) compounds to control insects and mites]. In another case, about 3,200 years ago, the Chinese were using mercury and arsenical compounds for controlling body lice. Ancient Greece and Rome historical records show that religion, folk magic and the use of what may be termed chemical methods were tried for the control of plant diseases, weeds, insects and animal pests. Farmers used smokes (burning various materials) and tar against insects and plant extracts such as bitter lupin or wild cucumber. Weeds growing among crop plants were controlled mainly by hand weeding but various “chemical” methods are also described such as the use of salt or sea water. Powder of pyrethrum (from dried flowers) was another physical substance that was used as an insecticide for many centuries. Many inorganic chemicals (copper sulphate, lime, salts of lead and mercury etc) have been used since ancient times as pesticides and copper mixtures are still used against various fungal diseases in farming.[3,4,5,6]


Figure 1. The agricultural revolution 10,000 years ago in the Fertile Crescent was the wide-scale transition of many human cultures from a lifestyle of hunting and gathering to one of farming and settlement in one place, allowing the ability to support an increasingly large population with adequate nutritious food.

Before the Second World War (1920s-1930s) inorganic substances, such as sodium chlorate and sulphuric acid, or organic chemicals derived from natural sources were still widely used in pest control. Agrochemicals such as nitrophenols, chlorophenols, creosote, naphthalene and petroleum oils, which were highly toxic, used for fungal and insect pests, whilst ammonium sulphate and sodium arsenate were used as herbicides. The growth in organic synthetic pesticides accelerated in the 1940s with the discovery of the effects of DDT, aldrin, dieldrin, endrin, chlordane, parathion, captan and 2,4-D. A well known example was the successful and extensive use of insecticide of DDT that exterminated very dangerous insects to food production and reduced insect-born diseases like malaria, yellow fever and typhus (Dr. Paul Muller won in 1949 the Nobel Prize in Medicine, for the use of DDT as effective insecticide) that shaved millions of human lives all over the world.[7]

Figure 2. A variety of agrochemicals for the protection of crops and the extermination of insects and fungal pests were used in the previous centuries of agricultural development, but synthetic pesticides from the 1950s became important in the dramatic increase of yields of basic crops.

After the 1950s, consumers and most policy authorities in developed countries for health and environmental pollution were concerned about the potential health risks of pesticide residues in food. Farmers developed various diseases and cancers from excessive use of pesticides and the lack of protective health and safety measures. Environmental problems from excessive and indiscriminate use of organic pesticides were highlighted by Rachel Carson in her book "*Silent Spring*" (1962).[8] The environmental problems caused by pesticides (water and soil pollution, ecosystem degradation, from toxic, carcinogenic and persistent pesticides) were projected in the last decades by various scientific research projects and new legislation in developed countries.[9-14]

World Population, Famine and Food Production

The world population grew from 2.5 billion in 1950 to 6.1 billion in the year 2000 and 7 billion in 2014. By the year 2050 the world population is estimated to reach 9.1 billion (between 7.7 and 10.6 billion, depending on estimates). This means that the population of Earth more than doubled in the past 50 years. In Western developed industrial countries this supply of energy calories is largely obtained from livestock products (meat, milk, fish), while in many developing countries the supply is primarily obtained from cereal grains, fruit and vegetables. Overall, 80% of the poor in developing countries live in rural areas and derive their livelihood directly from agriculture with diets that are deficient in micronutrients (minerals, vitamins, etc.), proteins and animal fat.[15] The future of adequate food supply with increasing global population remains a serious problem for the modern farming techniques and the use of fertilizers and agrochemicals for crop protection.[16-19]

Famines of large scale occurred even during the 20th century (1900-2000). It is estimated that 70 million people died from famines across the world, of whom an estimated 30 million died during the famine of 1958–61 in China. The other most notable famines of the century included the1942–1945 disaster in Bengal, famines in China in 1928 and 1942, and a sequence of famines in the Soviet Union (1932-1933, caused by the policies of Stalin). Other famous famines were the Biafran famine (southeastern Nigeria) in the 1960s, the Khmer Rouge-caused famine in Cambodia in the 1970s and the Ethiopian famine of 1984–1985.[20]

At the global level, significant progress has been made since 1960 towards improved nutrition and food security. Since then, world gross agricultural production has grown more rapidly than the world population, with an average positive production increase of food *per capita*. While food security improved significantly in East Asia, it became very unsatisfactory in sub-Saharan Africa and South Asia. This was the result mainly of the **Green Revolution** (1960-1980) that led to the development of new crops with high yields by international agricultural research centers in the developing world. Researchers with the help of western agricultural experts developed new impressive high yield seeds, but also increasing use of fertilizers, water and pesticides. The key breakthrough in Mexico was the breeding of short-stemmed wheat that grew to lesser heights than other varieties. The Mexican dwarf wheat was first released to farmers in 1961 and resulted in a doubling of the average yield. India for example was on the brink of mass famine in the early 1960s because of its rapidly growing population. Green revolution research developed a new variety of rice (IR8) that produced more grain per plant when grown with irrigation and fertilizers. Today, India is one of the world's leading rice producers and IR8 rice usage spread throughout Asia. Critics argued that the Green Revolution benefited large farm operations, and displaced poorer farmers from the land. Also, fertilizer and irrigation caused long-term degradation of the soil and water pollution. As a result the highly celebrated Asian (India, Pakistan, Philippines, Indonesia) and Mexican crop "miracles" masked the loss of agro-biodiversity, the massive reduction of water tables, salinization and erosion of soils, and the displacement of millions of peasants to fragile hillsides, shrinking forests, and urban slums.[21-26]

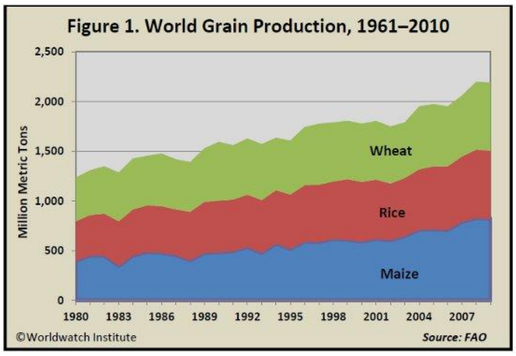

Figure 3. The Green Revolution in Mexico and Asian countries changed dramatically the agricultural technology and the production of basic crops. World grain (wheat, rice, maze) production in the last 40 years increased substantially in parallel with world population [source: Worldwatch Institute, Vital Signs, 2011, & FAO, http://vitalsigns.worldwatch.org/vs-trend/world-grain-production-down-2010-recovering].

Agrochemicals in Agriculture, Health and Safety Issues

Agrochemicals and especially pesticides became a standard technological part of agriculture production system in the last decades with many benefits for crop protection and high yields. [27] In the beginning (after the 1940s) the use of pesticides was without safety and health controls and regulations. But later toxicological studies and environmental pollution measurements proved that these chemicals do pose a potential risk to humans and other life forms and pollute the environment. Exposure to pesticide residues caused serious health effects to farmers in developing countries (through negligence, and ignorance/avoidance of protective measures and safety regulations). The high risk groups exposed to pesticides include production workers, formulators, sprayers, mixers, loaders and agricultural farm workers. Regarding the use of pesticides in developed countries, including European Union, USA, Japan, Australia and Canada, from the 1960s they applied stricter health and safety regulations, also approved new laws restraining the use of agrochemicals and introduced vigorous environmental and health criteria for their manufacture and agricultural distribution.[28-32]

The **European Union** has applied rules for the sustainable use of pesticides by member states to reduce the risks and impacts of pesticide use on people's health and the environment (Directive 2009/128/EC). The **European Food Safety Authority** (EFSA) is the agency that assesses the safety for consumers of the pesticide residues. The EFSA is also working with regulatory bodies, scientists, and policy makers. Pesticides usually contain more than one active substance and the agencies of the EU evaluate every active substance for safety before it reaches the market in a product. Substances must be proven safe for people's health, including their residues in food and effects on animal health and the environment. Under the new EU rules, it takes 2-3 years from the date of admissibility of the application to the publication of a regulation approving a new active substance, depending on how complex and complete the dossier is for the pesticide companies (Regulation 188/2011).[33]

In the **United States**, the EPA (Environmental Protection Agency) regulates pesticides at the national level under the authority of the Federal Insecticide, Fungicide and Rodenticide Act (FIFRA) and other laws. EPA register pesticides, educate applicators, monitor compliance and investigate pesticide health and safety problems. The federal agencies Food and Drug Administration (FDA) and U.S. Department of Agriculture (USDA) assess pesticide residues in food. The U.S Fish and Wildlife Service assess the risk of pesticides to wildlife and the environment. The National Pesticide Information Center and the Centers for Disease Control provide information on pesticides in the USA.[34,35]

Figure 4. The European Food Safety Authority, FAO/WHO Codex Alimentarius Commission, Food and Drug Administration (FDA, USA) and other international and national organizations control the potential human health and environmental effects of agrochemicals used in agriculture.

The EPA controls a wide variety of potential human health and environmental effects associated with use of pesticides. Manufacturers must provide data from studies that comply with EPA testing guidelines. The EPA has developed risk assessments that evaluate the potential for harm to humans, wildlife, fish, and plants, including endangered species and non-target organisms. Also, the EPA controls the contamination by pesticides of surface water or ground water from leaching, runoff, and spray drift. Potential human risks range from short-term toxicity to long-term effects such as cancer and reproductive system disorders. Under the Food Quality Protection Act (FQPA), EPA must ensure that all pesticides used on food in the USA meet FQPA's stringent safety standards.[36]

The **Organization of Economic Co-operation and Development** (OECD, Paris) assists countries in harmonising test methods for chemical safety and good laboratory practice for pesticides, in order to ensure high quality and reliable data for countries on a global scale. Chemical industries and national regulatory authorities benefit from the OECD agreement on Mutual Acceptance of Data and avoid duplicative testing. The OECD Guidelines for the testing of chemicals (including pesticides) are a collection of the most relevant internationally agreed testing methods used by governments, industry and independent laboratories to assess the safety of chemical substances. They are primarily used in regulatory safety testing and subsequent chemical notification and registration.[37,38] The OECD provides a platform for information and publication of national websites of various countries on pesticide health and safety laws, regulating authorities, national surveys and standards.[39,40]

International organizations, such as the World Health Organization (WHO), have formulated various policies and recommendations on improvements of health and safety for workers and farmers using pesticides. The WHO issued guidelines for registration, classification by hazards, safety measures for farmers and studies of health and risk assessments for residues in food. The WHO implemented sustainable epidemiological surveillance and monitoring of pesticide poisoning in clinical settings and communities. Also, developed community programmes that minimize risks of intentional and unintentional pesticide poisoning and improved the medical management care of people with pesticide poisoning.[41,42] The United Nations Environment Programme (UNEP) advanced in the last decade numerous studies of health effects of pesticides residues to children.[43]

Dietary Pesticide Residues. International Health Standards and Maximum Residue Limits (MRLs)

Maximum Residue Levels (MRLs) are not toxicological safety limits. They are a commercial standard, indicating the legally allowed maximum amount of an active ingredient which may be present as a residue in or on an unprocessed raw product to verify whether a crop protection product has been correctly applied or not. The Maximum Residue Limits–Codex Alimentarius (MRLs) that define the residue limits in food was established by the Food and Agriculture Organization of the United Nations (FAO, Rome) and the World Health Organization (WHO) in 1963 to develop international food standards, guidelines codes of practices, and recommendation for food safety. Currently the CODEX has 185 member countries and the European Union as a member organization.[44,45]

The MRL is the maximum concentration of a pesticide residue (in mg/Kg of food) to be legally permitted in food commodities and animal feeds. The Codex food standards are based on the best available science (toxicological studies) assisted by independent international risk assessment bodies or ad-hoc consultations organized by FAO and WHO (Joint Meetings on Pesticides). The Codex Alimentarius Commission develops harmonized international food standards, guidelines, and codes of practice to protect the health of consumers and to ensure fair practices in the food trade.[46,47]

International harmonization of MRL does not exist at a global level. Even though the Codex Alimentarius provides MRLs for most of pesticides, they are not statutory. National authorities hold the sovereignty in fixing these limits. Therefore these legal limits can widely vary across countries. Regarding pesticide residues, there are as many regulations as countries. The number of pesticides registered and the MRL set, greatly vary from one country to another. Some countries have adopted very severe rules with MRL well below the Codex settings and zero-tolerance provisions for disallowed or prohibited substances or for which a MRL cannot be established due to the lack of toxicological data. Some countries (e.g. the USA or the EU) have a very detailed list while others provide a limited number of pesticides but zero tolerance provisions or a very low tolerance level.[48]

International organizations have attempted to harmonize pesticide standards worldwide to facilitate trade of food and animal feed. In 1985, FAO adopted an International Code of Conduct on the

Distribution and Use of Pesticides; this was updated in 2014. The code is a set of voluntary standards that are especially useful where national pesticide legislation is inadequate or absent. The standards encourage responsible and generally accepted trade practices, ensure effective and efficient use of pesticides, and promote risk reduction in handling pesticides. Several other international instruments, which any country can adopt, have come into force to aid with pesticide management, including the Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and Their Disposals, the Rotterdam Convention on the Prior Informed Consent (PIC) Procedure for certain hazardous chemicals and pesticides in International trade, and the Stockholm Convention on Persistent Organic Pollutants.[49]

European Union regulations on MRLs. The EU legislation harmonises and simplifies pesticide Maximum Residue Limits (MRLs), and sets a common EU assessment scheme for all agricultural products for food or animal feed. MRLs apply to 315 fresh products and to the same products after processing, adjusted to take account of dilution or concentration during the process. Legislation covers around 1,100 pesticides used in agriculture in or outside the EU. The safety of all consumer groups is covered e.g. babies, children and vegetarians. The **European Food Safety Authority** (EFSA) is the scientific agency that assesses the safety for consumers based on the toxicity of the pesticide residues. The EFSA is also working with regulatory bodies, scientists, and policy makers worldwide to refine methodologies and provide risk assessors with new tools to determine possible combined effects derived from exposure to multiple chemicals through consumers' diets. [50,51]

Regulation (European Community) No 396/2005 establishes the MRLs of pesticides permitted in products of food commodities (plant or animal origin). MRLs are derived after a comprehensive toxicological assessment of the properties of the active substance; residue levels resulting from the good agricultural practices (GAP). An indispensable precondition for setting MRLs is a risk assessment demonstrating consumer safety (consumer intake not exceeding the toxicological reference values). The EFSA's Pesticides Unit is responsible for the risk assessment of MRLs in accordance with the legislation. The Unit has provided advice in the framework of setting European MRLs (2007). Furthermore, the Pesticides Unit in collaboration with Member States is reviewing the scientific basis of existing MRLs and performs consumer toxicological risk assessments to ensure that MRLs established in the past are compliant with the data requirements and are safe for consumers. Every year, EFSA publishes an Annual Report on Pesticide Residues in the EU based on the monitoring information on pesticide residues in food received from 27 EU Member States and two EFTA countries Iceland and Norway. The EU MRL monitoring programmes are one of the most comprehensive food survey programmes worldwide, covering more than 60.000 food samples every year which are analysed for up to approximately 800 different pesticides. Member States report more than 15 million determinations of pesticide residues on a yearly basis. The report also assesses the exposure of European consumers to pesticide residues through their diets.[52,53]

European Union Database of Approved Pesticides: Active substances approved by the EU are available to view via the website of the European Commission. Each substance can be searched for according to defined criteria and included is a reference to the relevant EU legislation with all toxicological information and MRLs in food and animal feed. The database can be found: http://ec.europa.eu/sanco_pesticides/public/index.cfm .

The **United States and MRLs regulation**. In recent years, tougher legislation on pesticides (lower MRLs). The USEPA also establishes tolerance limits; child health and the risks of pesticide exposure in numerous products must be considered. Tolerance exemptions are occasionally granted for a pesticide ingredient where the exemption is found to be safe. The US Department of Agriculture enforces tolerances for meat and certain egg products, and the US Food and Drug Administration implements tolerances specified for other foods.[53,54,55]

Japan and MRLs of agrochemicals. The Ministry of Agriculture, Forestry and Fisheries specifies standards for the required amount of active ingredients, the maximum permitted amount of hazardous chemical ingredients, and other requirements specific to each type of pesticide (scientific data regarding the physical and chemical properties, types of active ingredients, phytotoxicity, toxicity, and efficacy of the substance along with the provision of samples). The Ministry of Health, Labour, and Welfare is authorized to establish pesticide MRLs. Because Japan is one of the world's largest users of pesticides, a positive list system was introduced in 2006.[56]

China and national food standards. The Institute for the Control of Agrochemicals, Ministry of Agriculture (ICAMA), 1963, affiliated to the Chinese Ministry of Agriculture (MOA) is responsible for the pesticide registration, administration, quality control, bioassay and residue monitoring of pesticides, supervision of pesticide markets, information sharing, international cooperation and other services. China from 2014 established the national food safety standard No 4/2014 with the MRLs for pesticides in food (GB 2763-2014 repealing GB 2763-2012). In total, 371 pesticide items and 3,650 MRLs are defined, based on pesticide toxicity evaluation (i.e. acceptable daily intake: ADI), dietary structure (i.e. pesticide residues intake level), and actual residues on crops in farm produce (i.e. the monitoring data received on the field residues).[57]

The European Union Reports (EFSA) on Pesticide Residues in Food

The annual reports on pesticide residues from 1996-2006 were published by the European Commission, the annual reports 2007-2013 are published by the EFSA Journal. The annual reports of the last 5 years (2008-2012) analysed 67,000-78,000 food samples of pesticide residues from 200-750 food commodities. The results showed that 96-98% of samples were below the MRL standards and 50-55% contained no quantifiable residues at all (the sensitive chromatographic analytical techniques did not record the presence of any pesticide residues).[58]

The EU Reports of EFSA on the results of pesticide residues in 2013 was published in 2015 in the EFSA Journal.[59] The latest 2013 report summarized the results of 80,967 samples of a wide variety of unprocessed raw agricultural commodities (fruit, vegetables, beans etc) and processed food products (products washed, cooked, added preservatives, etc) and analysed residues of 685 distinct pesticides.
A substantial number of samples (8,270) were taken for food products from third countries [under the Regulation (EC) No 669/2009]. In the framework of the EU-coordinated monitoring programme, 11,582 samples of 12 different food commodities were analysed for 209 distinct pesticides. Overall results showed that 97.4% of the tested food samples fell within the legal limits and 54.6% of the samples contained no quantifiable residues at all. In unprocessed product (fruit vegetables) MRL exceedances were detected in only 2.8 % of the samples; 46.1 % of the samples contained measurable residues but within the legal limits and 51.1 % of the unprocessed products were free of detectable residues. Processed food products in general had a lower prevalence of pesticide residues and MRL exceedances. Among the 2,788 individual determinations that exceeded the legal limit, 878 determinations were reported for pesticides not approved in the EU. In most cases these MRL exceedances were related to imported products (659 cases) while for products produced in the EU and EEA countries non-approved pesticides were less frequent (186 results). Baby food samples. In total, 1,597 samples of baby food were analysed and 92.7% of the samples contained no detectable pesticide residues. Organic food products. In total 4,620 samples were analysed for 134 distinct pesticides. Pesticide residues were detected within the legal

limits in 15% of samples and only 0.8% of samples exceeded the MRL. In most cases the detected residues were related to pesticides that are permitted for organic farming, persistent environmental pollutants or residues of substances which may come from natural sources.[59]

Determination and Control of Pesticide Residues in Food, Greece

In Greece the determination of pesticide residues in food commodities and other scientific issues on pesticide control is the work of The **Benaki Phytopathological Institute** (BPI), especially by the specialized Department of Pesticides Control and Phytopharmacy, established in 1977 and its expertise covers all subjects and scientific issues related to the use of pesticides (Plant Protection Products, or PPPs), weed science and Biocidal Products (BPs). The BPI is mandated by the Greek government for the evaluation of pesticides and biocide products at national and European level. Scientific findings, ideas and expertise are continuously exchanged between researchers, regulatory evaluators and agricultural practitioners.[60,61]

The laboratory of BPI performs the determination of pesticide residues in plant protection products through the following methods: gas and liquid chromatography (GC, LG) triple quadrupole mass spectroscopy (TQ-MS) and time-of-flight mass spectroscopy (TOF-MS). Typical samples analysed come from: food of all types (both of primary production and processed), children's food, animal feed, water (potable, irrigation, underground), soil and empty plastic packaging of plant protection products. The number of food samples in Greece that were analysed for the EFSA reports in the last decade.

2000. Samples 1,636 were examined by 5 laboratories, including 1,472 samples of fresh and frozen fruit and vegetables and cereals, of which 63.8% did not contain detectable residues, 30.2% contained detectable residues below the MRL, and only 6% residues exceeding MRL (EFSA report 2000).

2008 Report. Samples 2,229 of which 82% no measurable pesticide residues, 14,6% detected below MRL concentrations, and only 3,3% above the MRL concentrations (EFSA report 2008).

2013 Report. Samples 2,361, of which only 2.2% exceeded the MRL levels and in 27.5% were lower than MRL and 70% contained no quantifiable residues (EFSA report 2013). Examples of pesticides residues that were detected above the MRL concentrations were: Chloropyrifos in peaches, at 1.44 mg/kg, whereas the MRL=0.2 mg/kg, Formetamate in strawberries, 0.55 mg/kg, MRL=0.3 and Acetamiprid in tomatoes, 0.22 mg/kg, MRL=0.15. [62]

Since 2008, Greece followed the EU Regulation 396/2005 for Maximum Residue Levels (MRLs) that have been harmonized for food or feed of plant and animal origin. Pesticide MRLs for processed or composite products are based on the MRLs of the raw agricultural ingredients. According to the new Regulation, the EU is divided in three different zones. Greece is included in Zone C (South) along with Bulgaria, Cyprus, France, Spain, Italy, Malta and Portugal.[63]

Other laboratories for analysis of pesticides in Greece. Food Safety & Quality Laboratories (including the Pesticide Residues Laboratory) of the **General Chemical State Laboratory**, Athens, Greece (Director, Despina Tsipi). The **Hellenic Food Authority** (Ενιαίος Φορέας Ελέγχου Τροφίμων,) (ΚΥΑ 349 ΦΕΚ183/Β'/11-2-2005) is the National Point Contact for the Codex Alimentarius in Greece.

The Authority responsible for reporting pesticide residues in food is the **Ministry of Rural Development and Food. General Directorate of Plant Products**. Directorate of Plant Protection Products. Department of Pesticides. Regional Center of Plant Protection and quality control of Thessaloniki Laboratory of pesticide residues. Kavala, Ioannina, Magnesia, Achaia, Pireaus, Iraklion, and Argolida, ESYD S.A. (Hellenic Accreditation System S.A.) [64]

The Pesticide Residue Control, Summary Reports, Greece (2011-2013) . The most recent report on pesticide residues in food was the 2013, which was published in National competent authority: Ministry of Rural Development and Food. Directorate of Plant Produce Directorate of Plant Produce Protection. Department of Pesticides, available at http://www.minagric.gr/ index.php/en/citizen-menu/foodsafety-menu or http://www.minagric.gr/ index.php/el/for-citizen-2/food-and-sequre/845-asfaleiatwntrofimvnefsa]. The report contained data from the previous years 2011 and 2012 reports.[64]

Year 2011. Number of samples 2,715 , of which 1,983 (73%) without detectable pesticide residues, 653 (24,5%) with detectable residues below the MPLs and 74 (2,7%) with residues exceeding MRLs.

Year 2012. Number of samples 2,797, of which 1,991 (71%) without residues, 754 (27%) detectable residues blow MRLs, and 53 (1,9%) with residues exceeding MPLs.

Year 2013. 2,361, of which 1,649 (70%) without detectable pesticide residues, 650 (27,5%) detectable but below MRLs, and 62 (2,6%) with residues exceeding MRLs.

Figure 5. The Benaki Phytopathological Institute, the General Chemical State Laboratory and Hellenic Food Authority in Greece are responsible for analysis, evaluation and control of pesticide residues in fresh and processed food products.

The report explained the sampling rules. Sampling strategy was based on "from the farm to the fork" rationale, taking into account the specificities of each region of the country. The sampling methods of pesticide residues were those provided for in JMD 91972/2003- Directive 2002/63/EC. Samples were taken by domestic production and imports, proportionally, covering points of collection, storage, packing and trade of products of plant origin. The official laboratories, analysing samples for pesticide residues are accredited and participate in the Community Proficiency Tests. The methods of analysis used by the laboratories comply with the criteria set out in relevant EU law provisions and other adopted technical guidelines. In a case of an MRL exceedance, before any administrative and punitive enforcement action is taken, a default analytical uncertainty of 50% is subtracted from the measured value. If this figure still exceeds the MRL, enforcement action relevant to the case is taken.[64]

Toxicological Studies. Are Pesticide Residues Harmful?

Pesticides can be toxic to man, animals, plants and the environment if improperly used. Several early insecticides and fungicides contained highly toxic heavy metals (arsenic, lead, and mercury) or carcinogenic substances. Most of these early pesticides are no longer approved for use and many were banned many years ago. Advances and developments in the agrichemical industry have turned out modern pesticides that are much less toxic to the applicator and environment but still control the pests that damage crops, livestock, forests, and home gardens. In the last decades new environmental laws and toxicological regulations for pesticides evaluate with scientific tests the potential to cause immediate harm to people through ingestion (eating), contact with skin, breathing fumes, or spills in the eyes. Harm from exposure to low doses (residues in food) over a long period of time is also examined. Also pesticides are evaluated for potential to cause birth defects, cancer, reproduction problems, and mutations. Only after the potential pesticide has been determined to be within the allowable, established limits of risk is the registration process continued. Tolerances are maximum legal limits (after rigorous toxicological tests to animals and long term use in the fields) of pesticide residues that can occur in food and are normally set 100 times below the level that might harm people or the environment.

Pesticide use and health effects on farmers and agrochemical applicators

The majority of toxicological studies on pesticides in the last decades focused on farmers, pesticide applicators, harvesters of sprayed crops and general populations living in the countryside, who have the most dangerous exposure to pesticides. A recent review on pesticide health effects (2014) to farmers covered more than 120 scientific studies (mostly epidemiological research). Cancer, neurological effects and acute toxicity (poisoning) were the most important health threats to farmers and agricultural populations.[65,66]

Pesticide poisoning is a global public health problem. In the 1990s the WHO estimated three million acute poisonings occur worldwide each year (with 220,000 deaths, most of them intentional), of which pesticide-related poisonings were more frequent and serious in developing countries. This was the result of lack of health and safety measures among farmers, lack of training and absence of state regulations on pesticides (it is estimated that 25% of developing countries do not have regulations).[67,68,69,70] Self-poisoning with agricultural pesticides represents a major hidden public health problem accounting for approximately one-third of all suicides worldwide. It is one of the most common forms of self-injury in the developing and Third World countries in Asia, Africa and Latin America. The WHO estimates that 300,000 people die from self-harm each year in the Asia-Pacific region alone.[71]

Long-term pesticide exposure and risk of developing cancer was is one of the subjects that was investigated during the last decades. Of the studies found in the scientific literature, more than 40 analyze the relation between direct exposure to pesticides and the risk of cancer. Most of the studies use the well known to the scientific community Agricultural Health Study (AHS) data (USA). The results showed that 12 of the studies report no significant evidence of increased risk of cancer among farmers exposed, while 31 studies concluded that exposure to certain pesticides significantly increases the risk of cancer. The heterogeneity of the results was related to the type of cancer, the time of exposure and the nature of the pesticides.[72-77]

Also, scientists explored the association of health effects of pesticide by indirect exposure among agricultural populations. From the data of 16 studies of cancer risk and indirect pesticides exposure no consensus is reached. Five studies find evidence associating pesticide exposure (environmental or prenatal exposure) with increased risk of childhood leukemia, while other studies found no significant association between increased risk for certain diseases and pesticide exposure.[78,79,80] In the case of depression and neurological effects associated with indirect pesticide exposure, 11 studies were found in the scientific literature. Research looked at neurobehavioural development, Parkinson's diseases, and effects on children's IQ scores. The results are mixed with some positive results for specific pesticides with long-time exposure. Even when exposure is indirect the risks of neurological damage may increase, especially for children whose exposure (organophosphates) takes place during early stages of fetal development. [81]

Pesticide residues health effects in fresh food and food products

The safety of toxic substances in food products is monitored by the so called the "**Total Diet Studies**" which analyze pesticide residues in a "typical meal." Information on what people eat or what is a "typical meal" is collected by various organizations (e.g. the USDA's Nationwide Food Consumption Surveys and the FDA). Food monitored in the Total Diet Studies may be analyzed for nearly 200 different pesticides (Cornell University Program on Breast Cancer and Environmental Risks Factors, Pesticide residue monitoring and food safety, http://envirocancer.cornell.edu/factsheet/Pesticide/fs25.foodSafety.pdf).

The Food Standards Agency (FSA) in Great Britain contacts rigorous safety assessment to make sure that any pesticide residues remaining in the crop will not be harmful to people. Pesticides are also reviewed regularly and if a review highlights any areas of health and safety concern then more data may be sought, or the approval may be modified or withdrawn completely. Pesticide residues in food and drink in the UK are monitored through an official surveillance programme conducted by the Chemicals Regulation Directorate (CRD) and overseen by the Defra Expert Committee on Pesticide Residues in Food (PRiF). [Food Standards Agency, Pesticides, https://www.food.gov.uk/business-industry/farmingfood/pesticides].

Most of the studies were related to cancer risks. Consumption of fish, water, seafood, milk and other dairy products were associated with a small but statistically significant association between cancer risk and some specific pesticide residues (DDT, organochlorines and PCBs (polychlorinated biphenyls). Most of these pesticides have been banned (the DDT from 1974) and were replaced with others that are more effective and less toxic. International and national authorities carefully evaluate human and ecological risks from exposure to pesticides, including occupational exposure. Also, they place special emphasis on children's health in making regulatory decisions about all pesticides. Some pesticides which are probably carcinogenic to experimental animals at high concentrations (much higher than residues in food) are regulated to be used on special cases and only when their concentration do not exceed MRL levels.[82,83] Some polychlorinated compounds that are allowed to be used under special regulations and for certain crops because of their effectiveness to stop highly damaging pests and insects in some countries. In most developing countries of Asia and Africa regulations on pesticide use are hard to apply and residues appear in some food commodities. [84,85,86]

The links between health risks and pesticide residue exposure depend on the concentrations observed in food. Margarine and butter in Poland were examined for organochlorine pesticides. Human and environmental health risk assessment was carried out by the estimation of lifetime average daily dose (LADD) and non-carcinogenic health hazard quotient (HQ). Results showed that OCP did not pose an immediate danger to the consumers' health.[87] Another study for the classification of risk to human from levels of contaminants in food in Belgium investigated also pesticides: The results showed that metals , such as As and Pb were classified as priority 1 (high concern) for food safety, Cd, MeHg, dioxins, PCBs priority 2 (medium concern), various pesticides, polychlorinated pesticides and metabolites, and polychlorophenols priority 3 (low concern).[88] Risk evaluation for human health was carried out for dietary intake of PCBs from edible fish. Experimental data were compared with the new EU legal level and the EPA risk estimations. The dietary intake of PCBs, as WHO-TEQ (toxic equivalency) per kg body weight (b.w.), was below 8 pg TEQ/kg body weight/week, while the new EPA approach suggested that the chronic effects do not represent any type of danger for human health.[89]

The pesticide residues found under the Good Agricultural Practices in food commodities, especially in fruits and vegetables, are relatively low. At the same time MRLs are set at 100 times lower (safety margin) than toxicological concentrations in experimental animals. Although some residues may remain at the time of harvest, residues tend to decline as the pesticide breaks down over time. In addition, as fruit and vegetables are washed and processed prior to sale the residues often diminish further. Sampling and new technological developments in pesticide use in agriculture showed in the last decade that only 2,5-3% of samples exceeded the MRLs (National Pesticide Information Center. Pesticide residues in food, Oregon State University and U.S. EPA, http://npic.orst.edu/index.html).

Risk assessment for pesticide residues in food was the subject of the Joint FAO/WHO Meeting on Pesticide Residues (JMPR). Health scientists, toxicologists and food specialists established a safe intake level for consumers taking into account the daily dietary intake. MRLs are enforced by national authorities to ensure that the amount of pesticide consumers are exposed to in the food they eat over a lifetime will not have adverse health effects (WHO. Pesticide residues in food? Available at http://www.who.int/features/qa/87/en/).

Several scientists in the USA argued with papers in the scientific literature that several independent exposure studies suggested that the FDA dietary pesticide residues are reasonable estimates of average human exposures. Using standard methodology and measured dietary pesticide residues in the TDS (Total Diet Study), the estimate of excess cancer risk from average lifetime exposure to synthetic pesticide residues in the diet appears to be less than one-in-a-million for each of the ten pesticides for which adequate data were available.[90]

Health Risks or Benefits from the Consumption of Fruit and Vegetables?

A high number of studies showed that daily consumption of five servings of fruit and vegetables is considered as an important part of diet for good health and prevention of premature morbidity and mortality from cardiovascular and cancer diseases, especially the digestive tract. Scientific evidence has accumulated supporting the notion that increased intake of fruits and vegetables reduce cardiovascular risk. It is clear that fruit and vegetables should be eaten as part of a balanced daily diet, as a source of vitamins, fiber, minerals and phytochemicals.[91,92]

A recent review, estimated the additional risk of cancer that can be attributed to increased consumption of fruit and vegetables with pesticide residues compared to the prevention of cancers by increasing consumption by one serving each per day (e.g. one apple). The cancer prevention estimates were derived using a published meta-analysis of nutritional epidemiology studies (many studies showed that fruits and vegetables contain antioxidants and other vitamins that prevent initiation of carcinogenic mechanisms). The cancer risks were estimated using U.S. Environmental Protection Agency (EPA) methods, cancer potency estimates from rodent bioassays, and pesticide residue sampling data from the U.S. Department of Agriculture (USDA). The resulting estimates are that approximately 20,000 cancer cases per year could be prevented by increasing fruit and vegetable consumption, while up to 10 cancer cases per year could be caused by the added pesticide consumption. These estimates have significant uncertainties (e.g., potential residual confounding in the fruit and vegetable epidemiologic studies and reliance on rodent bioassays for cancer risk). However, the overwhelming difference between benefit and risk estimates provides confidence that consumers should not be concerned about cancer risks from consuming conventionally-grown fruits and vegetables.[93] The health benefits of eating fruit and vegetables, in contrast to the perception of dangerous pesticide residues from conventional agricultural products, is supported by the consumer organizations of the USA and other developed countries. [94]

Fruit, vegetables, beans and nuts are very important for a healthy diet because they contain polyphenolic compounds, such as flavonoids. Flavonoids are bioactive compounds, with antioxidant, anti-inflammatory and anti neurodegenerative properties. Flavonoids are found in foods such as tea, chocolate, red wine, fruit, vegetables, beans, nuts and spices. A great number of studies linked higher intakes of specific flavonoids and flavonoid-rich foods with reduced mortality from specific cardiovascular diseases and cancers. However, the importance of flavonoids in preventing all-cause mortality remains uncertain.[95,96]

Figure 6. Flavonoids and vitamins in foods are bioactive compounds with very important properties: antioxidant activity, cell signaling, stimulate phase II detoxification enzyme activity, inhibit proliferation and induce apoptosis, inhibit tumour invasion, angiogenesis, anti-inflammatory action, decrease platelet aggregation, etc. [http://lpi.oregonstate.edu/mic/dietary-factors/phytochemicals/flavonoids].

The WHO recommends an intake of 5-8 portions (400-600g) daily of fruits and vegetables to prevent risk of cardiovascular disease, cancer, poor cognitive performance (neurodegenerative), and other diet-related diseases, as well as for the prevention of micronutrient deficiencies. Much of their potential for disease prevention is thought to be provided by polyphenolic compounds and especially flavonoids.[97,98,99]

Fruit and vegetables are important elements of a balanced diet offering vitamins, minerals, fibre, some energy (sugar) and certain antioxidant phytochemicals which are potentially beneficial for human health. Epidemiological studies have shown that high intakes of fruit and vegetables at a young age and in later in life are associated with a lower risk of chronic diseases. The problem of pesticide residues carries a much lower risk than avoiding to eat adequate portions of fresh fruit and vegetables. Despite the scientific facts, the presence of pesticide residues in apples (one of the fruits that is consumed daily) raises serious health concerns among parents, especially if they are consumed by children. A study in Poland followed a 9 years of investigation (2005–2013) of 696 samples of Polish apples for 182 pesticides. The study focused on the health risk for children, adults and the general population consuming apples and exposed to specific pesticide residues. A deterministic model was used to assess the chronic and acute exposures that are based on the average and high concentrations of residues. Children were the group most exposed to the pesticides. The results indicate that the occurrence of pesticide residues in apples could not be considered a serious public health problem. Nevertheless, an investigation into continuous monitoring and tighter regulation of pesticide residues is recommended.[100]

A recent European epidemiological prospective studies established that consumption of fruits and vegetables is associated with a lower overall mortality (450,000 participants, European Prospective Investigation into Cancer and Nutrition, 25,682 were reported deceased after 13years of follow-up). The study found that participants reporting consumption of more than 569 g/day of fruits and vegetables had lower risks of death from diseases of the circulatory, respiratory and digestive system (0.85, 0.73 and 0.60 respectively), when compared with participants consuming less than 249 g/day. Raw vegetable consumption was additionally inversely associated with death from neoplasms and mental and behavioural disorders.[101]

Detection of Pesticide Residues in Greek Food Commodities

Greece as a member of the EU reports every year collects samples of food products and performs analytical determinations of residues (Directive 2002/63/EC). The results of pesticide residues in food commodities and the statistical analysis appears in the annual Report of European Food Safety Authority. In addition, Greek scientists in the last decades extended their research and analytical techniques in the determination of concentrations of various pesticides of Greek agricultural products foods (olive oil, grapes, apples, fish, etc) and their processed food items.

Olive oil and pesticide residues.

Olive oil represents a very important agricultural product and Greece exports substantial amounts of olive oil and olives every year to other countries. Organophosphorous pesticides (OPs) and their metabolites were analysed (2004-2005) in 167 samples in virgin olive oil from various areas. 30% of samples contained detectable levels of OPs (fention, fenthion sulfoxide) and only one sample contained dimethoate above MRL. A cumulative risk assessment was performed using the hazard index and toxicity equivalence factor. The results indicated that there is neither acute nor chronic risk for the Greek population through olive oil consumption.[102]

Another study determined 35 pesticides in 100 olive oil samples. The highest detection rates were observed for the residues of fenthion, dimethoate and endosulfan. For the multiresidues of pesticides present in olive oil, there is no zero risk in consumption of olive oil. However, the exposure of olive oil consumers to each detected residue was far below the acceptable daily intake (ADI).[103] Olive oil samples from Crete (1997-1999) were analyzed for fenthion and dimethoate. All olive oil samples contained residue levels lower than the MRL' levels. The organic olive oil contained significantly lower concentrations of the two pesticides. The levels of fenthion and dimethoate in organic olive oils exhibited a decreasing trend following the implementation of the new cultivation method.[104]

An extensive study was contacted with a total of 815 samples from selected agricultural products collected in the period 2005-2009 by Bio-Hellas Control Certification Body. The study was sponsored by the research funds of the Center of Toxicological Sciences and Research of the University of Crete. Bio-Hellas is the biggest Control and Certification organism in Greece supervising a great variety of agricultural products such grapes, olives, olive oil, fresh fruits, vegetables etc. The study focused on 381 pesticides analysed by sensitive analytical techniques, GC/MS-MS and HPLC/MS-MS. Pesticide residues determination were for chlorpyriphos, cyfluthrin, a-cypermethrin, cyhalothrin, deltamethrin, diazinon, dimethoate, endosulfan, fenthion, fenthion sulfone, fenthion sulfoxide, malathion/Malaoxon, methidathion, methomyl, parathion, etc. Only 3.06% of total samples exceeded the MRLs. The problem occurred in the case of dimethoate (in grapes), endosulfan, a-cypermethrin, chlorpyrifos and diazinon (all in olive oil samples), etc. The most frequently detected pesticide residues were organophosphates, organochlorines, carbamates and pyrethrins. The risk assessment was performed by the HI method (Hazard Index). According to selected parameters for the risk assessment (food commodities, consumption, body weight) pyrethrins (HI value 0.05288) and organochlorine (HI value 0.087) presented a negligible hazard for the consumers. Organophoshates (HI value 0.34) and carbamates (HI value 0.38) were also far below one (1). All samples from biological cultivations (grapes and olive oil) presented a minimum acute risk from detected pesticides.[105, 106]

Virgin olive oil consumption and exports from Greece prompted the determinations of pesticide residues in the last decade. 48 samples of virgin olive oil (1999-2002) were collected directly from olive mills in the island of Corfu for insecticides sprayed from the air and/or from the ground. Determination was performed by GC-MS. The most common pesticides were fenthion and its oxidative metabolites. Concentrations of total fenthion in the positive olive oil samples were below the Codex Alimentarius MRLs. Only three samples contained total fenthion residues that exceeded the MRL[107] In the last decade some of the olive oil exported from Greece have protected geographical indication. Seventy (70) olive oil samples with protected geographical indication or designation of origin were analyzed for 51 pesticide residues by GC-MS. From the 70 samples only 4 contained pesticide levels exceeding MRLs. However, the investigated samples showed decreased occurrence and levels of pesticides residues in comparison with previous studies concerning samples from Greek conventional and organic cultivations. The Cretan olive oil samples showed the lower detection rates and the lowest average number of detected pesticides.[108] Virgin olive oil samples (167) were investigated for organophosphorus pesticide (OPs) and their metabolite residues during a 2-year (2004-2005) sampling campaign. The Hazard Index (HI) and Toxicity Equivalence Factor (TEF) were calculated, taking into account that OPs share the same toxicological mechanism. The results indicate that there is neither acute nor chronic risk for the Greek population through olive oil consumption.[109]

Fresh milk and cheese pesticide residues

Fresh milk and cheese are two food products that were analyzed for pesticide residues. In a study 38 samples of bovine milk and 28 samples of 3 types of cheese were collected throughout Greece during 1991-1992, and analyzed for 13 organochlorine insecticides, 3 herbicides, and one organophosphorus insecticide. Eleven milk samples (28.9% of analyzed samples) contained residues of one or more of the following: lindane, α-isomeric hexachlorocyclohexane, 1,1-bis[p-chlorophenyl]-2,2-dichloroethylene (metabolite of DDT, p,p'-DDE), and methyl parathion at concentrations below the MPLs. Overall all the samples of milk and cheese analyzed contained concentrations below the MRL of FAO/WHO Codex Alimentarius permitted by the European Union.[110] Another study investigated residues from samples of milk of Greek dairy sheep and goats, fed mainly with supplementary feed during the winter months. 200 milk samples from sheep and goats were collected from 10 farms representing conventional production and feeding systems (alfalfa hay, wheat straw, etc) in Greece. Milk and feed samples were analyzed for pesticides residues. The results showed that the ∑endosulfan (total endosulfan α and β) in all the concentrates samples (mean con. 5.36 $mg.kg^{-1}$) was much higher from the MRL. In the wheat straw, shrubs and pasture samples no pesticides residues were detected. No pesticide residues were detected in milk samples of sheep and goats. The results indicated that milk from the farms sampled presented no human health risks.[111] Organochlorine insecticide DDT (persistent insecticide) was banned in Greece from 1977. Despite the prohibition still residues of the metabolites have been detected in human tissues and milk. A recent study analyzed 196 cow milk samples from the Greek market by GC-MS analysis. In 97.4% of the samples at least one DDT isomer or one metabolites was detected, in levels well below the MRL levels. The health risk was calculated by using the Hazard Index (HI) for both carcinogenic and non-carcinogenic effects and was found below 0.109 (negligible). HI values of EDI/RfD (Estimated Daily Intake/Reference Dose) and EDI/PTDI (Estimated Daily Intake/Provisional Tolerable Daily Intake) were far below 1, which indicates no significant health risk for children. The values for cancer risk were well below the EPA threshold of unacceptable cancer risk.[112]

Pesticide residues in fruit and vegetables. Rapid Alert system in EU

Greece is the 4th largest producer of fruits in the European Union. According to FAO, in 2005 Greece produced 4.5 million tons (or tones=1,000 KG) (grapes accounted for 27% of total fruit production, oranges 22%, peaches and nectarines 15%). Also, Greece in 2005 was the 7th largest producer of vegetables in the EU (3 million tons) (tomatoes 56% of total vegetable, onions 7%, cabbages 6%, cucumbers 5%). In 2005 Greek fruit exports were €339 million for 696.000 tons, and exports of vegetables were €73 million for 53.000 tons. Also, Greece imports every year fruits and vegetables from other countries, such as bananas, apples, pineapples, lemons, tomatoes, potatoes, etc.

The European U requires that every of imported consignments of many plant products undergo phytosanitary inspection upon arrival in the EU unless the plant products qualify for reduced inspection levels. The requirements and products are detailed in Council Directive 2000/29/EC or plant health legislation. Border officials will check documents and a physical plant health check to verify compliance with EU import requirements. More detailed information can be accessed on DG Health & Consumer Protection's website: http://ec.europa.eu/food/plant/organisms/imports/inspection_en.htm .

The EU has one of the highest food safety standards in the world. A key tool to ensure the cross-border follow of information when risks to public health are detected in the food chain is **RASFF** (Rapid Alert System for Food & Feed).[available at http://ec.europa.eu/food/safety/ rasff/index_en.htm]. RASFF (started in 1979) enables information to be shared between its members (EU-28, national food safety authorities, EFSA, Norway, Liechtenstein, Iceland and Switzerland) and provides a round-the-clock service to ensure that urgent notifications are sent. Thanks to RASFF, many food safety risks had been averted before they could have been harmful to European consumers. Every year RASFF records the cases of excessive use of pesticide residue and issues notices to EU countries and imported food items from other countries. According to official EU data, Greece was once again among the member-states where fewer cases of excessive use of pesticide residue that have been detected in plant products [available at http://greece.greekreporter.com/2014/07/04/greek-fruit-and-vegetables-among-the-safest-in-eu/].

This section relied on a search of the scientific literature of the last 20 years on pesticide residues in fruits, and vegetables in Greek food products. It does not include validation studies of multi-residue methods for the determination of pesticide residues from Greek laboratories.

Greek apples, grapes, peaches, apricots

Greece is producing thousands of tones every year of apples, pears and other fruits. In 2013 total production of apples was 228,000 metric tons (MT), pears 39,500 MT (GAIN Report. Global Agricultural Information Network, gain.fas.usda.gov). A high percentage is exported to European countries. A study investigated 80 Greek apple samples with protected geographical indication or designation of origin from various areas of Greece. The study analyzed 51 target pesticides. 12 pesticide residues were detected in 75 positive samples. The highest detection rates were observed for chlorpyrifos (*n*=75), quinalphos (*n*=75) and parathion (*n*=73). Only 2 of the 80 samples contained pesticide residues (parathion-methyl) exceeding the MRLs.[113]

Figure 7. Greek agriculture produces very good quality fruits. In 2014 the total supply of apples was 248,022 (metric tons, MT), total exports 66,700. Pears 43,000, t.e. 4,510. Grapes 299,182, t.e. 69,000. Cherries ~60,000. Peaches ~750,000. Nectarines ~100,000 (selected from various sources).

Grapes is another important food commodity produced and exported in large amounts from Greece every year Total production in 2013 was 301,134 metric tons. An investigation aimed to assess residues of Azoxystrobin (fungicide) on fresh and washed grapes and raising following processing. The pesticide has an MRL of 2 mg/kg of grapes. The results showed that residues on grapes were at concentrations 0.49-1.84 mg/kg, and after washing 75% of the pesticide is removed. In raisings produced from seedless grapes after treatment the concentrations found were t 0.51-1.49 (treatment 1) and 1.42-2.08 mg/kg (treatment 2).[114] Other studies investigated residues of cypermethrin in field-treated grapes and raising under various conditions.[115] Also, the degradation of methamidophos on soultanina grapes was observe during refrigeration. The results were correlated with relevant results from the decomposition of the same pesticides on apples on the trees and during refrigerated storage. These correlations are suggesting that biological factors strongly affected the decomposition rate of azinphos methyl. On the contrary the decomposition of parathion methyl was mainly affected by environmental rather than biological factors.[116]

Peaches is another fruit produced in Greece in large amounts and exported annually. A study collected peaches cultivated by two methods. 150 samples of peaches were collected pre-harvest from Pella and Imathia districts, Macedonia (June–August 2001). 55 samples were from conventional and 95 from Integrated Crop Management (ICM) cultivation (more environmental friendly method with regulated pesticide application and management). The residue levels of selected insecticides, fungicides and acaricides in peach samples were determined by GC-MS. The concentrations of all detected pesticides were lower than the MRLs. Only chlorpyrifos residues were detected in 7% of peaches at levels higher than the MRLs grown by the conventional system. The present study indicated that ICM cultivation has a higher efficiency in terms of product safety and quality.[117] The study with peaches was extended and a total of 1,150 samples were collected (2002-2007) and analysed by GC-MS for 31 types of pesticide residues. Traceable levels of pesticides were lower than MRLs in all peach samples. 22 of pesticide residues were measured above detection limit, and 8 of pesticides were found present in peaches every year.[118] A highly specific analysis was applied to the analysis of 32 samples of apricots and peaches from Greece. The apricot samples originated from Corinth and Nauplion in the Peloponnese and the peach samples were from the area of Veria in North Greece. No residues of the target pesticides were detected in 22 of the samples. Chlorpyrifos was detected in one apricot and five peach samples at concentrations from 0.01 to 0.06 mg/kg (MRL 2.0). Parathion methyl was detected in four peach samples from 0.02 to 0.07 mg/kg (MRL 0.2). The pesticides detected in one sample only were bromopropylate 0.36 mg/kg (MRL 2.0), phosalone 0.14 mg/kg (MRL 2.0) and azinphos methyl 0.05 mg/kg (MRL 0.5).[119] Peaches from Greece were analysed for 23 pesticides by a multi-residue method using selected ion monitoring mode GC-MS. The analysis of 104 fruit samples collected under Integrated Pest Management (IPM) production during the 2006 cultivation period. Residues detected were lower than those established by legislation (MRL) for all pesticides, except diazinon, where one (1) positive sample was detected containing 0.03 mg/kg.[120]

The methodology used in the application of pesticides and adequate days before harvest (recommended by the manufrasturers on the label) is very important for lower residue levels in the fruits. A study in Greece investigated the residue levels of pesticides chlorothalonil, iprodione, bupirimate, pirimicarb, chlorpyrifos and fenoxycarb in different peaches and nectarine cultivars. Analysis by Gas Chromatography found that, with the exception of chlorpyrifos, the residue levels of all pesticides were lower than the MRLs in all samplers. The detected levels of chlorpyrifos were higher than the MRLs in one cultivar 7 days after application, but dropped to very low levels 27 days after application. All pesticides showed a reduction of concentrations over time. Overall, the pesticide regime (applied well in advance and with environmental friendly methodology) gave residue levels much lower than those of MRLs, in all peaches and nectarine cultivars.[121]

The agricultural sector is Greece contributes more than 3% of the GDP using up 12.6 % of the working population (2012). The agricultural sector in Greece uses private laboratories accredited to analyse its fruit, vegetables and other products for pesticide residues. The Hellenic Accreditation System (ESYD) was established by the Law 3066/2002 [http://www.esyd.gr/portal/p/esyd/en/esyd.jsp]. For example, The AGRALAB RDS laboratory (Markopoulo, Attika, Greece) is equipped with modern, very sensitive analytical instruments, such as Liquid Chromatography double mass (UPLC-Ms-Ms), Gas chromatography double mass (GC-Ms-Ms), HPLCs (High Performance Liquid Chromatography) and GCs (Gas Chromatography) with various detectors, to conduct analyses of fruit, vegetables, olive oil, tobacco, cereals, flour and pulses [http://www.agrolab-rds.gr/default.aspx?lang=en-US&page=297].

Greek vegetables, tomatoes, etc.

Greece is a big producer of vegetables, especially tomatoes, in Europe. There are three types of tomato production: industrial, from greenhouses and from open-field cultivation. Greece produces 1,3-1,5 million tons of tomatoes in the last decade, but also imports every year tomatoes from other Balkan countries. Canned tomato production is around 500,000 tons annually. Tomato growers in greenhouses use pesticides to control plant diseases. Most of the applied pesticides (procymidone, progargite) are degraded with time and their application is permitted some days from preharvest time.[122] Tomato processed products (ketchup, puree, homogenates, paste, etc) which are used in cooking contain lower concentrations of residues because thorough washing, storage and cooking accelerate the lowering of the concentrations. A study in Greece investigated the effect of storage at 5°C and of thermal processing by cooking at 100°C and sterilization at 121°C for 15 min on pesticide maneb residues (and its toxic ethylenethiourea, ETU, metabolite) in tomato homogenates. No significant loss of maneb (fungicide) was observed during cold storage for up to 6 weeks, but thermal treatment resulted in substantial degradation of maneb. After cooking, only 26% of initial maneb residues remained in the samples, whilst the conversion to ETU was 28%. Sterilization eliminated the residues of the parent compound giving rise to conversion to ETU up to 32% (mol mol^{-1}).[123]

It is well known that pesticide used in raw agricultural products are degraded substantially with time and their concentration gets lower after application. Normally, their application is recommended to be few days before harvest. Postharvest storage and food processing (washing, sun drying, boiling, milling, etc) involve aqueous hydrolysis, photolysis in water and air, biodegradability that can be achieved under aerobic and anaerobic conditions and especially during cooking at high temperatures.[124]

Spinach is used for baby and infants foodstuffs. The pesticides diazinon and pirimiphos-methyl (lately banned in Europe) are commonly used for the cultivation of spinach. But the process of trimming, washing, and boiling decrease substantially the concentrations of their residue on the final spinach product. Experimental crop of spinach was cultivated and collected using all steps of common agricultural practices. Residues were analysed by GC multiresidue method, as fresh or after washing and boiling. Insecticide residues dissipated considerably between 0 and the 7th day of sampling and all sample concentrations were found to be much lower than the MRLs. The risk assessment to the consumer of spinach foodstuffs was evaluated by standard methods. It was estimated that the long-term intake by consumers ranged from 0.27% to 13% of the acceptable daily intake (ADI) for diazinon and from 0.03% to 1% for the pirimiphos-methyl. In conclusion, these pesticide residues, despite their toxicity, decrease substantially in the prepared food at levels that have very low risk to consumers.[125]

Many scientific investigations were focused on pesticide degradation under various food processing techniques. It is well known that washing, sun-drying, peeling, thermal processing treatments (pasteurization, blanching, boiling), cooking, steaming, canning, scrambling etc. have been found valuable in degradation of various pesticides. Many other food techniques like refining, fermentation and curing have been reported to affect the pesticide level in foods to varied extent. Milling, baking, wine making, malting and brewing resulted in lowering of pesticide residue level in the end products. Post harvest treatments and cold storage have also been found effective in decreasing the concentration of pesticides below MRLs.[126]

Nitrate content of vegetables.

Nitrate is an essential plant nutrient found in soil that is taken in by all plants as a primary nitrogen source. Industrial fertilizers also contain nitrates and some of their residues can be found on the outside of fruits and vegetables. Some plants have much higher levels of nitrates than others. Nitrates in spinach, lettuce, parsley, beet leaves and roots, radish, carrot, cabbage, broccoli, cauliflower, cucumber, tomato, eggplants, etc., from the vegetable Market of Thessaloniki, were determined (1995-1996). Samples of the above vegetables had nitrate concentrations in excess of 500 mg/kg. Nitrate concentrations in all cases were lower than values reported in Northwestern Europe and more important, they were always below the maximum permissible values.[127]

Pesticide residues with endocrine disrupting properties

The concern for pesticide residues has increased lately for the organic virgin olive oil produced in Greece and the possible endocrine disrupting properties. An initial programme (2004-2005) examined 100 olive oil samples for 35 pesticides from the 10 main olive oil producing regions in Greece. 71 samples were taken from olive mills and 29 samples came from local markets. 10 of these samples were organic olive oil. 10 % of samples had no residues of the pesticides. The three most commonly detected pesticides were dimethoate, fenthion and endosulfan. Overall, results of the study suggest all estimated daily intakes were well below the acceptable daily intakes. Commercially and individually processed extra virgin olive oils contained the highest amounts of the targeted pesticides, possibly as a result of farmers protecting their crops against the olive fruit fly, a pest that has the greatest impact on the quality of the olive oil. 6 out of 10 organic olive oils samples contained no detectable pesticide residues (European Commission, DG Environmental News Alert Service. Science for Environmental Policy. Greek olive oils contain no harmful levels of pesticides [available at http://ec.europa.eu/environment/integration/research/newsalert/pdf/13si2_en.pdf].

Organic olive oil in Greece has been investigated for pesticide levels with endocrine disrupting properties. Studies showed in Greece showed that organic olive oil is safer, although not free from pesticides. Pesticide residues levels detected in olive oil usually do not surpass MRLs. However, MRLs for pesticides, that are known to endocrine disrupting compounds, should be reconsidered, since endocrine disrupting action of pesticides is activated even at very low concentrations. In the case of Fenthion (found in olive oil) concentrations as low as 10 and 1 ppm are sufficient to interact with male hormones. Lypophilic pesticides may accumulate in the body fat at levels sufficient to cause endocrine disrupting action. Given the known heath risks associated with exposure to pesticides, steps should be taken to minimize their presence in food. This can be achieved by adopting organic agricultural methods and using the codes of “Good Agricultural Practices”.[128]

GreenPeace in Greece as a policy of the NGO every year issues an alarming survey for pesticide residues in food and the dangers to human health. On the 24/9/2013 posted a report through its website “Comments on Toxic pesticides residues in fruits and veggies in Greece: news of the systemic, the banned and even the illegal”. In this report GreenPeace described an alarming state of their tests, “**…..**Between April and June 2013, Greenpeace tested 34 samples of 11 kinds of fruits of vegetables that are sold in the Greek market, most of them of Greek origin. The samples were found to carry 32 highly toxic pesticide residues that are (according to the investigator) “dangerous for humans and the environment**”**. For instance, the market authorisation of several of these pesticides warns that: Thiacloprid – found in apples and pears- is likely that can be carcinogenic to humans, or Linuron – found in carrots- can harm an embryo during pregnancy. It is obvious that Greenpeace presents an alarming view with residues found in food commodities at very low concentrations below MRL. The dangerous properties of these compounds are for experimental animals receiving high doses in toxicological experiments. The report states **"….**Half of the active substances that were detected belong to the category of systemic pesticides. Which means that they do not stay only on the external skin of the fruits and vegetables but can enter their tissue. In addition, 8 of these active substances are known to also harm bees while some the pesticides that were detected are banned in the European Union for use in agriculture. It seems that in Greece there is a real problem with illegal trade of chemical pesticides which has been worsened with the economic crisis…". [available at http://www.arc2020.eu/2013/09/concern-over-toxic-pesticide-residues-in-fruit-and-veg-in-greece/].

Health Campaigns for Increased Consumption of Fruit and Vegetables

National and international (WHO) health authorities and medical agencies all over the world consider high urgency to campaign for increasing consumption of fruit and vegetables, especially among children as important components of a healthy diet. Although the presence of pesticide residues is of concern it is considered as a secondary issue after all the scientific studies. International and national recommendations for fruit and vegetable intake vary considerably throughout the European Region and USA, Canada, Australia and other developed countries. There are already national initiatives in place aiming at increasing fruit and vegetable consumption in children.

Figure 8. Posters from various national campaigns to eat fruit and vegetables every day. 5 a Day for Better Health or 5 am Tag (servings) is the main slogan in all countries

Examples of campaigns for such national and local programmes are:
SchoolGruiten – The Netherlands (http://schoolgruiten.kennisnet.nl/)
Frugtkvarter – Denmark (http://www.frugtkvarter.dk/)
5 am Tag – Germany (http://www.5amtag.de/)
Fruitness – Italy (http://www.fruitness.eu/)
5 al dia – Spain (http://www.5aldia.com/)
5 a day – United Kingdom (UK) (http://www.nhs.uk/LiveWell/5ADAY/Pages/5ADAYhome.aspx)
Un fruit pour la récré – France (http://agriculture.gouv.fr/un-fruit-pour-la-recre)

WHO recommends a daily intake of 400 g of fruit and vegetables per person per day. Surveys showed that that only 8% of adult consumers in the USA have the daily recommendation in their daily nutrition. The USA started in 2007 the campaign "***Fruit and Veggies-More Matters***" and spent 3-5 million dollars per year in promoting the idea of healthy nutrition in schools and in other social events. [129] Most of the national projects in Europe aimed at increasing fruit and vegetable consumption in children are school-based. Implementing programmes in schools ensures wide participation and gives the opportunity to combine different types of activities, such as traditional classroom-based learning, school gardening, cooking classes and feeding. Distributing fruit and vegetable (one fruit free every day in schools) as well as involving parents, teachers and peers also improves the results of school-based interventions. Involving parents is of great importance since parental intakes, encouragement and home availability of fruit and vegetables are factors with strong influence on children's consumption. Clear fruit and vegetable messages, involvement of the family and using a theoretical framework as the basis of the intervention have also been demonstrated to be advantageous.[130,131,132,133]

Consumer Surveys in Greece on Nutrition

A cross-sectional survey collected through a questionnaire the attitudes of Greek consumers towards conventional food products (integrated pesticide management) and organic fruit and vegetables. The findings of the survey suggest that consumers' level of awareness and information towards certification systems for conventional and organic products in Greece is inadequate due to lack of information to the public. The topic of food quality is not widely discussed in Greece. The study also reveals consumers' willingness to pay more for organic fruit/vegetable products because they think are healthier. Information about food quality in Greece is disseminated by nutritionists, health institutes and popular magazines.[134]

Other scientists suggest that consumers will be more informed for food quality by certification, designation of origin (PDO), and traceability of food products. Consumers in Greece are willing to pay a premium for food with one or more of the above-mentioned labels and help producers and processors to adjust according to market demand. A study in 2011 identified consumers' awareness, attitudes, and buying intentions toward food quality of fresh fruit and vegetables with data collected by interviewing 400 consumers in Thessaloniki. Results indicated that the most important factors affecting willingness to pay a premium are mainly related to positive attitudes toward healthy food, level of awareness, and, to a lesser extent, several socioeconomic characteristics.[135] Another survey was conducted to assess the self-reported food safety knowledge and food-handling practices of Greek young university students They completed a questionnaire containing 32 questions (food safety, handling, cooking and hygiene). Students correctly answered only 38% of food handling and 37% of food safety. Females obtained considerably better food-handling scores than males. An educational background relevant to food safety was a significant predictor of responding accurately to a wide range of study question.[136]

From the 1990s the organic food industry in Greece became a niche market, but high prices and restricted points of sale, especially for fruit and vegetables compared to conventional products, kept the organic market share at around 8-10% of the total.[137-141] The domestic market in Greece for organic products was estimated to be around EUR 60 million (2010). The market for organic products was developing slowly until 2010, when growth was halted by the economic crisis. Between 2011 and 2013, consumption of organic products fell by almost half. Supermarkets and specialised organic shops covering about 40% of the market. About 30% of the organic food products are exported. Key products for the growing export market are olive oil and olives, wine and to some extent fresh fruit, vegetables and feta cheese. Greek organic production is certified according to EU legislation on organic farming and other regulations, which is fully implemented; some farmers have an additional certification (Demeter). Some organic products, usually those which are to be exported, are certified according to the private standards of other countries (e.g. Germany and USA)[available at IFOAM Organics International. Greece. Organic Agricultural Products, http://www.ifoam-eu.org/].

The demand for organic foods is constantly increasing mainly due to consumers' perception that they are healthier and safer than conventional foods and contain lower concentrations of agrochemical residues. With regard to other food hazards and hygienic conditions, such as natural chemicals, microbial pathogens and mycotoxins there no clear differences between organic and conventional. Also, organic products are not more nutritious than conventional. [142,143,144,145]

Traditional Greek products Greece is justifiably proud of the high quality and palatability of a wide range of traditional foods (fermented meats, dairy products, fermented vegetables) that have long made the traditional Greek diet distinct. In the past, the manufacturing of these products was craft and empirical and presented differences from area to area, resulting in final products with variable microbiological, physicochemical and sensory characteristics. But in the last decades, this situation has drastically changed and traditional Greek products are now produced in well-equipped industrial units under strict processing and hygienic conditions. Recently, in the context of EU funded project (www.truefood.eu), an extended retail survey was carried out for a variety of traditional Greek food commodities. Studies showed that the intrinsic properties of foods attained at the end of the process, together with other technological characteristics (e.g. use of preservatives, chill temperatures, modified atmosphere packaging, etc.) resulted in absence of pathogenic microorganisms, and consequently, Greek traditional foods could be considered to have a good safety record.[146]

The Latest Scientific Studies of Pesticide Residue and Human Health

Research on pesticide residues health effects is a permanent theme of numerous studies and surveys in developed countries. Pesticide residues in food commodities is monitored continuously in most countries and dietary record surveys, especially for fruit and vegetables, are evaluated for adverse health effects. In this section we present selected studies of the last three years (from European and other developed countries) in the scientific literature and their conclusions on risk assessment.

A Hungarian study (2016) on pesticide residues monitoring data of organophosphorus pesticides and daily intakes concluded that the cumulative acute exposure of the population was not a health concern.[147] A study of organochlorine pesticide residues in goat milk in Poland observed that the average daily dose for the sum of the organochlorine compounds were well below the acceptable daily intake (ADI) and their hazard quotient was very low, much less than 1, meaning no health concern ($3.4x10^{-3}$- $5.5x10^{-2}$).[148]

The average hazard quotients and hazard index for organophosphorus (OP) pesticide residues in fresh vegetables were estimated in a provincial area of China (Changchun).The results of the study showed that both were less than one and 0.46 respectively. The conclusion of the study was that inhabitants who are exposed to average OP levels may not experience adverse health effects from their diet.[149] Children's diets are of great concern. A study in Israel focused on the risks posed by pesticide residues in children's diets. Researchers investigated potential exposures to food pesticides in 301 urban Israeli children (2008-2010) through a food questionnaire. The Israel's national pesticide monitoring program estimates uptake for 26 pesticides in 27 fruits and vegetables. The surveyed children had higher potential exposures than the general population. Methamidophos, fenamiphos, iprodione, and oxydemethon methyl exceeded the Acceptable Daily Intake (ADI) even though the residues detected were below the statutory limit (children eat more fruit and vegetables and have lower weight than the average population).[150]

The total diet intake (TDI) was used in a study in Hong Kong for exposure to organochlorine pesticides (OCP) residues of adult population. The study included the most persistent organic pollutants-pesticides (Stockholm POPs Convention, "dirty dozen", such as aldrin, dieldrin, chlordane, DDT, endosulfan, lindane, mirex, etc). The lower- and upper-bound mean exposure estimates of OCP residues ranged from 0% to 0.5%. The results indicated that dietary exposures to the OCP residues would be unlikely to pose unacceptable health risks to Hong Kong adult population.[151] China's urban population increased dramatically in the last decade with similar increase in food consumption. In this part of the population there is growing concerns about food safety and stricter pesticide residues national standards. The attention of consumers has focused on vegetables and fish as they are an important part of the Chinese daily diet. A recent study used questionnaires distributed in the population of major regions of a northern Chinese metropolis. Samples of fruit-like, vegetables, leafy and root vegetables, and five species of fish (freshwater and marine) were collected and analysed for organochlorine pesticide residues (OPR). The results showed that the estimated daily intakes (EDIs) and hazard ratios (HRs) for OPRs in vegetables and fish in this area were inside safe limits.[152]

A large scale French study (2007-2009) assessed the chronic dietary exposure to pesticide residues using a total diet study (TDS). The study analysed 325 pesticides and their transformation products. Sampling corresponded to 194 individual food items that cover 90% of the adult and child diet. The study used 19,000 food products in 36 French cities. The results showed that 37% of the samples contained one or more residues, 73 pesticides were detected (pirimiphos-methyl, chlorpyrifos, iprodione, carbendazim and imazalil, mainly in fruit and fruit juices). For 90% of the pesticides, exposure levels were below the acceptable daily intake (ADI).[153]

The same research team extended the investigation with a large scale (2014) French Total Diet Studies (TDS) to measure exposure to food pesticide residues and other toxic chemicals in the French population aged 3 years and older. At the same time the French Agency for Food, Environmental and Occupational Health and Safety (ANSES) launched a specific TDS study on infants to complete its overall chemical food safety programme for the general population. More than 500 chemical substances were analysed in food products consumed by children (including nutrients, endocrine disruptors, polychlorinated biphenyls, dioxins and furans, brominated flame retardants, pesticide residues, bisphenol, etc.). Food commodities were selected based on results of a national population consumption survey. Also, a specific study on food was conducted on 429 households to determine which home-cooking practices are employed to prepare food consumed by infants. Chemical substances were analysed in more than 450 food samples, representing the purchase and home-cooking practices of over 5,500 food products. The sampling plan covered over 80% of the total diet. Specificities in infant food consumption and habits were therefore considered to define this first infant TDS.[154] Preliminary results of French system for dietary exposure to pesticide residues were published. The aims of the study was to assess acute and chronic risks to the general population and to identify food commodities with high concentrations of residues. The national monitoring programmes estimated the probability of exceeding the toxicological reference values (MRLs, for children and adults) for 522 pesticides and their metabolites. The majority of the pesticides (87%) were detected at low concentrations not exceeding MRL. This category scored at the lowest priority level 1. Scientists suggested that monitoring should be extended to include newly authorized substances in levels 2 to 4. Carbendazim, dimethoate, dithiocarbamates and imazalil merit particular attention as they scored at level 6 and are frequently quantified in fruits and vegetables, meaning that risk managers should take corrective measures in order to ensure safety.[155]

Despite the numerous studies and regulations on pesticide residues popular perception and environmental organizations argue that risk is very high and most of toxic pesticides should be banned. The difference according to some scientists is the meaning of scientific terms such as “hazard” and “risk”. Scientific studies on the potential health effects of hazardous pesticides base their classification (carcinogenic, neurotoxic, toxic, etc) on results with experimental animals (mostly rats and mice), and cell cultures. The adverse health effects depend on the dose and the route of exposure. But the same chemical can have different effects at different doses and the route by which the exposure occurs, e.g. ingestion, inhalation or injection. The extremely high doses used in experimental animals are not representative of the low doses that human are exposed from their diet. [156]

Other scientists argued that with global population projected to increase above 9 billion by 2050, food security—the availability of food and one's access to it—is increasingly important. Crop-protection products can help reduce yield losses caused by pests, pathogens, and weeds, to help feed the world's population sustainably. At the same time it is emphasized that given potential harm (for high concentrations and inappropriate use of agrochemicals despite the regulations for Good Agricultural Practices) for human health and the environment, regulation of pesticide use in agriculture has been controversial for many years.[157]

Recently high publicity was given to a new research from Harvard School of Public Health and Massachusetts General Hospital (research team by Dr Chiu) that presented important new data on semen quality in relation to dietary pesticide exposure via fruit and vegetable intake. The study utilized a novel approach that classifies fruits and vegetables into high versus low-to-moderate pesticide residue groups based on data from the Pesticide Data Program of USDA. Data obtained from a validated food frequency questionnaire concerning intake of fruits and vegetables with higher pesticide residues. The authors found that men (155 persons) who ate greater amounts (1.5 servings per day) of fruits and vegetables had 49% lower sperm count and 32% lower percentage of normal sperm than men who ate the least amounts (less than 0.5 serving per day), a finding which could have clinical and public health implications.[158]

The results promoted discussion among the scientific community. Other scientists were skeptical with the limitations of the study hoping that these findings should not discourage the beneficial consumption of fruit and vegetables. They focused on a number of important caveats (and limitations) to this research paper: i. the actual concentrations of pesticides present in the test groups’ bodies was never measured, and the researchers didn’t measure the actual level of pesticides on the fruit and vegetables being consumed either. That means that this study can only suggest a casual link between fruit and vegetable consumption and pesticides affecting changes in sperm quality, ii. test group’s diets were only assessed once (those diets could have changed), and iii. there were limitations with testing sperm quality at fertility center, the researchers used only a small sample of men who were probably attending these centers because they already suspected they had fertility problems. Other scientists suggested that other factors could be influencing the lower sperm counts, given that the Harvard results came from an observational study.[159,160]

In the last decade there were many studies of men visiting fertility clinics with semen quality problems. The results showed various problems (obesity, lack of exercise, life-style factors, diet, etc). Expert scientists commending on the results of the Harvard study note that know the findings can not be generalised to men in the general population. They emphasized that researchers did not have information on whether or not the food was grown conventionally or organically, and the exposure to pesticides could have been misclassified as it was not measured precisely for every individual man.
But despite the relatively small sample size and exposure assessment limitations, the paper makes a convincing case that dietary exposure to pesticides can adversely impact semen quality and need to be replicated in other settings and populations.[161]

Pesticide Residues in the Diet and Children's Health

From the 1960s the scientific community investigated the association of pesticide residues in food and children's adverse health effects. There was always a growing concern about children's exposure to pesticides in agricultural areas but also their special susceptibility to residues in food products. Children are not little adults, and may have higher exposures and greater vulnerability at both high and low levels of exposure to pesticide residues. Already, the review contains some studies on children's health. In this section we present some recent studies in the last 5 years.

A recent study (2015), following similar investigations, examined urinary 2,4-D (2,4-dichlorophenoxyacetic acid, herbicide, "possible carcinogenic", IARC 2B, with restricted use in many countries) and two measures of OP (organophosphate) pesticide exposure. The study found that were lowerurinary concentrations in children eating an organic diet (vegetables). Other frequently detected metabolites for pyrethroids, diethyl OP pesticides, and the herbicide metolachlor were not significantly lower during the organic diet phase. The study also found that diet was not an important exposure source for other pesticides (e.g., diazinon, malathion) in this population. According to researchers additional research is needed to clarify the relative importance of dietary and non-dietary sources of pesticide exposures in young children and determine the proportion of urinary metabolite excretion attributable to preformed metabolites.[162]

Following concerns for adverse gastrointestinal effects in children, prospective studies in USA, Germany and UK were designed to investigate whether there is any association between gastrointestinal effects and pesticide residue exposure (as measured by metabolite levels in urine and faecal samples) in young children. The study in UK examined 107 children (1-4 years of age) with background baseline samples and 26 provided samples when suffering from gastrointestinal symptoms. The results showed that there were no statistically significant differences between samples from children when healthy or unwell (they were in agreement with US and Germany). Samples from children suffering gastrointestinal symptoms were no more associated with anti-cholinergic pesticide metabolite levels or rotaviral infection than samples from healthy children.[163] Another U.S. study investigated exposure to OP pesticide residues from consumption of fruits and vegetables by children of farmworkers and non-farmworkers and across seasons (eastern Washington State, US) and urinary measurements . There were a few significant trends between dimethyl metabolites (DMAP) and fruit, vegetable or apple juice consumption; however, no clear pattern held across seasons or occupation. No significant trends were found between fruit and vegetable consumption and urinary measurements of diethyl (DEAP) metabolites.[164]

A recent study (2016) in Australia investigated the concentrations of persistent organic pollutants (POPs) and pesticide residues in infant foods and exposure by dietary intake. Fruit purees, meat and vegetables, dairy desserts, cereals and jelly foods were from Brisbane, Australia. The concentrations were very low (pictogram, 10^{-12} g/g) with most of analytes below the limit of detection. The study found that consumption of a 140 g meal resulted in intake ranging from 0 to 4.2 ng/day (1 ng $=10^{-9}$ g) of some POPs. Results from this study indicated that exposure for infants via dietary (in contrast to dust and breast milk) intake in Australia contributed only a minor component to total exposure.[165]

A Romanian study examined the behavior of 12 pesticides used in the treatment of a variety of apples (in a Romanian orchard) and exposure for adults and children. Five treatments were applied in recommended dosage with 23 days intervals between treatments. Pesticides degraded quickly in apples during the first days, when 30–50% from the initial concentration is lost. Pesticides residues at harvesting were below the MRLs applied in the EU, except for tebuconazole and chlorothalonil. The estimated lifetime exposure dose was calculated based on pesticide concentrations in apples at harvesting, and average fruit consumption. These doses for adults and children were below the reference dose (RfD) for each pesticide, suggesting a negligible risks for consumers.[166]

International and national health organizations, such as WHO, UNEP, National Resource Council (USA), National Pesticide Information Center (USA), Food Standards Agency (UK) have special information sites for advice and promote research on the effects of pesticides residues to children's health. [167-171] Rigorous legislation, monitoring and control of pesticide residues in food applies in Germany. The latest details can be found in the online database of the Federal Office of Consumer Protection and Food Safety (BVL, Bundesamtes fur Verbraucherschutz und Lebensmitelsicherheit). The BVL contributes to food safety in Germany, coordinates research programmes and ensures the application of EU legislation on pesticides and the European Rapid System for Food and Feed.[172] Public perceptions of pesticides in food by Germans were published by the Federal Institute of Risk Assessment .[173]

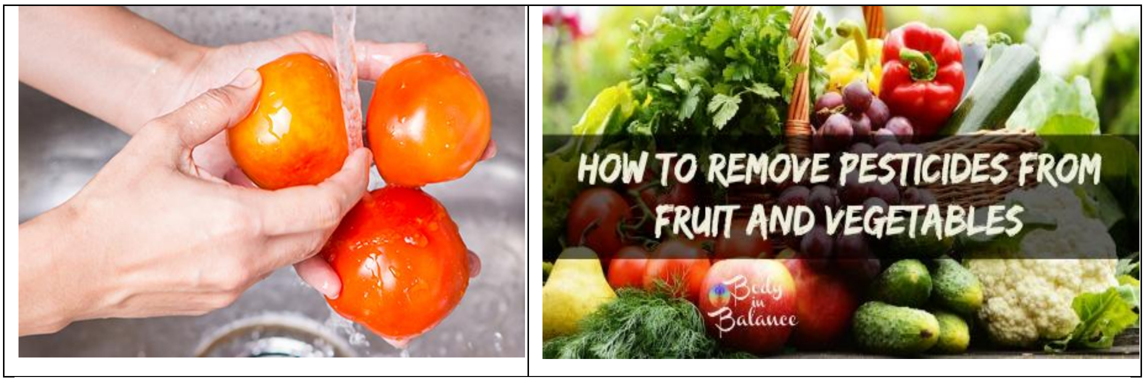

Figure 9. According to the Center for Science and Environment (CSE, USA), it helps to wash your fruits and vegetables with 2% of salt water. This should remove most of the contact pesticide residues that normally appear on the surface. CSE claims that if done diligently, washing with cold water should be able to remove 70% to 80% of all pesticides [http://www.healthychild.org/produce-purification-101-can-washing-fruits-veggies-remove-pesticides/].

Conclusions

Global population is now 7 billion and rising very fast. At the same time global food production must increase substantially to cover feeding needs of the rising population. FAO has already issued a sobering forecast on world food production which will need to rise by 70% in order to overcome rising energy prices, growing depletion of underground aquifers, the continuing loss of farmland to urbanization and desertification, and increased drought and flooding resulting from climate change. Advances in agricultural science and technology (fertilizers, pesticides, irrigation, green revolution, hybrid seed varieties, increased yields, etc) have contributed to remarkable increases in food production s by 2.5–3 times over the last 50 years and let food production keep pace with human population growth. Innovative and reformed agricultural food systems will also need to navigate complex resource limits imposed, in part, by environmental degradation to which modern agriculture has contributed.

The Integrated Pest Management (IPM) is a very important agricultural scheme that combines targeted use of agrochemicals (fertilizers and pesticides) with growing practice and biological techniques to control pests. This has been proved very effective in improving crop yields while reducing overall pesticide use. So, in this respect less toxic pesticides which are used in the last decades have a crucial role to play in agriculture. At the same time pesticides can protect non-urban populations from parasitic diseases. Most scientists agree that improvements in the control of weedy competitors of crops, crop diseases and pathogens, and herbivores could significantly increase crop yields, especially the three cereals (wheat, rice and corn), which provide 60% of world human food. Agrochemicals, such as (herbicides, antibiotics, etc) are protecting effectively agricultural food production. Evolving resistance within a decade there is a need to breed for new disease resistance and to discover new less toxic pesticides. This can be achieved by crop rotation and the use of spatial or temporal crop diversity.

From the 1960s FAO, WHO, most of developed countries and the European Union countries have set rules for the sustainable use of pesticides and regulations on the reduction of risks and impacts of pesticide use on people's health and the environment. Maximum Residue Limits (MRLs) for pesticide residues and residues of veterinary drugs are the maximum concentrations of residues to be legally permitted in or on a food and have been adopted on the basis of scientific expert advice. MRLs are adopted from most countries and regulate the safety of food products on a global scale. These levels of residues (on raw materials) are set 100 times lower than the toxicological effect measurements in experimental animals. Various shades of the Precautionary Principle have been integrated into the EU's regulatory system. Public opinion and understanding of risk is misrepresented. Natural constituents of food are "good", synthetic substances are "dangerous". Life-style attitudes (smoking, obesity, lack of exercise, excessive consumption of animal fat, salt, sugar, etc) are not perceived as unhealthy. The heart of the controversy is the debate over risk versus hazard.

Pesticides residues decrease substantially after washing, peeling and processing of food commodities. Food products are monitored for concentrations of pesticide residues in all developed countries continuously, and an international alert system informs effectively national food safety agencies. Statistical data showed that only 2-3% of analyses of residues exceeded the MLRs concentrations. In this respect, most of the studies presented in this review showed that fruit, vegetables, olive oil, cheese, meat etc, have very low concentrations of pesticide residues with minimum of risk to health. Health authorities in all developed countries have active campaigns for increased daily consumption of fruit and vegetables by children and the general population because of the substantial benefits to their health. This review presents important scientific studies and consumer food reports on the perceived adverse health effects associated with consumption of fruit and vegetables.

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