WORKSHOP REPORT

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Wastewater Reuse Applications and Contaminants of Emerging Concern

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ORGANISED BY:

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DARE-EU COST Action TD0803

NIREAS International Water Research Center

Wastewater Reuse Applications and Contaminants of Emerging Concern

Workshop Report – White Paper

On the 13 and 14 September 2012, approximately 80 scientists gathered in Limassol, Cyprus, to discuss the challenges of wastewater reuse practices with regard to contaminants of emerging concern, their transformation while in the environment, their potential uptake by plants and crops, the effects that these contaminants may induce in the environment, the evolution and release of antibiotic resistance, and technologies that are able to remove such contaminants from wastewater.

The workshop aimed to address the following themes:

- Which are the contaminants of emerging concern that are relevant to wastewater reuse applications? (e.g. antibiotics and other licit and illicit drugs, transformation products, disinfection byproducts)
- What are the new concerns related to reuse applications? (direct and/or indirect effects)
- What technologies can enhance the 'conventional' treatment by removing such contaminants?

The workshop was held jointly by the NORMAN Network, COST Action TD0803 (DARE) and the NIREAS-International Water Research Center.

NORMAN, a network of reference laboratories and research centres for monitoring emerging environmental pollutants, is an independent and competent platform in the field of emerging pollutants. NORMAN facilitates an exchange of information, debate and research collaboration at a global level with official recognition from institutional agencies of the EU. (www.norman-network.net)

The main objective of "DARE", EU COST Action TD0803, is to identify and characterise environmental hotspots for the emergence of antimicrobial resistance, the spread of antibiotics and antibiotic resistance patterns, aiming to develop measures to control the evolution of

antibiotic resistance. Great emphasis is given to Wastewater Treatment Plants, which constitute a major source for the release of (newly evolved) antibiotic resistances into the environment. (www.cost-dare.eu)

NIREAS-International Water Research Center is co-financed by the European Regional Development Fund and the Republic of Cyprus through the Cyprus Research Promotion Foundation (DESMI 2008). It was created with the objective of leveraging scientific and engineering expertise in order to tackle water-related problems. (http://www.nireas-iwrc.org).

The workshop consisted of the following three sessions, each of which concluded with a panel discussion:

Session 1: Applied Practices and Current Challenges

Session 2: WWTPs and Antibiotic Resistance

Session 3: Uptake of Contaminants, Treatment Technologies

Session 1: Applied Practices and Current Challenges

Dr. Symeon Christodoulou, Associate Professor and Head of the Department of Civil and Environmental Engineering, and member of the board of directors of Nireas-IWRC (University of Cyprus), welcomed the participants and made a brief introduction of the current aims and activities of the research centre.

Dr. Despo Fatta-Kassinos, Assistant Professor and Director of Nireas-IWRC (University of Cyprus) then introduced to the audience the current challenges in the field of wastewater treatment and reuse, and summarised the related open questions. She stressed that the aim of the workshop was to answer the following key questions:

- *Is there a reason to deal with contaminants of emerging concern? Which are the most relevant?*
- *Is wastewater reuse practice causing accumulation of these compounds or other negative impacts?*
- *Are there best treatment technologies?*
- *Should treatment plants upgrade?*
- *Are there environmental or human-health consequences from exposure to these compounds or different mixtures of compounds in the long term?*

Professor David Waite (University of New South Wales, Australia) made a presentation on the provision of water in arid environments and talked about the lessons learnt in Australia in water use and reuse. He provided information on: the current status of water supply and demand in Australia; the major reuse schemes and large desalination plants; and major recycling projects.

Professor Dror Avisar, (Tel Aviv University, Israel), talked about the occurrence and chemical fate of pharmaceutical residues in the Israeli aquatic environment. He presented the effects on nearby aquifers of the operation of fish ponds at coastal sites. He concluded that the phreatic coastal aquifer, which is also a major drinking water source, is affected by the intensive aquaculture industry – which is mostly located above it – because of the presence of residues of pharmaceuticals administered to the fish. Oxytetracycline (OTC), an antibiotic commonly used for veterinary therapy in aquaculture, mainly used as a growth promoter and to prevent infections, is usually considered as an immobile compound (strongly absorbed). But his study showed a relatively rapid mobility of this compound, which appears to travel through the pond floor and the

aquifer beds towards the local groundwater because of the specific and unique hydrological conditions (creation of preferential saturated flow pathways) which developed under fish ponds and which enable the OTC to leak and rapidly reach groundwater. He also presented various examples of pharmaceuticals forming transformation products while present in the environment.

Professor Juliane Hollender (EAWAG, Switzerland) discussed the options that were assessed as part of the national "Strategy Micropoll" study in Switzerland for upgrading of wastewater treatment plants via integration of an advanced treatment step to improve the surface water quality. She presented the methodology applied to select the priority compounds for which environmental quality standards would be set. Three different activities were identified that could potentially hinder the introduction of these substances in the environment, one being the upgrade of selected treatment plants. She presented two technologies (ozonation at Regensdorf and activated carbon in Lausanne) and the way their efficiency was evaluated. Chemical analysis and bioassays were applied. Unwanted by-products formed during the ozonation, such as bromates and nitrosamines, were also monitored. The cost and the energy demand of the upgrade and their impact on the overall cost of the treatment were among the parameters taken into account for the selection of the recommended treatment processes. She concluded that technical measures in Switzerland should be taken at: a) large WWTPs to reduce high loads (>80 000 inhabitants), b) WWTPs at surface waters with a high wastewater load (> 10%) to improve the ecological status (> 8 000 inhabitants, > 24 000 at lakes), and c) WWTPs at surface water that are used for drinking water abstraction (precautionary principle).

Dr. Kevin Thomas (NIVA, Norway) analysed the presence of illicit drugs in wastewaters. He presented the potential sources of this class of compounds and the methods applied for estimating drug use through sewage analysis. He demonstrated through his presentation that illicit drugs are part of the urban water cycle, and that there are significant loads released from all the cities examined. He also explained that, although various treatment plants are efficient in removing such compounds, others are not that efficient. This results in the presence of illicit drugs in surface water. Illicit drugs have also been detected in finished drinking water.

The last speaker of the 1st Session, ***Dr. Susan Richardson*** (U.S.A), discussed the potential effects of the wastewater inputs and source water impairment, and the implications for water reuse. Furthermore, she presented the various stresses on the water supplies, the emerging contaminants found in drinking water, and the exposure routes, with emphasis on disinfection by-products

formed during disinfection processes. She elaborated on the formation of such by-products from other pollutants such as pesticides, pharmaceuticals, oestrogens, etc. She highlighted the fact that the increased complexity of chemicals in total drinking water exposure, the formation of disinfection by-products, and the fact that several of these compounds are not removed completely at WWTPs or via other advanced treatment processes, may lead to 'surprises' as to what can be found in the treated wastewater that is destined to be reused. It is therefore recommended to look beyond the regulated chemicals.

The session closed with a **panel discussion** led by **Dr. Norbert Kreuzinger** (Vienna University of Technology, Austria) and **Dr. Luigi Rizzo** (University of Salerno, Italy). The major concerns expressed, and the most significant questions raised, are the following:

- Re-used water might be 'dangerous' to humans' health.
- The multiple treatments applied currently might be sufficient.
- Our current knowledge of the applied systems might not be complete. Especially in relation to the removal of emerging contaminants.
- Wastewater must be checked at the treatment plants efficiently.
- The overall impact of the treated sewage should be assessed.
- There is a need to check the sources of wastewater before treatment.
- The impact of sewage overflows should be evaluated.
- Wastewater treatment plants must include a certain toxicity assessment methodology in their daily routines.
- There is a need for effect-based tools, better analysis of these tools, more sensitive tools.
- It is important to investigate risks at the sub-lethal levels.
- Serious challenges exist in how to convert conventional technologies into more advanced ones.

Session 2: WWTPs and Antibiotic Resistance

This session was opened by ***Professor Thomas Berendonk*** (Technische Universitaet Dresden, Germany), who presented the European COST Action TD0803, entitled "Detecting evolutionary hotspots of antibiotic resistances in Europe"). Bacterial resistances to antibiotics are increasing and pose a serious threat to public health. Currently, it is largely unknown to what extent the environment serves as a reservoir for the evolution of new antibiotic resistances. The evolution of new resistances is a local process with global impacts on society and the environment and therefore the network is ideally suited to investigate these processes. Through cooperation across disciplines and between scientists from different countries, this Action is trying to establish the first comparative database on antibiotics and antibiotic resistances in the environment, wastewater treatment plants, animal production, and clinics. This Action tries to identify the key evolutionary and ecological processes that lead to the evolution of antibiotic resistances in the urban and natural environment. This will contribute to guidelines for European and regional measures to reduce the evolution of antibiotic resistances in the environment, and will allow a better prediction of the effectiveness of newly developed antibiotics.

Professor Celia Manaia, (Universidade Católica Portuguesa, Portugal), then talked about antibiotic resistance in urban wastewater treatment plants (UWTP), environmental contamination and the risks in water reuse. She introduced to the audience the problem of the development of antibiotic resistance, and presented detailed examples of how UWTPs can function as bioreactors for antibiotic resistance enrichment. She also talked about the consequent introduction of resistance and the genetic determinants in the environment. She presented the standing unanswered questions, such as the unknown resistome, and how important this is in the environment, the factors triggering gene transfer and what makes a resistance gene a success.

Dr. Eddie Cytryn (Volcani Agriculture Research Center, Israel) described the impact of treated wastewater irrigation on antibiotic resistance in soil microbiomes. He presented a study performed in Israel, which examined the impact on soil antibiotic resistance of treated wastewater used for irrigation. The study revealed that treated wastewater-associated antibiotic resistant bacteria and genes do not appear to enhance antibiotic resistance in the irrigated soils. The soil bacteria were characterised by significant levels of native antibiotic resistance. Nevertheless, he underlined the importance of developing risk assessment models to predict the horizontal transfer

of antibiotic resistance genes and to look at antibiotic resistance genes in mobile genetic elements.

A **panel discussion** was then held, led by **Dr. Marie-Noëlle Pons** (CNRS-ENSIC, France) and **Professor Hemda Garelick** (Middlesex University, UK). The discussion identified the following points and questions as the most important and relevant ones with respect to the evolution/release of antibiotic resistance bacteria/genes in the environment:

- The long-term responses in relation to antibiotic resistance in the environment should be identified.
- How the above question relates to concerns about the emergence of new diseases should be determined.
- The relevance of environmental bacteria when present in different media/habitats should be investigated.
- Different transfer pathways may exist for different antibiotic and bacterial species.
- Different effects may be caused in the organisms of the same species.
- Statistics on ecological analysis are required.
- Wastewater must be studied as a potential source for the transfer of various genes into the environment.

Session 3: Uptake of Contaminants, Treatment Technologies

Professor Ana Agüera, (Universidad de Almería, Spain) presented analytical methodologies for evaluation of advanced tertiary treatment processes applied for water reuse purposes. She discussed the objectives of this evaluation, which include the understanding of degradation mechanisms and the identification of transformation products, the combination of chemical and toxicological analysis for the interpretation of toxicity and biodegradability results, the detection of undesirable compounds, the comparison of the efficiency of different treatments, and the design and optimisation of the various processes applied for wastewater treatment. She underlined the fact that many studies include only one compound, high concentration and modelled sampling matrices, i.e. conditions different from the real environmental conditions. She presented results from various relevant studies, including studies of the analysis of organic compounds in soils and crops irrigated by treated wastewater. She highlighted the fact that field studies (real conditions) are required in order to derive concrete results and recommendations.

Dr. Josep Bayona (IDAEA-CSIC, Spain), talked about the uptake of organic micro-contaminants by vegetables from irrigation waters, and presented results obtained from *in vitro* and field studies. *In vitro* studies showed that plants can uptake micro-contaminants from the growing medium by roots and leaves when they are exposed to contaminated water. Uptake kinetics by roots depends on the physico-chemical properties of the contaminant, and concentrations in leaves decline following a time lag attributable to their metabolisation. Field studies were conducted in greenhouse conditions where secondary and tertiary reclaimed water was used for irrigation of different crops (e.g. lettuce, green beans and carrots). Crops irrigated with secondary treated effluents exhibited a higher frequency of detection and higher concentration of micro-contaminants than well water. Finally, crops (e.g. alfalfa and apple tree) from the NE of Spain irrigated with reclaimed river waters showed the lowest concentrations, probably because of the removal processes occurring in soil.

Professor Dionysios (Dion) D. Dionysiou, (University of Cincinnati, U.S.A), talked about the use of oxidation processes to destroy cyanotoxins, taste and odour compounds, and selected pharmaceuticals in water treatment and reuse applications. He presented a number of examples of research studies of his group, providing examples on the application of Advanced Oxidation Processes (AOPs) for the abatement of pesticides, PPCPs, antibiotics, cyanotoxins, etc., and the transformation products formed. He emphasised the need to develop new AOPs, or improve

existing AOPs, develop new catalysts, etc. He underlined the fact that new advances in the field of nanotechnology and reaction engineering show promising results and encouragement for the removal of contaminants of emerging concern, but detailed mechanistic aspects are yet to be understood. Moreover, the effective coupling of processes can yield targeted removal efficiencies of such compounds but optimisation schemes are necessary for specific water sources. Finally he closed his presentation by highlighting the fact that all the relevant ongoing efforts for the development of new AOPs or improvements of existing AOPs face many scientific challenges.

Professor Gianluca Li Puma (Loughborough University, U.K.), made a presentation on the advanced oxidation of oestrogens, antibiotics and other emerging contaminants in water, and the potential for solar detoxification and water reuse. He concluded that solar-powered advanced oxidation can be considered as an effective process for the treatment of emerging contaminants in WWTP effluents. Large diameter Compound Parabolic Collector reactors treat such contaminants efficiently using low catalyst concentrations. The optimisation of solar reactors for emerging contaminants treatment relates to the use of the lowest catalyst concentration, the size of the reactor, the total pollutant load, and issues related to the cost of the catalyst recycling. He emphasised the fact that more engineering pilot and large-scale demonstration studies should be carried out at different scales, using home/domestic, semi-industrial, industrial and municipal wastewater. He also discussed the issue of photocatalytic reforming of biomass waste, which may provide an efficient and low cost method for production of renewable hydrogen from waste biomass.

Professor Luigi Rizzo (University of Salerno, Italy), discussed the disinfection of urban wastewater by solar-driven and UV lamp – TiO_2 photocatalysis and the effect on a multi-drug resistant *E. Coli* strain. He therefore introduced to the audience the need for checking the removal of antibiotic resistance during the treatment process and the capacity of photocatalysis to achieve this objective. The role of urban wastewater treatment plants (UWWTPs) in the spreading of antibiotic resistance into the environment has not yet been fully investigated, and poor information on the role of disinfection processes is available. In this study, the effect of an advanced oxidation process, namely TiO_2 photocatalysis, was investigated for the inactivation of an antibiotic resistant *Escherichia coli* strain selected from an UWWTP effluent.

A **panel discussion** was then held, led by **Dr. Christa McArdell** (EAWAG, Switzerland) and **Professor Dionysios Dionysiou** (University of Cincinnati, U.S.A.). The major concerns

expressed and the most significant questions raised are the following:

- Prior to the decision on which technology should be applied, the source of water should be considered (e.g. ozonation – formation of bromates).
- Chemical compounds that could act as indicators for the performance of the treatment process are needed.
- The available technologies might be inadequate in removing all classes of micropollutants? (Example: ozonation is not efficient in removing X-ray contrast media).
- Technologies could be ranked according to the drawbacks and advantages.
- There is a need for clear conclusions / recommendations on the technologies to be applied: investors need to be assured that their investment is viable and sustainable for the next 25–50 years.
- Toxicity assessment needs to be standardized.
- Validated modelling tools are needed to evaluate and compare the performance of the different technologies.

Afterwards, nine short talks were delivered on various important issues as follows:

10-Minute Presentations

Predicting the fate of emerging contaminants in sewage treatment plants: evolution of the SimpleTreat model, **Dr. Antonio Franco**, Unilever, U.K.

Monitoring of water quality using photometric technique. Case study: Application of Hungarian developed Robotic Water Analyser, **Dr Ákos Rédey**, University of Pannonia, Hungary

Organics removal and minimisation of degradation products during treatment of pharmaceutical wastewater, **Dr Giuseppe Mascolo**, Water Research Institute, Bari, Italy

Required ozone doses for achieving 90% removal of pharmaceuticals from Swedish WWWTP effluents, **Dr. Maria G. Antoniou**, Cyprus University of Technology

Ecotoxicity assessment of wastewater containing mixtures of active pharmaceutical ingredients, **Dr. Marlen Ines Vasquez**, University of Cyprus, Cyprus

Environmental impact assessment of antibiotics mixtures: a risk evaluation in urban wastewater treatment plant effluents, **Dr. Süreyya Meriç**, Namık Kemal University, Turkey

Prevalence of Tetracycline Resistance in the wastewater treatment plant, **Dr Magdalena Popowska**, University of Warsaw, Poland

Persistence and dissemination of the multiple-antibiotic-resistance plasmid pB10 in complex environment matrices, **Dr Christophe Merlin**, Université de Lorraine, France

Comparison of photocatalytic degradation for series of dyes, **Dr Marie- Noëlle Pons**, Université de Lorraine, France

FINAL PANEL DISCUSSION

Led by Dr. Jaroslav Slobodnik (Environmental Institute, Slovak Republic)

The discussion can be summarised as follows:

- Long-term effects of micropollutants, including those at the sub-lethal level need to be assessed and understood.
- Information on the effects of the inadvertent chronic exposure of organisms to micropollutants is scarce and incomplete.
- The tests and methods for assessing the adverse effects of micropollutants in wastewater are not standardised (validated protocols are required).
- The uptake of micropollutants by plants and crops during wastewater reuse applications has been proved in many cases. More systematic studies under real environmental conditions are required.
- Reuse applications should integrate phytoxicity/ecotoxicity tests.
- Effluent quality criteria are required in order to assess the various technologies in a uniform way.
- More studies on pilot and industrial scale are needed.
- The cost of the various technologies is not always known.
- Expert systems and algorithms/decision support tools may have a major role to play in selecting best technologies.
- Life cycle analysis could be applied for the assessment of wastewater treatment technologies.
- Treatment technologies need to be evaluated, using the same experimental conditions, in respect of their capacity to remove micropollutants, antibiotic resistance, toxicity, etc, produce unwanted by-products and remove/induce toxicity.
- Natural organic matter present in wastewater needs to be thoroughly examined and assessed in respect of its capacity to generate toxic by-products during the application of advanced chemical processes.
- The information on the capacity of the currently applied wastewater treatment systems in removing antibiotic resistance is still scarce.

- Antibiotic resistance genes have to be considered as contaminants of emerging concern
- Concerning antibiotic resistance, model sites in various countries should be examined.
- The tests and methods for assessing antibiotic resistance removal are not standardised (protocols are required).
- The effects of newly released drugs and thus new chemical mixtures on the resistome are a challenge.

What is the next step?

Scientific meetings such as this one on this particular topic should be organised on a regular basis under a specific framework.

A dedicated Working Group within the NORMAN network and activities should be established.

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