Executive summary:

The contamination of drinking water infrastructures (catchment areas, raw water transfer systems, treatment facilities, treated water reservoirs and distribution networks) with CBRN (chemical, biological, radionuclides) as a result of malevolent acts of sabotage represents one of the major threats that security has to face with. Rapidly restoring the functionality of such infrastructures, and the access to safe drinking water represents another major concern for regulatory agencies and water utilities. Indeed, the damage resulting from impairment of drinking water services would seriously impact the quality of life of many people not only by directly harming them but also making water systems unusable for a long period of time with a risk of societal disorder (similar situation as with any accidental contamination events or natural disasters).

Such accidental or malevolent contamination events determine a crisis situation, which affects or is likely to affect a water utility or its provided services, and require more than the usual means of operation and / or organisational structures to deal with it. The ISO 11830 standard on crisis management describes the fundamentals of a crisis management system (ISO, 2011). ISO 11830 provides general guidance on how a crisis should be dealt with ("crisis phase"), on how to re-establish services (post-crisis phase), and on the best way to draw conclusions and revise procedures for future events. A large number of "preventive" actions have to be taken / implemented on a routine basis before the crisis, and recovery activities can start during the crisis phase. At the same time, specific scenarios for supplying potable waters (distribution of bottled waters, water tankers, emergency water treatment mobile systems, and so on) also have to be developed.

In this context, SECUREAU (a European project for restoring distribution system functionality after deliberate CBRN attacks involving 12 partners from 6 countries) has defined four research and development objectives:

a. tools for detecting water quality changes by combining generic non-specific and specific sensors for measuring unexpected / abnormal signal variations (and to be integrated in an Early Warning System); methods for identifying the best locations for sensors for full coverage monitoring; methods for treating the data collected every five minutes by the network of sensor
b. methods for rapidly identifying the source(s) of any intentional contamination thanks to accurate mathematical procedures and new software
c. multi-step strategies for cleaning distribution systems: pipe walls / biofilms / deposits and waste (water bulk and deposits extracted from the network).
d. analytical methods for confirming cleaning procedure efficiency taking into account especially the pipe walls.

The main results of SECUREAU are organised under ten items:
1. Sensors and sentinel coupons (non-specific sensors; specific sensor; biofilm sensor; sentinel coupons)
2. Optimal location of water quality sensors and data treatment
3. Optimal distribution of sentinel coupons
4. Identification of the sources of contamination and the contaminated areas
5. CBRN analytical methods
6. Modeling sorption and desorption

7. Pipe wall cleaning and decontamination
8. Handling of decontamination sludge and water
9. Validation of decontamination
10. Decision tool

The main socio economic impacts of the SECUREAU project are described in four main items:
(i) the security and safety synergy reinforcement for the benefit of EU citizens;
(ii) the development of EU industry and leadership on environmental sensors networks;
(iii) the development of modern analytical tools for contamination surface analysis and
(iv) the decontamination strategies combining complementary technics.

Project Context and Objectives:

The importance of water and water infrastructures to human and ecosystem health and to the functioning of our economy makes water systems targets for terrorism. Their contamination with CBRN agents as a result of malevolent acts of sabotage represents one of the major challenges that security has to face with. The damage is done not only by hurting people but also rendering water systems unusable for a long time with a risk of societal disorder.

In order to restore rapidly safe drinking water distribution, SECUREAU (an European project for restoring distribution systems after deliberate CBRN attack involving 14 partners from 6 countries) has defined three levels of research & development:
(i) developing early warning system (EWS) for drinking water distribution systems. It means an integrated system for deploying a monitoring technology (specific and non-specific sensors for detecting unexpected/abnormal signal variations), mathematical models for positioning these sensors, analysing and interpreting the huge mass of results given by the sensors to make decision while minimizing unnecessary concerns and inconvenience within a community.
(ii) allowing a rapid localization of the source(s) of intentional contamination thanks to accurate methods and softwares.
(iii) defining multistep strategies for cleaning the distribution system: pipe wall/biofilms /deposits. Curative treatments will concern pipes, and wastes (water bulk and deposits), as well as methods for controlling the efficacy of the decontamination procedure.

SECUREAU serves as a research project for designing and implementing an effective and timely response action after drinking water distribution system contamination which may be integrated in the management of a crisis phase (ISO 2011). The restoration of the network will have to provide maximum and rapid benefits to the users with limited environmental effects. The SECUREAU programme was organised in 8 workpackages:

1: Project management
2: Early warning systems for rapid detection of deliberated intrusion
3: Off-line rapid detection methods in water and biofilms/deposits
4: Modelling contaminant accumulation
5: Decontamination procedures of water distribution systems
6: Controlling the efficacy of decontamination of deposits
7: Case studies and data support analysis
8: Dissemination, exploitation and, transfer

As a preliminary objective, we aimed to get a "state of the art" related to early warning systems, sensors, etc.. Five main questions were clearly identified:
- What are the key characteristics (accuracy, robustness, price ...) of commercially available sensors for early warning systems (EWS)?
- How to organise pilot bench scale to control biofilm disinfection and sensors testing?
- What methods can be used or in development for cleaning contaminated networks?
- How to improve sensitivity of analytical methods for controlling rapidly water bulk (several known CBRN or surrogate)?
- How to design sensor location to monitor contaminant distribution and/or the origin of injection in the distribution systems?

During the year 2 of the project SECUREAU, five objectives were addressed:
- Definition of the key characteristics (accuracy, robustness, price...) of commercially available sensors for early warning systems (EWS) and improvement of such devices to prepare their implementation in networks and pilots during year 3 and year 4.
- Finalisation of pilot bench scale to control biofilm disinfection and sensors testing.
- Sensitivity improvement of off-line analytical methods used for detection of CBRN threat agents in water and following adsorption to biofilms and other deposits occurring in water distribution networks
- Continue the development of methods for cleaning contaminated networks (both chemical and physical methods)
- Design sensor location to monitor contaminant distribution and/or the origin of injection in the distribution systems.

Six main objectives were carried out in the third year of SECUREAU:
- Analytical methods were adapted from literature and validated for the detection of CBRN in the matrices of interest, having special attention to matrix effects. Three groups of chemicals or microorganisms or radionuclides were concerned:
  paraquat, chlorfenvinphos, carbofuran, pentabromodyphenylether (BDE-100) and methylmercury in deposits and/or biofilms.
  alpha, beta and gamma radionuclide emitters both in drinking water and in biofilms and deposits
  various bacterial pathogens (Y. pestis, E. coli, Bacillus), including surrogate species (F. tularensis, Y. pseudotuberculosis, B. subtilis) in drinking water.
- Commercial on-line sensors (MSS, Optiquad and Neosens) and coupon monitoring devices were tested in three loop systems.
- Three new sensor prototypes measuring (i) pH, temperature, chlorine, conductivity, (ii) organic matter and turbidity, and (iii) radionuclides, were developed for early warning systems (EWS); Their implementation in two large drinking water networks (one rural, one urban) was planned as well than testing their behaviour in different pilots.
- Optimal placement of sensors was also a main objective of this work period. It was planned according to several optimisation criteria for two selected sites in Europe (one rural site and one urban site). Mathematical models were developed to define both the contamination source origin, and the contaminated area.
- Different decontamination procedures were planned to be tested and evaluated. As an example, the applicability of cavitation method to remove bacterial spores from contaminated water and pipes surfaces, by taking into account both engineering and economical aspects was planned. The development of chemical methods (dissolution, chlorination, dispersion, complexation and Fenton-like) was also taken in account. In particular, very promising results were reached with Fenton-like oxidation, which proved to be effective for degradation of SECUREAU chemical contaminants in water. Pipe deposits also revealed to act as catalysts in this process, which is already being tested at pilot scale.
- Implementation of methods and/or strategy for the treatment of contaminated water and deposits is also taken into account.

The efforts spent during year 4 have permitted to continue and to improve and to conclude the work initiated during the previous periods, and also to communicate via per review publications; technical meetings; European conference. Seven main objectives were clearly identified for this year 4:

- Final experiments (focused on wall deposits and biofilms) were carried out to determine the kinetic parameters for adsorption and desorption of chemicals, radionuclides and microorganisms in biofilms and inorganic deposits. Values were obtained for sorption of: (a) paraquat to clay and iron oxide materials; (b) mercury to iron-based and calcium-rich deposits; (c) Po-210 and Am-241 to biofilms and to deposits scraped from cast iron pipes; (d) Francisella philomiragia, Yersinia pseudotuberculosis, Francisella tularensis and Bacillus subtilis to drinking water biofilms.
- The modelling part was divided in two parts. One was the software development to simulate contaminant transport along drinking water networks. The other was the development of software tools to identify the source of contamination (together with the optimization of sensor positioning, see WP2).
- Decontamination methods of water systems and handling of obtained sludges was one of the year-4 objectives. In the reporting period experiments were carried out both in laboratory scale and in three pilot loop systems. Results aimed to select the methods, which are closed to practical application, and to evaluate their applicability in real situation (economical, technical or safety reasons).
- Selected decontamination methods were tested with the aim to clean/decontaminate three pipe reactors, each of them being experimentally contaminated with either a very soluble organic pesticide (glyphosate), or spores of Bacillus subtilis, or not contaminated (then the target was the autochthonous biofilm).
- In addition controlling the efficacy of decontamination procedures was planned according three steps (i) to develop a concept for positioning coupons and sensors at representative and indicative locations in the distribution system, (ii) to optimize and to verify the decontamination efficacy by use of the sensors/coupons and (iii) to use the information as generated by the earlier tasks of WP6 in order to provide a decision tool.
- The installation of two networks (one rural and one urban) of 40 sensors each was planned to get field information and to generate a know-how to install an early warning system in real cases.
- In terms of communication, the main objective for the months 37 to 48 was to ensure a clear and effective dissemination, exploitation and transfer of the results to (i) partners inside the group, (ii) the international scientific community, (iii) the industry and (iv) the general public (in particular thanks to a European Conference).

All over the project, recommendations provided by external reviewers (Walter Biederbick and Clive Thompson) following two scientific reviews (Feb 2011 and Feb 2012) and have been taken into account.

Project Results:

Introduction: context and actions.

The contamination of drinking water infrastructures (catchment areas, raw water transfer systems, treatment facilities, treated water reservoirs and distribution networks) with CBRN (chemical, biological, radionuclides) as a result of malevolent acts of sabotage represents one of the major threats that security has to face with. Rapidly restoring the functionality of such infrastructures, and the access to safe drinking water represents another major concern for regulatory agencies and water utilities. Indeed, the damage resulting from impairment of drinking water services would seriously impact the quality of life of many people not only by directly harming them but also making water systems unusable for a long period of time with a risk of societal disorder (similar situation as with any accidental contamination events or natural disasters).

Such accidental or malevolent contamination events determine a crisis situation, which affects or is likely to affect a water utility or its provided services, and require more than the usual means of operation and / or organisational structures to deal with it. The ISO 11830 standard on crisis management describes the fundamentals of a crisis management system (ISO, 2011). ISO 11830 provides general guidance on how a crisis should be dealt with ("crisis phase"), on how to re-establish services (post-crisis phase), and on the best way to draw conclusions and revise procedures for future events.A large number of "preventive" actions have to be taken / implemented on a routine basis before the crisis, and recovery activities can start during the crisis phase. At the same time, specific scenarios for supplying potable waters (distribution of bottled waters, water tankers, emergency water treatment mobile systems, and so on) also have to be developed (Loo et al. 2012).

In this context, SECUREAU (a European project for restoring distribution system functionality after deliberate CBRN attacks involving 12 partners from 6 countries) has defined four research and development objectives:
a. tools for detecting water quality changes by combining generic non-specific and specific sensors for measuring unexpected / abnormal signal variations (and to be integrated in an early warning system); methods for identifying the best locations for sensors for full coverage monitoring; methods for treating the data collected;
b. methods for rapidly identifying the source(s) of intentional contamination thanks to accurate procedures and software;
c. multi-step strategies for cleaning distribution systems: pipe walls / biofilms / deposits and waste (water bulk and deposits extracted from the network).
d. analytical methods for confirming cleaning procedure efficiency.

The Deliverable 8.4 "Methodological guide for end users" (public level) aims to highlight, along with Deliverable 6.8 (Decision tool Suitable for Assessment to Approval of Successful decontamination) (confidential level), some of the methods selected in SECUREAU for each phase defined above, i.e. pre-crisis, crisis and post-crisis.

During the pre-crisis phase, besides daily routine tasks and usual network management procedures, specific devices (e.g., reliable and robust sensors) are dedicated to monitor water quality. Any abnormal change in water quality (unexpected changes, taking into account historical data) should raise an alarm and determine the course of actions to be undertaken. Traditionally, an "abnormal situation" corresponds to a change three times greater than the standard deviation of the baseline, for the parameters measured (as usually done in analytical chemistry).

Insofar as specific sensors of any potential contaminants could not be developed, the aim of SECUREAU was to develop sensors taking into consideration traditional parameters for monitoring water quality, as well as some specific parameters such as radionuclides. Sensor positioning was optimised by modelling, taking account of both the hydraulics of the network and specific assumptions made by operators (protection of the population at risk, economic situation to minimise the number of sensors, and so on). Besides, installation of dormant sentinel coupons was considered. These coupons would be installed in the water supply system and get colonised by biofilms and deposits just like pipe walls. They would then be used to validate the cleaning procedures applied throughout the network during the crisis phase but also during "normal" operation of the network (pre-crisis).

As a result the research under the SECUREAU project was conducted in four directions, being relevant to the pre-crisis phase. De facto, we developed an Early Warning System (EWS), that is to say an integrated system for deploying the monitoring technology, analysing and interpreting the results, and utilizing the results to make decision ... while minimizing unnecessary concerns and inconvenience within a community:
- sensors, both specific and non specific. Respectively, they measure the radionuclide content in water and, thanks to the determination of traditional parameters (conductivity, chlorine, pressure, etc.), they may indicate any abrupt change in water quality; other sensors (OptiQuad, FS-900 and SkidSens) are able to monitor the deposits on the surface of the pipes.
- sentinel coupons. Sentinel coupons of polymeric materials (HDPE, EDPME, etc.) are to be installed in water distribution systems for deposits and biofilms to form on their inner surface. The extraction and analysis of these coupons, carried out without the water flow being interrupted, allow the concentration of contaminants associated with deposits to be estimated. At a later stage during the crisis phase, such coupons allow contaminant accumulation and cleaning procedure efficiency to be measured.
- optimal positioning of the sensors and coupons thanks to new models taking into account economic, strategic and technical assumptions made by the operators.
- methods for handling data provided by the sensor network (one data each five minutes for each sensor) and their treatment.

During the crisis, actions are taken in order to identify the contaminant, the contaminated area and the source(s) of contamination. Obviously, decontamination strategies are needed for both the water bulk and the pipe walls, which represent a challenging objective. In order to bring to an end to the crisis phase, analytical methods - again adapted to pipe wall analysis – should be carried out. During the crisis phase, some tools developed in SECUREAU should be used:
- water quality sensors to continue monitoring water quality and to measure the extent of contamination (if the water flow has not been interrupted),

- sentinel coupons to monitor the concentration of the pollutant adsorbed onto the pipe walls,
- mathematical models to determine, thanks to measurements carried out throughout the network, the areas which have been contaminated and the sources of contamination,
- various cleaning methods, both traditional and new ones, are to be applied to decontaminate the network, especially pipe walls.

The Post-Crisis Phase is the complete changeover from crisis management to normal management including normal supply of the water utility services. Part of this phase is carried out under crisis management and part under normal management (ISO 2011). No specific tools were developed for such a phase as it is a matter of management and restarting routine actions.

The main results of SECUREAU are presented below in 10 independent sections:
1. Sensors and sentinel coupons (non-specific sensors; specific sensor; biofilm sensor; sentinel coupons)
2. Optimal location of water quality sensors and data treatment
3. Optimal distribution of sentinel coupons
4. Identification of the sources of contamination and the contaminated areas
5. CBRN analysis
6. Modeling sorption and desorption
7. Pipe wall cleaning and decontamination
8. Handling of decontamination sludge and water
9. Validation of decontamination
10. Decision tool

1 Sensors and sentinel coupons

Fourteen commercial non-specific sensors have been evaluated regarding 12 technical criteria. Three of them were selected and are devoted to water analysis (free chlorine, pressure, temperature, conductivity, organic matter, turbidity and radionuclides). Five other devices are used for assessing the accumulation of deposits on the surface of the pipes. Sensors devoted to water analysis have two purposes. The first is to rapidly detect any abnormal changes in water quality, and the second is to provide assistance to the operator to manage the drinking water network under normal operational conditions.

Sentinel coupons have three main objectives: (i) characterise deposits on the coupon, (ii) identify the adsorbed contaminants and (iii) validate the cleaning and / or decontamination procedures.
These devices (sensors and sentinel coupons) should be obviously installed during the pre-crisis phase in drinking water distribution networks. Nevertheless, the drawback is that it creates more access points to the distribution system, and hence could weaken it (i.e. more points that can be tampered with).

The development and design of Kapta 3000 AC4, Kapta 3000 OT3 and Kapta 3000 RAD1 were carried out during the SECUREAU project with technical specifications meant for allowing rapid installation in drinking water networks. The sensors were chosen according to some criteria as life-time and maintenance, energetic independence, wireless communication, no water loss, sanitary conformity, easy installation for all canalisation diameters.

The development of MSS (Mechatronic Surface Sensor) was mainly carried out during the SAFER project (FP5 project EVK1-CT-2002-00108). Its design and adaptation to drinking water networks were carried out during the SECUREAU project.

The other three sensors (OptiQuad, FS-900 and SkidSens) are prototypes or commercial products elaborated by private companies outside of the framework of the SECUREAU project.

1.1. Non-specific sensors (Kapta 3000 AC4 and Kapta 3000 OT3)

The Kapta 3000 AC4 sensor measures free active chlorine, pressure, temperature and conductivity (Deliverable 2.1.2, version 2, June 2012). In the context of SECUREAU, two European drinking water systems were equipped with 80 sensors each which measure water quality online and send results every two hours to operational control centres thanks to a GSM communication module.

The Kapta 3000 OT3 sensor, thanks to two LEDs, measures transmission at two wavelengths: 254 nm for organic matter and 625 nm for turbidity measurement. Two optic paths are used for each wavelength in order to compensate for fouling.

The Kapta 3000 OT3 probe is currently under development and all its specifications are being evaluated for a commercial product expected available by 2014.

Both sensors are designed to be easily and quickly installed in drinking water distribution networks.

1.2 Specific sensor (Kapta 3000 RAD1)

A new type of online sensors for measurement of traces of gamma emitters in solution has been proposed in SECUREAU.

The detection limits (Bq/kg, measurement for 10 seconds) with the NaI(Tl) detector are presented. Using an integration time of 10 s, the sensor allows an alarm to be raised before the maximum permitted levels of radioactive 60Co and 137Cs are reached. On the other hand, the limit of detection is too high for 241Am. In order not to exceed the maximum value of 10 Bq L-1, the integration time should be around 90 min (241Am is essentially an alpha emitter and the system is not well adapted). This sensor is designed to be easily and quickly installed in drinking water distribution networks.

1.3 Biofilm sensors (MSS, Optiquad and Neosens FS-900 / SkidSens)

Allowing deposits to be monitored online is one of the key aspect as contaminants, depending on their nature, may adsorb onto pipe walls and deposits. Biofilm/deposit sensors appear very useful for evaluating the effectiveness of a cleaning procedure throughout the network, meaning they have to be installed during the pre-crisis period. Three commercially available sensors - MSS, FS-900 and SkidSens – and one prototype - Optiquad - were tested over a relatively short time period of one month to monitor the deposits formed on the pipe surfaces in a drinking water distribution network. However, their interest is quite limited in a drinking water distribution system for three main reasons:

- First, their high cost does not allow them to be used in large numbers in a network,
- Secondly, the low sensitivity of FS-900 and SkidSens does not allow them to be used in drinking water systems.
- Third, the OptiQuad sensor can only detect the first deposition events. The latter act as a screen and inhibit the spread of the signal, thus preventing the sensor from distinguishing between thin and thick deposits.

The four systems need a power supply (Optiquad: 230 V, FS-900 and SkidSens: 24 V or 230 V) and calibration step (to convert the signal to cell counts or deposit thickness, for example).

1.4 Sentinel coupons

Sentinel coupons should be installed during the pre-crisis phase in drinking water distribution networks so as to allow, during and after the crisis period, (i) the deposits on the coupons to be analysed, (ii) the adsorbed contaminants to be identified and (iii) the efficiency of the decontamination procedures to be assessed. The materials used for the coupons should not be corrodible as they will be in the pipes for years.

Our experiments show that coupon-monitoring devices are suited to follow deposit/biofilm formation in drinking water distribution systems as well as to investigate and confirm the successful removal of deposits from surfaces. Such devices have to be installed proactively on bursts / leaks (in specific chambers) during pipe repair or installed in each district meter area in meter / pressure reducing valve chambers. They can also be fitted in a chamber equipped with other sensors.

2 Optimal location of water quality sensors and data treatment

The installation of water quality sensors in a drinking water system allows an alert to be issued rapidly when abrupt changes in the quality of water are detected. It is necessary to ideally distribute these sensors taking into account a number of considerations. Indeed, water supply providers may consider expenses (limited budgets), efficiency (willingness to detect a contamination event within less than x hours), protection of specific groups of consumers (e.g., children, hospital patients or retirement home residents), etc. These considerations can of course be combined, always with the same purpose in mind: to alert as quickly as possible (early warning concept). Objectives and constraints need to be defined to ensure that sensors are distributed in the best possible way: some are sensor specific (technical considerations for installation), others are to be considered when it comes to cleaning, while others are peculiar to population vulnerability and financial costs.

For the early-warning design (optimal location of sensor), two different software should be run. The first one generates the contaminant events by a Monte Carlo method. It consists of an executable code linked with the Epanet DLL. It is necessary to have an INP Epanet file describing the network and the hydraulics conditions (demand, tank level, valve status, pumps...). The second step is to solve a multi-objective INLP problem, with the contaminant generated from the first step. The two complementary softwares solutions are free of use but are not documented except information given in the deliverable 2.2.2. So using it is not an easy task. There is no access right to the source code.

Two European drinking water systems (Urban and Rural) have been equipped by February 2012 with sensors which measure water quality online and send results every two hours to operational control centres (Network U was equipped with 44 Kapta 3000 AC4, and 15 Kapta 3000 OT3. Network R was equipped with 29 Kapta 3000 AC4, and 8 Kapta 3000 OT3). Firstly the installation of the 2 sensor networks gave some valuable information and generated an indispensable know-how to install an early warning system in real case. That shows the feasibility of the installation with a limited budget. It also allowed finding the best compromise during the installation between prices, feasibility, remote data efficiency optimization and interest of points for the targeted application.
The installation of a probe network is just one part required to create an early warning system against the terrorist attack on a drinking water distribution network. Once the equipment is installed and the data are correctly transferred, a significant work has to be done to transform all this data (up to 126,000 data points per day, as an example!) into valuable information. To be efficient, several steps have to be considered. The first one is to better understand how the water quality normally changes in the network. Indeed, before the probe network was installed we did not have such detailed information. The second step is to optimize the operation of the water quality management in the network in order to improve water quality and stabilize the water quality behaviour. Finally, the survey can be implemented with the help of know-how of the operators and with algorithms developed in the same time.

It is possible to evaluate the impact of operational practices on the water quality thanks to the sensors network. For example, a change of water source in one of the two networks caused a decrease of chlorine level for 2 months. Moreover, the survey of the network, through the data handling methods developed, shows that several changes or events in water quality were detected during the year of functioning. For example, some incidents at the drinking water treatment plant have consequences on chlorine level (sudden drop and raise of the concentration) and are visible on non-contiguous areas of the drinking water network.

3. Optimal distribution of sentinel coupons

Sentinel coupons are simple tools which can be used to control the efficiency of decontamination and to identify the composition of deposits throughout the network under normal operational conditions. The coupons must be positioned optimally throughout the network so as to be representative of water quality interaction with pipe surfaces while minimizing costs. Consequently, the mathematical models dedicated to the spatial optimisation of sentinel coupons differ from those dedicated to the spatial optimisation of generic water quality sensors, because the objectives and constraints are not the same.

A new mathematical problem, namely the contaminant accumulation coverage model, is formulated for placing surface sensors and coupons to control the efficiency of decontamination. The design relies on the principle that good quality at a water sample entails a good quality at immediate upstream nodes. Moreover, the places where contaminant concentration is higher and the shear stress is lower are ideal candidates. This approach is very simple to implement and has, as a one and only prerequisite, the existence of a network model for hydraulic solving. This problem belongs to the class of maximum coverage problem. Hydraulic modeling is used to calculate the nodal wall shear stress and the coverage matrix.

Additionally, a Monte-Carlo simulation is run to generate contaminant events and calculate the corresponding concentrations. It was found than large network are tractable with only few minutes on the Neos server by cloud computing.

For the decontamination surveillance design (optimal placement of coupons) this is a two-step process. Firstly, the shear stress is worked out at any place, in parallel the contaminant events are generated and the objective and constraint are written in an output text file ready to use with GLPK. Then, contaminant accumulation coverage model formulation, an ILP programming problem, is solved with the GLPK (GNU Linear Programming Kit) package on a personal computer (PC). The prerequisites are an INP Epanet file describing the network, the hydraulics conditions (demand, tank level, valve status, pumps...). The first software is free of use. GLPK is a freeware solver. There is no documentation file other than the deliverable 6.5. There is no access right to the source code.

4. Identification of the sources of contamination and the contaminated areas

If a contamination event (accidental or deliberate) occurs in a drinking water network, it is essential to identify the sources of contamination and to determine the area which is likely to be contaminated, in order to isolate and decontaminate the affected area only, as well as keep supplying drinking water in non-affected areas, depending of the nature of the contaminant.

Different mathematical approaches combined with hydraulic models were applied within the SECUREAU project to determine, as quickly as possible, the possible contamination sources and the respective probable contaminated areas. Such determination is made possible thanks to information provided by the water quality sensors installed during normal management of the networks. The precision and rapidity of the determination are directly correlated with the number of sensors installed.

4.1. Localization of the sources

Different strategies were implemented during this project to identify the location of the contamination sources. The approaches considered: a) a deterministic method based on successive positive readings of sensors; b) methods based on artificial neural networks (ANNs) for single and multiple contamination events; c) stochastic methods such as least-squares solving with Tikhonov regularisation or minimum relative entropy solution (MRE); and d) a method based on the analysis of flow data.

- The deterministic method based on successive positive readings of sensors is based only on the analysis of the residence time of water in pipes and it only requires a binary sensor status over time. The results for the localization of contamination sources are given sequentially, being updated each time a new sensor detect a change in contaminant concentration. Furthermore, in some situations, this method enables the verification of the occurrence false negatives and false positives. It was observed that the occurrence of false positives or negatives did not affect the results related with the real detections and it was possible to detect the sensor which suffered this anomaly.
- The method that used ANNs to identify the contamination sources in single contamination events is able to identify the correct contamination

source and to predict the correct time of contamination associated to each possible contamination source, even in the case of large and highly complex DWDSs, following two approaches. The tests performed with contamination scenarios considering water demand uncertainty demonstrated that the method has shown good performance even in situations that are not described by the hydraulic model used in the development of the ANNs. Both approaches required a very low time of computation to obtain the results for real DWDSs, generally less than 5 seconds in a 3.10 GHz processor, which is a great improvement to the solution of this problem that demands the computation of the results as quick as possible.

- The method that extended the application of ANNs for multiple contamination scenarios achieved very satisfactory results for real DWDSs. The method was generally able to determine correctly the simulated source and to define a very restricted set of possible contamination sources, even when considering hydraulic scenarios with demand uncertainties. However, the estimations of the time of contamination for scenarios under demand uncertainties showed larger deviation in relation with the simulated contamination sources. The time of computation required was generally very low, which makes this method very suitable for application in real contamination scenarios.
- The source identification with general inverse problem methods developed by Irstea partner is a two-step enumeration/exploration method. Firstly, the in/out transport matrix is worked out with a backtracking method and then, minimum relative entropy method, without any assumption for the pdf (probability density function) distribution, or the least squares method with Tikhonov regularization are used to refine the results and give mean of being a source as a confidence interval for this mean. The backtracking algorithm yields good results in giving very quickly the full list of potential node sources of contamination at the different times, and the in/out (transport) matrix that gives the relation between the potential source and the detecting sensors. That matrix can then be used either on a minimum relative entropy method or a Tikhonov method. Both produces good results, the real contaminant source was always determined as potential source. The minimum relative entropy method looks discriminate more the potential nodes than the Tikhonov method (with selection criterion on strictly positive expected values – other are possible). For the Tikhonov the positivity constraint was relaxed. As a result, negative expected values are possible, and may be interpreted as non-important potential source contamination.

The method based on analysis of nodes where contamination is detected and flow directions has been also tested in different cases. The results allow concluding that the method gives good results especially for cases when a single contamination event happened. The method is fast and does not require any prior adjustment or "teaching".

4.2. Spread issues

Several approaches have been used to develop software tools for simulation of contamination transport. These approaches considered: a) an off-line software tool based on equations governing bacterial regrowth that is affected by sorption, desorption, chlorine and substrate concentration; b) a software tool considering sorption developed using MATLAB, VBA and EPANET with models for the evaluation of contaminant concentrations; c) an on-line software tool that uses flow direction data for tracking contamination spread.

- The off-line software tool is supported in a model developed in Epanet-MSX model. The model contains differential equations defining functions

of attached bacteria, bulk bacteria, substrate and chlorine concentrations as functions of time and considered also the phenomena of pathogen adsorption/desorption. The model parameters are user-adjustable as various types of contaminants have different adsorption and desorption coefficients. Graphical user interface of Epanet software has been added to the model. The graphical user interface allows operator to modify model parameters, set initial conditions (e. g. contamination sources), view results in graphical or tabular form as well as visualize distribution of contamination over the network. The advantage of the off-line software tool is that it contains a comprehensive model that besides convection takes into account adsorption/desorption and regrowth of bacteria as well as chlorine and substrate concentration. The model can be used to run simulations and study proliferation of contamination, long-term effects including sorption, effects of chlorine disinfectant addition and substrate concentration. The best application of the off-line software tool is to simulate contamination scenarios, look for vulnerable parts of the networks that, if contaminated, allow the contamination to spread to a large part of the network, test effects of different chlorine concentration.
- The software tool developed on Matlab and VBA enables testing the effect of sorption phenomena on contamination spread in drinking water distribution systems and studying long-term behaviour of a partially adsorbed contaminant in a drinking water distribution system. It has been shown that the proposed method is suitable for the study of the effects of the sorption phenomena in the modelling of the transport of contaminants in real drinking water distribution system.
- The on-line software tool enables running simulations of contamination transport in a water distribution network based on flow direction data. The concept of the on-line software tool is based on the idea that in case of contamination accident the affected area of the network is mostly determined by flow directions rather than flow magnitudes. Flow direction data can be obtained by means of flow direction sensors or by hydraulic simulation. Combined approach (flow direction sensors installed in some pipes, simulation is used for other pipes) is also possible. The advantage of the method is that in case flow direction sensors are used, the software tool uses real-time data from the network and therefore provides more robust simulation results.

5. CBRN analysis

Analytical methods were developed or improved in the SECUREAU project for a number of typical CBRN pollutants. Some of them were developed for SECUREAU teams' own purposes while others could be used in the case of contamination. All the methods used provided accurate results when determining the nature and / or concentration of a contaminant in water. However, determining the nature and / or concentration of a pollutant adsorbed onto the walls of the network was systematically found to be a major difficulty.

5.1 Organic chemicals

Analytical methods for the SECUREAU's chemicals in waters were implemented and fully validated, either for rapid detection of a deliberate contamination (higher limits of detection) or for controlling the efficacy of the decontamination (limits of detection compatible with safety requirements). For paraquat, chlorfenvinphos and carbofuran, existing methods were adapted, but for BDE-100 an entirely new analytical method was developed and validated, which has the advantage over the existing ones of requiring fewer sample volume. BDE-100 method consists in a dispersive liquid-liquid microextraction technique prior to gas chromatography-mass spectrometry analysis. A detection limit of 0.5 ng/L for BDE-100 was achieved. Additionally, this method has the advantage of being extended to the quantification of other polybrominated diphenyl ethers (BDE-28, 47, 85, 99, 153, 154 and 183).

If lower limits of detection need to be achieved, a method for the detection and quantification of carbofuran and chlorfenvinphos in waters was also developed. This methodology consists of a dispersive liquid-liquid microextraction followed by gas chromatography-mass spectrometry analysis and allows the detection at lower levels of concentration. Under the optimized conditions, the detection limits were 0.04 μg/L for carbofuran and 0.02 μg/L for chlorfenvinphos.

Concerning chemicals quantification in inorganic deposits, different approaches were taken depending on the chemical and on the nature and composition of the deposit. Different inorganic deposits were tested: iron rich deposit (APP1), manganese deposit (P45), calcium carbonate rich one (BayCa) as well as kaolin:

- Paraquat revealed an extremely high affinity to kaolin and only drastic conditions, such as reflux with concentrated sulfuric acid (during 4 h) were able to remove paraquat from it. Since the interaction between paraquat and real inorganic deposits (APP1, P45 and BayCa) is lower, a simple extraction with saturated ammonium chloride is sufficient to carry the analyte from the solid to the liquid phase, where it is quantified.
- A methodology was successfully developed for the quantification of chlorfenvinphos in kaolin. Acetonitrile was used as a desorption solvent of the pesticide from kaolin. The same solvent showed to be ineffective to desorb chlorfenvinphos from the real deposits tested (BayCa and P45). This result sustains that a matrix-matched calibration approach should be used for each deposit, for quantification purposes. Carbofuran adsorption to inorganic deposits showed to be slower than chlorfenvinphos. As for chlorfenvinphos, acetonitrile was not able to desorb carbofuran from the contaminated deposits.

Chlorfenvinphos extraction from biofilms was also studied. Acetonitrile was the solvent used in the desorption step , and it was able to achieve high levels of desorption of chlorfenvinphos from the biofilm, providing that the determination of the level of contamination of a particular biofilm is possible.

Determination of methylmercury (MeHg) was started with the addtion of sulphuric acid and NaBr to liberate MeHg from the matrix and to form a neutral MeHgBr complex. MeHgBr complex was extracted either with pure hexane from water and biofilms or with 50% dichlromethane/hexane from deposits. After extraction a volatile phenyl derivative of MeHg was formed. MeHg quantification method was based on GC-HRMS technique using isotope dilution. For water samples MeHg method showed good sensitivity (LOD of 0.21 ng/l) and 96-111% recovery. The method for biofilm/deposit samples showed 15-66% recovery yield depending of deposit matrix.

5.2 Biological agents

Methods for the detection of microbiological contaminants in water have been successfully developed and validated. These methods have been tested for their reliability and accuracy, particularly in the detection of

viable but non-culturable (VBNC) bacteria which can often be missed in routine measurements but could still pose a threat to public health.
- For Escherichia coli O157:H7, microscopy and molecular methods were found to be effective. The use of highly specific peptide nucleic acid (PNA) probes in a fluorescence in situ hybridisation (FISH) assay, combined with cell elongation (direct viable count, DVC), allowed detection and differentiation of viable populations (including VBNC). In addition, real time PCR (qPCR) combined with propidium monoazide (PMA) could be successfully applied, with a detection limit of 1 cell ml-1.
- Francisella philomiragia (as a surrogate for F. tularensis) and F. tularensis subsp. novicida could not be detected by DVC-PNA-FISH due to their small cellular size and extended growth time. However, the molecular technique of PMA-PCR could be successfully employed, with clear differentiation of live and dead populations in water.
- DVC-PNA-FISH was developed and validated for use in detecting Yersinia pseudotuberculosis (as a surrogate for Y. pestis) and PMA-PCR was validated directly on Y. pestis.
- To assess the applicability of these methods on a spore-forming bacterium, Bacillus cereus E33L was used as a surrogate for B. anthracis (as the closest molecular relative). PMA-PCR could not be used due to inability to ensure efficient PMA pre-treatment leading to unreliable results, although qPCR on its own could be used for general detection (with no viability assessment). In contrast, PNA-FISH combined with DVC was found to be highly effective at detecting both vegetative cells and spores, as it caused spores to germinate and show clear elongation.

For all bacterial species tested, the detection limit for PNA-FISH based assays was 2 x 104 cells ml-1. This could be improved by filtering larger volumes of water (e.g. 100 ml samples are routinely tested by water companies compared to these developmental assays where only 1 ml aliquots were used) and by using an automated microscopy system to allow complete scanning of filter membranes. The detection limits for PMA-PCR were as low as 1 cell ml-1 and were dependent upon DNA extraction efficiency. Improved filtration and automation of systems could allow detection of single bacteria in larger water volumes.

The use of amoebae in co-culture assays was assessed as a screening tool for the deliberate contamination of water. This was found to be effective in establishing bacterial viability and pathogenicity but due to the time required (1 week), it was decided that this would not be a useful tool in a time-limited, emergency contamination event.

The same microscopy and molecular methods were also tested for use in detecting bacterial pathogens in biofilms and deposits. DVC-PNA-FISH could be successfully used for E. coli O157:H7, Y. pseudotuberculosis (as a surrogate for Y. pestis) and B. cereus E33L (as a surrogate for B. anthracis) with the same detection limit values as for water samples. The specific PNA probes could clearly differentiate individual target bacteria within complex biofilm structures and by combining with DVC, viable populations could be quantified, including Bacillus spores. This was also observed for loose deposit samples but particular care was required to monitor autofluorescence.

With regards to PMA-PCR, for biofilm samples, low levels of target pathogens could be detected within these communities following efficient DNA extraction. The biofilm matrix did not affect results. PMA-PCR could not be used on biofilms spiked with Bacillus sp.. For the same reasons as the water samples and further work is required. When considering deposit samples, results were more variable. The composition of the various deposits could affect both the efficiency of DNA extraction and the light activation of PMA treatment. Although this technique could be applied to these sample types, careful validation is required to ensure efficient detection of pathogens.

Combining the microscopy and molecular techniques has provided a toolbox of methods which can be applied to water, biofilm and deposit samples to give accurate and rapid detection of viable target pathogens. These can be used for detection and for assessment of disinfectant and remediation efficacies. It thus appears that the only useful methods for detection of pathogenic microorganisms in deposits are molecular biology ones. However, many interferences lead to a high detection threshold for deposits, but PNA-FISH while appearing less sensitive was more reliable than qPCR.

In summary, total cells can be determined quickly, which is essential for rapid confirmation of an event. The determination of viable cells take longer: but this can be done at a more leisurely pace when it is more important to confirm successful decontamination efficacy.

5.3 Radionuclides

A range of rapid, off-line sensitive radiometric methods were selected and adapted to be able to confirm the intrusion or presence of radionuclides in drinking water networks (water and pipeline deposits). Each method was chosen for its potential to be analytically robust and for its capability in providing rapid screening to accurately detect and quantify alpha, beta and gamma emitting radionuclides, either singly or collectively. All methods were required to be free of any risk of generating false-negative or false-positive data for the broad range of SECUREAU list radionuclides (Cobalt-60, Strontium-90 + Yttrium-90, Iodine-131, Caesium-137, Iridium-192, Polonium-210, Radium-226, Americium-241, Californium-252).

- High-resolution gamma spectrometry (HRGS) is the first analytical choice to investigate unknown materials. As most of the relevant nuclides are ?-emitters the use of ?-spectrometry will ensure that these nuclides can be identified and quantified without any loss of time. The method is characterised by simple sample preparation and provides good identification and accurate quantification (with sufficient detection limits) with one measurement. Even though ?-spectrometry cannot detect all relevant radionuclides (only ?-emitters) it is highly recommended that ?-spectrometry is employed in the case of an assumed or real contamination incident on a drinking water distribution system in addition to LSCcounting or alpha spectrometry.

- LSC measurement or gross alpha-/beta-determination (GABD) is the most convenient off-line analytical method for the verification of a real or assumed attack on a water distribution system. Liquid scintillation counting coupled with spectral analysis (LSC-Spectra+) is preferred to GABD when analysing waters directly or allied with simple acid treatments (leading to dissolution/digestion/decolorisation) for biofilms and deposits. The technique is capable of identifying and quantifying the target radionuclides with limits of detection of 1 Bq/g or better with preparation and counting times taking less than 3 hours. The sensitivity is acceptable for the detection of relevant concentrations of nearly all of the SECUREAU target nuclides.

- A complementary analytical approach to the rapid LSC LSC-Spectral+ method described is alpha spectrometry that uses the specially developed ADAM method (Advanced Deconvolving of Alpha Multiplets). Simplified sample manipulation with sophisticated analysis methods is advantageous when obtaining the results rapidly is of importance. In a radiation emergency event the heavy elements, like uranium and thorium, are rapidly detected using ICP-MS technology. Liquid scintillation spectrometry has advantages for detection of alpha and beta emitting radionuclides simultaneously. However, identification of radionuclides in liquid scintillation spectrometry is challenging because spectrums and peaks overlaps. In spite of that, information from alpha and beta emitting radionuclides can be achieved. Where more detailed identification of alpha particle emitting radionuclides can be achieved by water evaporation, alpha spectrometric measurement and ADAM analysis. In the case of Deposits and biofilms Simplified sample manipulation with sophisticated analysis methods is advantageous when it is necessary to obtain results rapidly. However, straightforward biofilm detection cannot entirely replace the radiochemical sample processing because thick sources may prohibit the analysis. In such cases real deposits cannot be analysed without some sample pre-treatment.

Overall, in the event of an emergency situation the simultaneous application of ADAM, LSC with Spectral Analysis and HRGS provide the most effective way of rapidly acquiring radionuclide data in the event of an incident. All of the SECUREAU-list radionuclides could be effectively identified with sufficient analytical sensitivity to be able to detect activities well below the recommended safe guidelines for drinking water quality (at least down to the maximum permitted levels defined in the Euratom Regulation 2218/89). The same methods, with some modifications, could also be applied to the more challenging case of biofilms and deposits. In some cases the methodologies allowed a rapid screening for many of the radionuclides (alpha, beta and gamma) that could be followed up with specific but slower methods for identification and quantitation. The methods are acceptably sensitive, detecting activities below the recommended safe guidelines. If only one radionuclide is introduced in the system, there is no longer need to identify them each time. For such a purpose LSC is interesting, with automatic sample changing and a near 100% counting efficiency in addition to contamination control (LSC vials can be closed more securely than gamma vials).

6. Modeling sorption and desorption

In case of contamination of the water in a drinking water distribution network, the obvious response is discarding the contaminated water volume and to rinse the system. However, as the contaminants will interact with the surrounding surfaces (with biofilms) and deposits by adsorption and desorption, such phenomena should be assessed in order to predict the opportunity of cleaning methods.

The complexity of the sorption and desorption processes is given by the different sorption properties of the pipe material surface, abiotic mineral surfaces and the biofilm as a three-dimensional gel-like biofilm matrix which acts like a sponge. What makes biofilms even more complex is the fact that the highly hydrated extracellular polymeric substances (EPS), the cell surfaces and the cytoplasm of the biofilm cells represent three different compartments, again with different sorption properties and capacities.

In principle, pipe materials and their deposits act as a sink for a certain proportion of the contaminants. However, this sink is not irreversible but over time, when the water phase is rinsed free of the contaminant, they will follow a concentration gradient and re-contaminate the water to an unknown extent and period of time until everything is washed out. Of course, it is impossible to determine the sorption and desorption behavior and kinetics of every thinkable contaminant, but with the concept as developed in the project, reasonable approaches for detection and sanitation as well as a decision tool could be generated.

In order to address this situation, experiments have been carried out to assess the sorption and desorption kinetics of model substances, representative for toxic metals, radioactive substances and microbial pathogens with particular consideration of biofilms. The following main contaminants were selected with the aim to illustrate methodologies for assessment of adsorption-desorption kinetics, test mathematical models and provide values of kinetic parameters that will contribute to simulate the impact of sorption phenomena on the water safety:
(i) Chemicals - paraquat and mercury (II) chloride (preliminary tests were also made with Chlorophenvinfos and Carbofuran);
(ii) Biological agents - Spores of Bacillus subtilis (surrogate to Bacillus anthracis), Yersinia pseudotuberculosis, Francisella philomiragia (surrogate to Francisella tularensis) and Adenoviruses.
(iii) Radionuclides - Am-241, Po-210, Sr-90, Ra-226;

The kinetic parameters obtained from these experiments have been used in the overall modeling of the drinking water distribution network, in order to simulate and predict the spread of contamination in case of a deliberate harmful intrusion.

The modeling approach of the sorption kinetics followed in the present work does not discriminate the mass transfer effects inside the sorbent, nor specify possible chemical or biological reactions within the sorbent structure. Therefore, the correspondent kinetic parameters describe, from a general macroscopic viewpoint, all the process occurring inside the solid phase. However, external mass transfer processes are discriminated, in order to take into account the different hydrodynamic conditions that may prevail along the network.

The "observed" rate at which the overall contamination process occurs in the liquid and solid phases is determined by the balance between the rates of the two individual processes: (1) external mass transfer rate in the water, and (2) sorption kinetics inside the sorbent. Since these two processes occur in series, the slower one will limit the overall observed adsorption rate. For example, if the hydrodynamic conditions in the liquid promote (by using intense agitation or high flow velocity) mass transfer rates that are potentially much higher than the local adsorption rates, the overall rate of the process will be practically equal to the local adsorption rate in the sorbent. For lower mass transfer velocities, the bulk liquid phenomenon may become the rate-limiting step of the overall process.

The results highlight the following conclusions:
- Important effect of the surface area available for adsorption of chemicals: suspended particles offer a much larger surface than attached deposit layers (for contact with the contaminated water), and that

explains the much higher rates of adsorption to particles in suspension (almost 2 orders of magnitude) than to attached deposits.
- Attached biofilm layers display adsorption kinetics more similar to suspended particles and much higher adsorbed amounts than in abiotic deposits, which may be due to the considerably higher porosity (greater than 90%) and internal surface area of biofilms.
- Mixed suspensions of particles (kaolin + iron oxide) have faster adsorption than simple kaolin suspensions. Once again, the surface area effect seems to predominate due to the very small dimensions of the Fe oxide particles (0.5 µm, versus 15 µm for kaolin particles).
- Adsorption to (and desorption from) clean pipe surfaces, without any attached layers, occurs usually at very low rates. The same is true for desorption from biofilms.
- The kinetic values for adsorption of mercury (II) chloride to removed deposits (here called "scraped deposits") APP1 and P-45 are much higher than for paraquat adsorption to the same sorbents, although the agitation was more intense in the paraquat tests. The size and shape of the two contaminant molecules may be a possible explanation for these clear differences.
- When comparing the adsorption of Hg2+ and paraquat to attached corrosion layers, the equilibrium concentration is higher for paraquat, but the rate constants (k1 and k2) are clearly larger for Hg2+ adsorption. This should be further investigated, but it is possible that the corrosion deposits used in the two kinds of experiments (mercury and paraquat) had different structural characteristics (porosity, tortuosity) that could not be assessed.
- The experiments with different hydrodynamic conditions (different agitation speeds) indicate that there is a slight effect of external mass transfer on the "apparent" rate constants. This effect is clear when comparing agitation versus no-agitation examples, but tends to become negligible for rotation speeds above 200-300 rpm (paraquat sorption). Mass transfer coefficients in the water were calculated for different agitation velocities, using correlations available in the literature, and they show a slight increase from 330 rpm (1.0 x 10-4 m.s-1) to 550 rpm (1.16 x 10-4 m.s-1). However, for no agitation (0 rpm), the mass transfer coefficient decreases to 1/3 of these values (3 x 10-5 m.s-1). Additionally, the first-order rate constant (k1) was compared with the mass transfer coefficient for the test carried out with a sorbent/contaminant ratio of 40 mg/mg and rotation speed of 440 rpm. In order to convert the dimensions of the rate constant (s-1) to dimensions of mass transfer coefficient (m.s-1), k1 was multiplied by the ratio of liquid volume to sorbent area, resulting in k1*(VL/A) = 1.2 x 10-6 m.s-1. This shows that the adsorption rate constant is 2 orders of magnitude below the mass transfer coefficient in the same experiment (1.1 x 10-4 m.s-1), i.e., external mass transfer effects are not relevant except when there is no agitation (or only very weak agitation).
- The two adsorption models, pseudo-first and pseudo-second order, fit quite well the experimental data.

7. Pipe wall cleaning and decontamination

Overall strategy proposed in SECUREAU was to carry out in situ cleaning (i.e. inside the pipe) and then flush neutralised sludge out of the system for safe disposal. There are some risks with most of these methods, namely, if cleaning is very intensive, pipes can be damaged and contamination will leak out to groundwater. It is therefore in cleaning step we avoided to apply "aggressive" methods such as plasma discharge. On other hand flushing should be more effective than traditional methods, which is mostly focusing on removing loose deposits. It is because corrosion layer inside the pipe will absorb some of contaminants and the only way to remove it is to flush it out with incrustation layer.

The limitations of currently used methods for cleaning drinking water distributions systems were taken into account and effective but simple solution "how to deal with adsorbed CNBR agents" were proposed. Depending of the methods, some of them were applied only in laboratory scale, other also in pilot scale, whereas one of them was tested also in full scale in an old, abandoned network.

Moreover in selection between different methods, it was clear that cleaning should not be to external expert demanding, since the first work will be done by water company staff and their major expertise is relevant in flushing and disinfection. Assuming that the first reaction can be decisive, the technologies proposed here are not requiring too much of professional experience in dealing with hazardous wastes.

Experience from contamination events in drinking water distribution systems showed that the procedures of decontamination are cumbersome and time-consuming when attempting to reach acceptable levels of safety. A number of challenges should be solved to make decontamination methods more efficient, among which are persistence of contaminants in pipes and deposits, recycling of contaminated water and exposure of emergency teams to contaminants. The following steps should be followed to ensure efficient cleaning:
Selecting the most suitable decontamination method should depend where the contaminant is located (the red cross on the picture on the right):
- at the surface, associated with loose deposits and / or biofilms. If such is the case, can traditional techniques (e.g. water/air flushing) allow this surface layer to be removed; or can disinfectants be effective in such complex environment?
- deeper in the deposits, due to an effective diffusion of the CBRN agent in the material. If such is the case, which method can be used for removing the deposit as it is unlikely that traditional methods will not work !

7.1. Bacterial agent decontamination

Both spores, non-spore forming bacteria, and viruses were used as models for testing decontamination procedures which are summarized.

Shock-chlorination was studied by adding high concentration of chlorine and keeping to reach optimal CT (concentration multiplied by time) value. Results showed that 5 log10 inactivation of B. subtilis spores are achieved within 3 hours using 200 mg/L of free chlorine (pH adjusted to 6) in the bulk. Although reasonable efficacy was observed in water, biofilm were not effectively removed. Moreover very high CT value can decrease the integrity of the pipes. Experiments with non-spore bacteria showed that disinfection with shock chlorination providing sufficient contact time is an effective method for neutralising bacteria both in water and biofilm. To ensure that this kind of treatment killed also viable and not culturable bacteria the lack of metabolic activity was confirmed with molecular viability methods (e.g. FISH-DVC).

The use of NO treatments to trigger biofilm dispersal events in drinking water systems was found to be ineffective. We would conclude that there is no added benefit in using NO treatment as part of a remediation toolbox following a deliberate contamination event.

Ultrasound cavitation may be used to detach spores from surfaces towards a further procession (i.e. DNA analysis or quantification) but it should be followed by other disinfection methods, which together is too expensive to use in case of contamination of distribution system. Thus, in general the study showed that application of cavitation for removal of spores from drinking water distribution networks did not show significant advantages comparing to conventional chemical disinfection methods.

One of the most promising results on surface disinfection was the regime alternating free chlorine (200 mg/L) and sodium hydroxide (1.5%). This technique is based on spore disinfection in a bulk and afterwards releasing of spores adhered to the surface.

Advanced oxidation process (oxidation through reactions with hydroxyl radicals) was successfully tested in SECUREAU. To take advantage of iron and cupper in water distribution systems and in biofilms, Fenton-like of advanced oxidation reaction was selected to inactivate B. subtilis spores in water and indigenous bacteria biofilm. The results showed that it is possible to inactivate B. subtilis spores rapidly and effectively in water using copper as a transition metal and ascorbic acid as a catalyst (without pH correction), or iron from the corroded pipe walls.

7.2. Decontamination from organic agents

Paraquat (a highly soluble (620 g/l) and highly toxic pesticide), Chlorphenvinphos, and BDE were used as model substances of organic agents in SECUREAU. To follow the principle of in situ cleaning we used Fenton's reaction to mineralised organic to inorganic carbon by means of peroxidation. The process involves a complex mechanism in which the parent molecules are oxidized into other organic compounds and ultimately into carbon dioxide and water. The organic matter oxidation is promoted by the hydroxyl radicals formed in the reaction between hydrogen peroxide and iron. The catalyst, Fe (II), is restored by the reaction of hydrogen peroxide with ferric iron.

In order to reduce the amounts of reagents used in the process (namely ferrous solutions), and trying to avoid the formation of iron sludge, the possibility of using iron-containing materials, or solids with iron attached to its surface, has been widely addressed by several authors through the so-called heterogeneous Fenton-like processes. Four real pipe deposits with different chemical compositions and morphological properties were tested as catalysts in paraquat heterogeneous peroxidation. Results showed that some of the deposits taken from water networks can be used as catalysts in the peroxidation of paraquat. The deposits are quite complex and the reasons for their completely different performances are not yet completely clear. Probably the form of iron oxides presented in deposits and also presence of other metals and substances plays important role. A better degradation is achieved when water was acidified to the pH of 3.0. Depending on the types of solids present in the network, in situ treatment can be made without insertion of chemicals apart from the oxidant, in case of a contamination event.

Concerning paraquat dichloride, a maximum mineralization degree of 60 % was attained after 240 min for dark classic Fenton (i.e. oxidation of paraquat and its intermediates up to CO2 and H2O), although the pesticide was completely degraded in much less time. It was possible to identify three intermediate compounds resultant of the process: oxalic acid, isonicotinic acid and 4-carboxy-1-methylpyridinium ion. From the 40% of organic carbon remaining, the conversion values estimated were 11 % to oxalic acid, 13 % do 4-carboxy-1-methylpyridinum ion and 3 % to isonicotinic acid. This means that 27 % of the 40 % of remained organic matter is known. However, the identified compounds are less toxic than the original one (paraquat).

Chlorfenvinphos and carbofuran conversions of 35 % and 52 % into CO_2 and H_2O were, respectively, achieved when dark classic Fenton was used as decontamination process. Again, the parent molecules were completely degraded by this advanced oxidation process. Experiments with BDE-100 proved that degradation of similar water contaminants is a good approach as a first and simple measure of water decontamination.

7.3. Decontamination from inorganic agents

Mercury was used as model substance of inorganic agent in SECUREAU and several methods were tested (water flushing with chlorinated or non chlorinated water; ice pigging). The results showed that in smooth pipeline materials (such as PEX) large part of Hg^{2+} was sorbed to the biofilm surface. Then an effective removal of biofilm also removed large major portion of Hg^{2+}. A 5 min high water flow flushing reduced the concentration of Hg^{2+}in the biofilm only by 58 %. Use of low flow chlorinated water (10 mg/l Cl_2) flushing for 1 hour reduced the Hg^{2+}concentration in the removed biofilm more 92 %. Total removal rate taking into consideration the fraction remaining in the pipe surface after biofilm removal was 82% with chlorine flush. Flushing time with chlorine should have been longer as Hg^{2+}concentration in both water and biofilm were still clearly decreasing after 1 hour. Higher chlorine concentrations, higher flushing time and possibly also higher flow rate might have helped to reduce the Hg^{2+} concentration in biofilms and water to an acceptable level. Also, more effective biofilm and corrosion scale removal methods such as ice pigging + Comprex would most likely be very useful. Decontamination of corroded cast iron tube by mixing with ice cubes was found ineffective as 40 % of Hg^{2+} was bound in the deeper layers of the corroded surface already after 2 hours of incubation that were not removed by shear forces of modest ice flow. Highly effective means that remove the corroded surface of pipelines, such as Comprex with small stones added, should be used for decontamination.

7.4. Decontamination from radioactive agents

Radionuclides can be introduced as in water supply by dissolving in water and depending on their type and species they can strongly absorbed adsorb on the surface of pipes (e.g. cast iron). Thus, their should be desorbed and later concentrated for the safe disposal.

In SECUREAU, chemicals (calcium acetate, sodium bicarbonate, sodium citrate, tri-n-octylamine, ethylenediaminetetraacetic acid and sodium hypochlorite as an traditional disinfectant) as release agents of radiological agents (Americium, Polonium, Radium and radiocolloids) from pipe material (stainless steel, polyvinyl chloride, high density polyethylene, polypropylene) and real pipe deposits was tested.

Acetate is a widely used anion in inorganic, organic and analytic chemistry because it is able to form several compounds. $NaHCO_3$ (or baking soda) is safe and common compound and it has a generally recognised as safe (GRAS) status by FDA. CO32- and HCO3- ions have been reported to make complexes with different radionuclides and they have been used for cleaning radioactive waste waters. Citrate is also used for decontamination of radioactive materials, either by itself or mixed with EDTA. In addition to selected chemical agents, we decided to include NaClO in the tests to find out how this traditional disinfection chemical affects the contamination.

The efficiencies of decontamination were compared to decontamination (desorption) efficiency by water which was the control sample in our tests. Sodium bicarbonate was the most effective chemical for cleaning 210Po and 241Am from selected pipe materials. Compared to other decontamination chemicals, NaHCO3 is also cheaper and safer chemical to use. The average for tested chemicals are given. Even though, similar decontamination results were obtained by EDTA as bicarbonate, its use should generally be avoided. As a complex ion, EDTA tends to form stable complexes with different types of metals. When released to the natural waters, EDTA may complex heavy metals from sediments and hence reintroduce them into the food chains. Real pipe deposits were difficult to contaminate by 90Sr, 210Po or 241Am in the first place. It was only possible by lowering the pH-value to less than2.5. The deposit contaminated this way could not be decontaminated by the studied chemical agents.

8. Handling of decontamination sludge and water

In SECUREAU, the applicability of the following sludge handling technologies were investigated: i) separation of solids from water with flocculation and sedimentation, ii) removal of radioactive substances with sorbents; and iii) removal of organic substances with advanced oxidation.

During decontamination process large amount of sludge are generated. If reservoirs to store them are not available nearby a viable option is to use transportable plastic containers. The advantage of this using theses type of containers is more safety against aerosolisation of contaminants present in the removed water. Some of them are more (e.g. spores) dangerous in air- then in water-phase. Removed sludge usually contains 99% water which makes volumes high; it is therefore necessary to decrease the volume. Sludges should dewatered to 20-25%, which allow them to be stored and neutralised.

Several flocculation agents were tested for their efficiency to improve the sedimentation of loose deposits removed during the distribution networks cleaning with air scouring method. Results showed that suspended incrustation from cast iron pipes have very good flocculation and sedimentation behaviour using a combination of two products: flocculants and flocculants aid. Turbidity after flocculation and sedimentation process decreased from 500 NTU to less than 0.2 NTU using cationic polymers. This is reasonable effectiveness of flocculation; 99.63% respectively 99.83% of the solid matter is deposited and settled as sludge.

The potential of magnetic ferrous sorbents to adsorb radionuclides from water and biofilm was studied. Ferrous sorbents with improved adsorbing properties were produced by consortia of sulphate reducing bacteria grown in continuous culture with magnetic feedback enrichment. The sorbent was used to study radionuclide removal in the laboratory scale. The ability to produce relevant magnetic ferrous sorbents was achieved, showing that up scaling of culture methods could produce large amounts of sorbent; furthermore this material was shown to be highly magnetic and consisted mainly of vivianite. The results of the adsorption experiments demonstrate a range of adsorption abilities and rates. However for most of the radionuclides tested there was a minimal change in such adsorption abilities across an environmentally relevant pH range of pH 5.5, pH 7 and pH 8.5. Adsorption levels observed were as high as 100% for Pb-210 and Ra-226, but as low as 14.4% and 16.1% for Cobult-60 and Cesium-137, respectively, at pH 8.5 after a 7-day incubation period.

The Fenton reaction was used to degrade two chemicals: paraquat and Chlorfenvinphos (CFVP). Total organic carbon removal (i.e. mineralization of the chemicals up to CO_2) with this advanced oxidation process was also studied in lab-scale. To study the degradation of either compound, variations in the initial concentration of H_2O_2, Fe^{2+}, initial pH and temperature were done; such detailed parametric studies allow optimizing the process performance. CFVP degradation depended on the operating conditions employed, and this significantly improved with the increase in the temperature; regarding the effect of pH, it was observed that the degradation is better when using the initial value of 3.0, although higher values can still be employed. For the other parameters, i.e., initial concentration of Fe^{2+} and H_2O_2, it was found that they affect the degradation of the compound in a positive or negative way, i.e., depending on their values and on the ratio between them. Similar results were found for paraquat. The Fenton process and the mechanism is complex, but those results can be explained by the existence of undesirable parallel reactions when one of the species is in excess or by the slowdown when one of the reagents is present in a deficit ratio. For both chemicals, results obtained are promising; thus, the implementation of this advanced oxidation process should be attempted in pilot-scale.

9. Verification of decontamination

In SECUREAU, several solutions and strategy for effective in situ cleaning based on using simple reagents were selected:

- Chemicals such as pesticides as well as pathogens and autochthonous bacteria could be removed using H_2O_2 for generating free radicals (Fenton's reaction). Metal pipe surface (iron and copper) can be used as catalysts of the reaction. In some cases, the treatment can be carried at pH near the neutrality;
- Removal of resistant microorganisms such as Bacillus spores from pipe surfaces could be achieved by alternating treatment with sodium hydroxide and chlorination;
- For removal on radionuclides, desorption by sodium bicarbonate solution and flushing of the system for safe storage or utilisation offer a remediate option.
- Ice slugs and gravel in combination with water flushing are effective methods for removing both loose deposits and corrosion layer of pipes. Such a treatment is needed as soon as chemicals are suspected to diffuse and sorb easily in the deposits.
- For separation of sludge from water chemical coagulation with addition of flocculants was shown to be effective for handling of decontamination waste.
-

Whatever the solutions, verification of the decontamination level of the pipe wall is requested either by deposits, biofilms or scale analysis (from in situ samples or sentinel coupons analysis); or by using sensor signals.

9.1. Off-line detection of contaminants in deposits

9.1.1. Chemicals

Acquisition of sufficient amounts of biofilms and the interference of abiotic deposit components with their heterogeneity is a considerable challenge but could be achieved. Methods were developed for paraquat, chlorfenvinphos and methyl mercury in deposit and for methyl mercury in biofilms. Validation included the linearity of the response, the limits of detection and quantification, the precision as expressed by the variation coefficient, the accuracy evaluated by the percentage of recovery from artificially contaminated samples and by the matrix-matched calibration. Paraquat as a representative for herbicides with highly polar properties is strongly adsorbed to the deposits. Therefore, drastic methods were applied for extraction, indicating a low recontamination potential. Chlorfeniphos was more easily extracted and, thus, is more likely to be leached out of a deposit after adsorption. Methyl mercury was determined in both biofilms and deposits and could be reliably detected.

9.1.2. Pathogens

Two general detection strategies have been pursued and compared with more conventional culture methods. One was cultivation based. This method of culture detection of microorganisms is the so-called "gold-standard". However, it is well known for a number of limitations, the most important one being that lack of culturability does not mean lack of presence and possible recovery of microorganisms. Such transient loss of culturability has been termed a "viable but non-culturable" (VBNC)-state and is understood as a stress response. Two main alternative culture-independent and molecular-biological approaches were assessed for application on water samples; the microscopy based combined techniques of cell elongation (DVC) and peptide nucleic acid/fluorescence in situ hybridization (PNA-FISH), and the combination of propidium monoazide with real-time PCR (PMA-PCR). In SECUREAU, these approaches were applied both on drinking water biofilms and on loose deposit samples. Tested pathogens and surrogate bacteria could be detected, and viable populations quantified, using the DVC-PNA-FISH method. Tested pathogens and surrogate bacteria could be detected, and viable populations quantified, using the DVC-PNA-FISH method. These methods are not trivial and require validation. Quantitative polymerase-chain reaction (qPCR) was suited to detect low levels of target bacteria when DNA extraction had been successful, although it cannot distinguish between viable and dead cells. Both of these methods (DVC-PNA-FISH and qPCR) allowed to detect lower levels of target spike bacteria than culturation methods alone, indicating the presence of VBNC cells. In addition, a new protocol for the permeabilization of Bacillus spores was developed and tested. The application of this method will allow rapid and accurate staining of spores and permit easier detection in these complex sample types. The advantages and disadvantages of these methods on a range of samples is now understood and will enable these techniques to be for pathogen detection and quantification in water, biofilm and deposit samples.

9.1.3. Radionuclides

The methods for detection of radionuclides as adapted were acceptably sensitive, even at activities below the recommended safety guidelines.

- Liquid scintillation counting coupled with spectral analysis together with acid treatment of biofilms and deposits are shown to be effective.

The technique offers a rapid approach for the identification and quantification of radionuclides in pipeline deposits in an emergency situation.
- The analytical methods are suitable for rapid off-line determination of radionuclides both in abiotic deposits and biofilms.
- Simplified sample manipulation with sophisticated alpha spectrometric analysis (ADAM method) is advantageous if it is necessary to obtain rapid results.

In general, the suitability of the methods for detection of radionuclides down to levels permitted by the safety guidelines was successful.

9.2. Sensors

To obtain information about surface deposits in drinking water pipes, the most direct way is to sample pieces of the pipe and analyze the deposit on their inner surface. However, this requires a construction pit and the replacement of a piece of the pipe. Furthermore, for assessment of cleaning success, another piece of pipe has to be extracted and analyzed.

For deposit analysis, basically three options are possible as already described for fouling monitoring in water systems:
i) Sampling of surfaces by gaining physical access and removing the deposit by scratching or wiping from a defined surface area with subsequent analysis in the laboratory.
ii) Exposure of test surfaces ("sentinel coupons") which are located at representative sites and removed after given periods of time or after treatments, again with subsequent analysis in the laboratory.
iii) Installation of sensors which possibly provide information on deposit formation and removal in situ, on-line, in real time and non-destructively, which was considered an optimal way of monitoring.
To overcome these problems, the use of so-called sentinel coupons is one possible option. Such a coupon monitoring device should fulfill the following requirements: small and easy to exchange surface pieces; easy to sample; possibility for multiple samplings; possibility for easy offline analyses in the laboratory; surface characteristics, geometry and hydrodynamics similar to and representative for real pipe surface.

A simple system of small pieces cut from a drinking water pipe (coupons) put together in a metal holder and assembled into a housing to be installed in the drinking water system was developed to meet the above mentioned requirements.

In addition to sentinel coupons enabling the off-line analysis of pipe wall deposits, online monitoring by deposit sensors providing real-time information on pipe wall deposits is desirable.

Such online deposit sensors should fulfill the following requirements: online, in situ and real-time measurement of surface deposits; detection of presence of deposits; quantification of deposits; characterisation of the deposits; no requirement for sample removal, staining or any other secondary procedure.

As part of the SECUREAU project, online sensor systems as well as coupon monitoring devices were developed and tested.

In comparative investigations it was possible to demonstrate the applicability of the sentinel coupon monitoring devices to follow the build-up and removal of biofilms in a pilot test system. Therefore, it could be deduced that coupon-monitoring devices are suitable to follow the deposit/biofilm formation in drinking water distribution systems as well as to investigate and validate the success of removal of the deposits from the surfaces.

Online sensor systems (OptiQuad from Krohne Optosens GmbH and SkidSens/FS-900/FS-1000 from Neosens SA) were tested for their ability to detect and quantify surface deposits as well as to confirm their removal. For these sensors, the trend in sensor signal and the biofilm growth curves fitted well. Good correlations between fluorescence signals of OptiQuad sensor and total cell counts (R^2 greater than 0.9) were observed. In the case of NeoSens system the measured deposit thickness showed a correlation to the determined total cell counts but some deviations from the OptiQuad sensor signal and the off-line total cell counts were observed in some cases.

In addition, both sensor systems as well as the coupon monitoring systems were successfully used in verifying the success of the tested disinfection/cleaning procedure. Therefore, it could be assumed that the systems (sensors and coupon devices) are suited to evaluate rehabilitation measures in drinking water systems.

The OptiQuad sensor was further able to detect inorganic particulate deposits by using the scattering signal measurement and a correlation between the sensor signal and thickness of the inorganic deposit was shown. Furthermore, using the different measurement principles of the sensor in combination, differentiation and selective quantification of biotic (biofilm) and inorganic deposit constituents was possible.

10. Decision tool

The main goal of drinking water supply is to provide to customers an uninterrupted supply of water with a high aesthetic quality and with a minimal risk to human health. However, because of potential hazards in the raw water, treatment and distribution parts, water supply systems and consumers may get exposed to a wide variety of risks. In addition, crisis situations as e.g. a terrorist attack on the water supply system are potential risks with high impact. As a result, water companies and authorities have to continuously adapt to manage those risks and to be prepared in such crisis situations.

In general, existing crisis management structures give only the organizational framework but not specific tools or regulations neither for specific crisis situations nor for appropriate and effective countermeasures in such cases. The SECUREAU project provides more detailed methods and tools to be included in the pre-crisis phase (preparedness) as well as in the crisis phase (response and recovery). One important part of a crisis management will be the "decision making" which is in the focus of the present deliverable.

In case of a deliberate attack, the drinking water supply should be restored in a way to ensure secure and hygienically proper drinking water to the customers. Therefore, the absence/level of the contaminant in the water phase as well as the removal of the contaminant from the distribution system (pipes, deposits/biofilms) has to be evaluated. In any case, the "absence" of the contaminant could most likely not be proved to be zero but only to be below a certain limit (detection limit, acceptable level, etc.). Thus, after a decontamination procedure (or even before) it has to be judged, if an acceptable low concentration of the contaminant in the water and in the deposits has been reached. Only if this criterion is reached, the clearance can be given to distribute drinking water to the customers again and to return to "routine operation" (post crisis phase).

To support the responsible persons/authorities to take such a decision, a decision scheme was developed in the SECUREAU project.

For the validation of a decontamination success, the absence of the contaminant or at least the decrease of the contaminant concentration to a sufficiently low concentration has to be verified. Therefore, in the first place the identity of the contaminant must be known and a suitable and sufficient sensitive analytical method must be at hand. Both information should be available from the inventory phase. Second, the site(s) where to take samples and to analyse must be known. This information should be available from the modeling approach and the analytical data in the inventory phase. Third, the validation has to be performed under consideration of target values or acceptable contaminant concentrations derived from health based risk assessment.

For the validation of the success of a performed decontamination measure, two points have to be addressed: in the first place, the water supplied to the customers must be acceptable, thus the contaminant must be absent or below a concentration derived from health based risk assessment. In the second place, the network (pipe surfaces, deposits) must be in a state that a recontamination of the water by possibly still contaminated deposits on piping surfaces is excluded. Thus, the contaminant should be either removed from the pipes and deposits or the deposits itself have to be removed if there is no other solution.

To validate the first requirement (no contaminant in water), the investigation of water samples taken from the network and customer taps is an appropriate approach. The main problem in this task will be the necessity to prove the good water quality at sufficient places to ensure the absence of the contamination within the entire distribution system. Thus, many sampling points (their number depends, e.g. on the DWDS, on the contaminated zone, or other factors) must be selected inside the contaminated zone and the adjacent zone including worst case locations e.g. terminal end pipes, low flow regions, house hold connections, etc.

To validate the second requirement (no contaminant in pipes/deposits) a sampling of the pipes and/or deposits is necessary. This may include real pipe pieces and sentinel coupons. In addition, data from online deposit sensors should be included. Again, the problem will be to include sufficient sampling points to ensure the absence of the contaminant in the network. Therefore, at least several representative places of the network as well as worst case locations (high deposit amount, high corrosion rate, low flow regimes, etc.) must be investigated.

In both cases, the validation has to be done in combination to health risk assessment. This would be most easy in case of the water phase, since uptake, daily dose, exposure etc. can be more or less well defined. For the deposits on pipe surfaces it is more difficult to define target values on a health based approach. Factors like desorption, sudden release events such as sloughing, release due to maintenance works on the network etc. are difficult to predict or to calculate. This is especially a problem if the contaminant was not been fully removed from the deposit or the deposit including the contaminant could not be removed. In this case it might be a constant reservoir of recontamination until the entire amount of the contaminant is washed out. This is even more complicated if the contaminant is a microbial agent with a potential to grow (mainly bacteria). Such bacteria remaining in the deposit may survive there and can even multiply, thus bearing a constant risk of recontamination of the water phase.

The application of cleaning procedures coupled with analytical methods in water and on deposits is used to finally decide if the network is back to a normal, "healthy" state, i.e. the water supply can be reopened to all consumers.

Potential Impact:

- 1. Security and safety synergy reinforcement for EU citizens benefit
SECUREAU project opens the way for a new drinking water network every-day management, as well as a new way for rapid detection and cleaning of contaminated networks. The alliance done between new information and communication technologies and drinking water expertise allowed to re-invent drinking water supply in several ways. SECUREAU project provides solutions designed for "security" applications (detect terrorism attack). Nevertheless, the Early Warning System (EWS) based on the multi-parameter sensor network developed has another purpose: this second use is dedicated for day to day operations. Allowing abnormal change detection of water quality, the EWS allows operators to have a better control of the water quality impact of standard waterworks: washing tanks, pipe renewal, valves closing... Moreover, this system is able to detect incidents such burst, under/over chlorination dosing, pressure control system failure... all the major events affecting the water quality and the water supply services. Finally, such tools could be used to improve asset management strategies. Usually the pipe renewal schemes are only driven by failure rate or burst model forecast; a water quality criteria could be now added to sort the list of pipes to be renewed in priority. As investments of municipalities are under constraint, optimizing the renewal effort without decreasing the safety of the water supply for the European citizen is a major output from SECUREAU.

The feasibility of both purposes "safety" and "security" were demonstrated within the project in two networks in a huge European case study (rural network: 50,000 inhabitants, and urban network, with 400,000 inhabitants) and at pilot scale. This double purpose allows municipalities to amortize the security investment by operation saving. The day to day use of such tool reinforces safety of supply water by a better traceability of waterworks impact. Moreover, the full scale demonstration brought by the urban and rural networks case study validates the strategic approach selected within the project as a good alternative of US homeland security program. Contacts with national "Civil Security and Defence" department confirmed this purpose. The EU expertise could be exported to developing countries where security issues are under focus: middle east, China, south America.

- 2. Development of EU industry and leadership on environmental sensors networks
Another benefit of the project is the sensor approach. Sensors suitable to determine surface and deposit contamination have been evaluated. Removable surfaces ("sentinel coupons") have been exposed which had the same history as the water system they survey. These can be sampled at suitable points in time, e.g. to verify a contamination and to verify cleaning success. The technical development of such sensors is a typical strength of creative SMEs and will provide them with a selection advantage on the market. More sophisticated devices allow for on-line, real-time, in situ and remote sensing and to survey very valuable systems. Finally, mathematical modeling tools have been developed which allow for optimal positioning of sensors. Such models improve the application of the sensors dramatically as they can be positioned in the most representative locations and improve their early-warning capacity. SMEs or stakeholders which implement these models will be competitive for protection of water systems. As soon as the sensors can be produced more cheaply, they will be powerful instrument to detect biofouling in

industrial systems (e.g., heat exchanger, membrane treatment) early, allowing for timely countermeasures and optimization of efficacy. Additionally the high level of technology developed within SECUREAU project in addition of full demonstration reinforced the industrial tissue of EU. For instance: Kapta 3000 AC4 probes as well as GSM communication modules, even if they were designed by a Swiss company, commercialized devices are now produced in England. Kapta 3000 OT3 components were provided by an innovative French SME. During the project, vertical integration of some strategic SME within a worldwide European company has to consequence the European leadership improvement in environmental sensors network market. These devices will be included in global offer for drinking water supply monitoring dedicated to water Safety and Security.

This offer includes device furniture, installation, maintenance, communication and visualization thought web access service. Kapta 3000 RAD is a co-development between CEA and VEOLIA at a proof of concept stage. All specifications required for this device were not already totally reached, especially in term of miniaturization and low power consumption allowing fully wireless solution compatible with drinking water supply market. Several impacts are expected: first of all, the technology developed reinforced drinking water supply safety and security. This solution provides new information to a better prevention in case of nuclear power incident (such Fukushima) or in case of intentional contaminations. Secondly, this device could be used also for radionuclides matter survey, for instance within hospital in order to control wastewater disposal (radiological scan service).

- 3. Development of modern analytical tools for contamination surface analysis

SECUREAU had a conceptual impact on any water system decontamination approach by taking the role of surfaces and their deposits into account. They act both as sink and source of the waterborne contaminants. Due to sorption processes, contaminants can be sequestered from the water phase. Due to desorption and deposit decomposition processes, sorbed contaminants can be mobilized in the water phase. Therefore, a new paradigm has to be implemented into practice: do not neglect the surfaces.

In practice, this means that surface and deposit samples are analysed and the efficacy of cleaning and decontamination measures can be verified in depth, providing better safety of such measures. The benefit for the society to take into account this new paradigm is the optimization of decontamination. Such approaches will make SMEs more competitive because they will be more effective. Moreover, optimizing the decontamination strategy and methods will benefit to the consumer's health.

In the event of an emergency crisis or terrorist attack resulting in the contamination of drinking water supplies, rapid response techniques are required that will allow any potential CBRN contamination events to be detected and quantified taking into account an even negligibly small risk of false-negative or false-positive results. In order to bring to an end to the crisis phase, analytical methods - again suitable for water and adapted to pipe wall analysis – should be carried out.

Analytical methods were developed or improved in the SECUREAU project for a number of typical CBRN pollutants. Some of them were developed for SECUREAU teams' own purposes while others could be used in the case of emergency contamination. All the methods used provided at least one acceptable choice for obtaining accurate results when determining the nature and / or concentration of a contaminant in water. However, determining the nature and / or concentration of a pollutant adsorbed onto the walls of the network was systematically found to be more difficult than for water sample analysis.

Quantitative and qualitative methods developed for chemical measurements were for the own purpose of the SECUREAU project. The analytical procedures developed were too complex for a rapid response in emergency situations. Nevertheless, quantifying the chemicals adsorbed onto deposits and biofilms is not a matter of emergency, but is needed for waste disposal purposes after an event is detected. In that sense, the developed methodologies were in accordance with the objectives of the project.

As some chemicals strongly attached to the deposits (depending on the nature of the contaminant and deposit), extraction techniques were required (e.g., hot acidic reflux with concentrated sulphuric acid). The extraction yields greatly depended on the contaminant / deposit pair, making any a priori predictions impossible. In addition to extraction, deposit-specific derivatization problems also were apparent with calcium-rich deposits.

Analyses of biological contaminants must allow low levels of bacterial pathogens to be detected, including non-spore forming and spore forming species, and provide a measure of their infectivity so that the public health risk can be accurately assessed. The use of highly specific peptide nucleic acid (PNA) probes in a fluorescence in situ hybridisation (FISH) assay, combined with cell elongation (direct viable count, DVC), allowed detection and differentiation of viable populations (including VBNC). In addition, real time PCR (qPCR) combined with propidium monoazide (PMA) could be successfully applied, with a detection limit of 1 cell ml-1. However, PMA-qPCR successfully differentiated live and dead populations in water. Consequently, one of several rapid methods developed for the detection of microbiological contaminants in water or biofilms/deposits can be chosen for routine or emergency monitoring of water distribution supplies. In particular, these are suitable for viable but non-culturable (VBNC) bacteria which can often be missed in routine measurements but could still pose a threat to public health. As such the rapid FISH and qPCR techniques are suitable for regular monitoring and rapid emergency detection of distribution supplies without waiting at least 24 hours for a culture recovery result, while the combined viability steps of including DVC or PMA permit the discrimination between live and dead bacteria following remediation treatment. The latter is essential when determining whether target pathogens have been successfully killed, rather than sub-lethally stressed to the VBNC state, to allow sign off and safe reuse of the distribution supply.

To confirm the intrusion or presence of radionuclides in a drinking water network, robust screening methods are required for detection and quantitation of alpha, beta and gamma emitting radionuclides, both in water and in biofilms / deposits. SECUREAU showed that liquid scintillation counting, coupled with spectral analysis and simple acid treatments of the biofilms and deposits which lead to dissolution / digestion / decolourisation, was effective. The technique enables the target radionuclides to be detected with limits of detection of 1 Bq/g or less with preparation and counting times taking less than three hours.

Therefore, these advances provide modern tools to ensure confidence among water companies and government regulators that they can reassure the general public that the drinking water and the distribution pipework have been successfully remediated following an emergency incident. This will impact on the public by protecting their health and reducing hospitalisation, morbidity and mortality costs. It will also reduce hysteria and save the expense and logistical difficulty of supplying bowser or bottled water to large sections of the population in a declared emergency area.

- 4. Combined decontamination strategies

Decontamination strategies needed first to define precisely the extent of contaminated zones. Then the research studies for localization of contamination and spread issues in drinking water distribution systems brought new knowledge with significant technical, public health and economic impacts.

One of the technical impact of SECUREAU work is related with the development of more reliable models able to predict the localization of contamination sources and the contaminated areas in real drinking water distribution systems, after accidental/deliberate contamination events. These models are incorporated in software tools that enable to obtain answers in very short periods of time. The new mathematical approaches developed under this project mitigate the impact concerning the public health because, in cases of contamination events, the populations that are really affected are more clearly identified. Moreover, predict the contaminated area will allow acting on the drinking water network before the contaminated water can be distributed in the tap water, safeguarding the health of the citizens.

As final consequence, the economical impact is also minimized because the necessary actions for solving the problems caused by the contaminated events (examples: the isolation of affected areas, the change of piping of affected areas) are defined in much more detail.

In this project practical applicability and safety (to staff and consumers) were most important criteria for selection of decontamination technologies, providing that the costs are within reasonable limits. The practical applicability is important because in most cases decontamination will be carried by water companies, and their staff is not necessary trained or equipped to address such situations.

As a result, the following combination of techniques were selected as the more appropriate regarding the nature of contaminants sorbed onto pipe walls. Indeed biofilms and deposits represent a sink for most of the contaminants and are more complicated to decontaminate than water treatment or waste.

- Fenton-like reactions with hydrogen peroxide taking advantages of iron and copper in the deposits were shown as very promising both for organic chemical and pathogen decontamination (even at quite neutral pH respectful of the system integrity).
- Sodium hydroxide followed by high chlorination allowed both effective spore desorption and disinfection.
- Radionuclides could be desorbed using sodium bicarbonate solution and flushed out of the system for safe storage and treatment.

- When necessary, first step allowing scale and strong deposit removal could be effectively carried out by pigging with Ice slugs or gravel in combination with water flushing.

- 5. Dissemination activities and exploitation of results

The important SECUREAU outcomes have been disseminated through 12 main actions, reported in the Deliverable 8.3.2 (public deliverable, available on the SECUREAU web site) and summarised hereinafter:

- European and national level conferences

The partners involved in SECUREAU have demonstrated a strong participation in conferences, both nationally and internationally by more than 42 oral presentations.
A strong effort has been done by SECUREAU partners to communicate efficiently, outside their country, mainly in Europe (9 countries, 37 conferences) even if four conferences were done outside Europe, one in Russia and three in USA.

- Poster presentation

A total of 21 posters were presented, mainly during two conferences: the 4th Water Contamination Emergencies (WCEC4) held in 2010 and the 5th Water Contamination Emergencies (WCEC5) held in 2012, both in Mulheim an der Ruhr (Germany). Three posters have been recognised as the best scientific poster presentation and received the Poster Award 2010 (in the WCEC4 conference, first price for Sandra Wilks and Bill Keevil, SOTON, UK and second price for Martin Strathmann, IWW, Germany) and the Poster Award 2012 (in the WCEC5 conference, for Florence Gosselin UL, F – Luis Miguel Madeira, Uporto, P – Talis Juhna, RTU, LV).

- Peer review and technical papers

By the end of 2012, 20 peer review and technical papers were submitted (11) or published (9) in journals or part of books.

- Workshop, seminars, technical meetings

Six partners have participated to oral presentations during 15 technical manifestations, both linked to specific WP or more generally concerned the overall SECUREAU project. Participation to such events permits both to diffuse information to selected public, as explained below through few examples:
- Meetings occurred at Ministère de l'écologie and Ministère de la Santé et du Développement Durable in France (two meetings in 2011 and two in 2012) to transfer specific, classified results to national French security agencies. One meeting was also done with the "détachement central interministériel d'intervention technique DCI-IT" in 2012. The same strategy was applied in Finland, with a specific seminar organised by STUK (December 2012) and one organised by SECUREAU in Germany, during the WCEC5 meeting;
- Results and contacts were also taken during meetings held by ASTEE (Association Scientifique et Technique pour l'Eau et l'Environnement, France), mainly with industrial partners;
- Presentation devoted mainly to SME was done, for example under the Pôle de compétitivité Alsace/Lorraine sur la qualité et la gestion de l'eau Hydreos (conference held by October 2012 in Metz, France);
- Cluster meetings with other European projects / coordinators also occurred, for example during meeting held in Stockholm (SRC'09 conference, Sept 2009), in Ispra (ERNCIP conference, Dec 2012), by direct contacts;

- Presentation to Technical committees was done during WCEC conferences (Mulheim, Germany) and ERNCIP conference (Ispra, Italy)...

Such actions were important (i) to feed the network of contacts and competences, (ii) to diffuse selected information to specific target identified (national authorities; SME; stakeholders; (iii) and to fulfil SECUREAU obligation written in the DoW.

- Participation in forum and organisation of workshop
(a) World forum for security and fire prevention (Essen, Germany)
SECUREAU was present at the "World forum for security and fire prevention" (Essen, Germany, Sept 2012) on the European Commission stand. It was the opportunity to have contacts with the other European projects on the EC stand (8 projects), the EC colleagues and industrial visitors, regarding technical SECUREAU solutions exposed. Many tools were presented at this occasion : sensors developed by VERI (Endetec) and IWW, coupon sentinels, models developed by UPORTO (slides were presented continuously on a 42" flat screen), Propella reactors...

Around 10 to 20 contacts or visits per day came in the SECUREAU area, including visit of M. Ralf Jäger (Minister of Municipal Affairs of Nordrhein-Westfalen), Paul Weissenberg (Deputy Director-General of the Enterprise and Industry Directorate-General), Marco Malacarne (Head of Unit for "Security Research and Development" in Directorate-General for Enterprise and Industry of the European Commission)... A 15 minutes conference on SECUREAU was also presented during this forum.

(b) 5th Water Contamination Emergencies: managing the threats
SECUREAU has co-organised with the WCEC5 the "5th Water Contamination Emergencies: managing the threats". During this conference, SECUREAU group has organised a closed workshop (requiring security clearance to hear confidential information). It was a very successful meeting: 150 participants from 26 countries. Around 25 participants were affiliated to Universities, around 25 to national or international health centres and around 100 to the industry. More than 50 people attended the session 8 "Security SECUREAU session" devoted only to the presentation of classified results. See Deliverable 8.5 (on-line on the SECUREAU web site) for more information regarding this workshop.

- Masters – PhD thesis – Post doctoral
A total of 13 young researchers were supported throughout SECUREAU budget (Five master thesis, Seven PhD and one post-doctoral formation).

- Website
The website (see http://www.SECUREAU.eu online) is a tool used for many purposes:
- Dissemination of non-confidential information (scientific objectives, expected impacts, structure of the consortium, public deliverables, main events with SECUREAU members participation),
- Document storage area devoted only for SECUREAU members (login + password needed) for scientific documents (e.g. deliverables not public, meeting reports, DoW, slides presented during the meetings...) as well as administrative documents (e.g. financial guide, audit and certification guidelines, project reporting guide, Grant agreement, consortium agreement...);
- Dissemination of scientific communications, conference and abstracts of publications. The scientific community is the main target of this action.
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- Industrial and final products
Two main items has been developed or improved during SECUREAU: sensors and mathematical models and are reported above.

- Press releases, interviews
Few actions related to press release or interview occurred within SECUREAU project.
An article entitled " SECUREAU : faire face à une attaque terroriste sur les réseaux d'eau potable " was published (see http://eureka.lorraine.eu/jahia/Jahia/cache/bypass/pid/1968?actu=20643 online) on the French Lorraine web site. Two video interviews were realised, one for the European Commission (2009) followed by a DVD freely distributed (Video illustration on the Security Research Theme) and one done by the Latvian National TV Chanel, dedicated in general to RTU research and in particular to SECUREAU project.
Last, SECUREAU is also referenced (see http://ticri.inpl-nancy.fr/wicri-lor.fr/index.php/SECUREAU online) on the Wicri/Lorraine web site, which refers all research activity in which one partner (industrial, research organisation, SME...) came from Lorraine (France).

- Methodological guide for end-users
The Methodological guide for end users (Deliverable 8.4, public document available on the SECUREAU web site) aims to highlight, along with Deliverable 6.8 (Decision tool Suitable for Assessment to Approval of Successful decontamination), some of the methods selected in SECUREAU for pre-crisis, crisis and post-crisis phases. Their main interests or novelty and their limits are also discussed in the document.

- Classified results
Special attention has been paid to the secured dissemination of classified results. First, preconisations given both by the European Commission and by the national security agencies were strictly respected: no classified information on the Web site, even on a restricted area; no diffusion of this information on the EC Participant Portal)... Specific meetings were organised to diffuse this classified information, already described above in the Chapter 4: Workshop, seminars, technical meetings and chapter 5: Forum participation and workshop organisation.

- Deliverables produced
The Deliverables produced are sorted by (i) 8 public deliverables (available freely on the SECUREAU web site), (ii) 35 restricted and confidential deliverables, i.e. not public documents, not available, and (iii) 4 classified deliverables. Systematically these documents were reviewed by the steering committee and by the dual use advisory board for classified deliverables. 70 % of these documents were also revised by external experts during two scientific reviews (one held for 2009-2010 period and the second one for the 2011 period).

List of Websites:

http://www.secureau.eu