AGENZIA NAZIONALE PER LA PROTEZIONE DELL'AMBIENTE

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'MODELLING ENVIRONMENTAL PROCESSES'

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Contents

- 1. Preface
- 2. Introduction
- 3. Literature review
- 4. Development of DAGES Code, a Monte Carlo Algorithm to evaluate dose rate in air from gamma emitters in soil
- 5. Application of DAGES Code
- 6. References

1. Preface

The present document reports the activities carried out in the first year of work in the framework of the scientific collaboration program: *Modelling Environmental Processes*. This program is developed in the frame of the agreement between the National Environmental Protection Agency, Italy (ANPA) and the National University of San Luis, Argentina (UNSL).

This program, which has a total duration of three years, has as its main aim the development and application of a general system based in a Monte Carlo algorithm (DAGES code) to evaluate the external photon exposure from surface contamination on the ground.

This first partial report includes a summary of the principal methodologies applied to determine the external exposure. The flux diagram and the physical principle used in the development of DAGES Code are presented and some preliminary applications of the program are shown.

The data contained in the present summary have been provided by the National Environmental Protection Agency (ANPA) and the software used by the National University of San Luis (UNSL).

Reproduction of the data and the methods contained in this report is authorised provided the source is acknowledged.

2. Introduction

External gamma irradiation from surface contamination on the ground can be one important source of exposure to radionuclides which have been released to the environment.

The calculus of this irradiation is extremely complex due to innumerable environmental factors, which have an important influence in the photon fluence on a detector situated in air above contaminated soil. Some of the most important factors to consider are the following: the soil vertical distribution of radionuclides, their mobility, the ground roughness, the soil bulk density and its eventual change with depth. The energy of the source determines the way of interaction between the radiation and the medium, but the previous factors are important to determine the energy and the angular distribution of the gamma radiation on the detector.

If these factors are not taken into account, the results of the determination of the external exposition (or absorbed dose or equivalent dose) could conduce to not sufficiently accurate results. For example, the U.S. Nuclear Regulatory Commission (USNRC77) uses the approximation that the radionuclides deposited on the ground remain on the ground surface until removal by radioactive decay. This assumption leads to overestimates of external dose to exposed individuals because the activity will normally be transported into the soil and the resulting layer of soil between the source and the receptor locations will provide an important shielding. Uncertainties related to ground roughness and laterally non-uniform activity distribution should also be taken into account to determine the dose contribution with more precision.

Bunzl *et al.* (1996) have shown the importance that migration of radiocaesium into the soil has on the attenuation of the gamma radiation by overlaying soil layers.

The determination of the external irradiation implicates the previous determination of the spectral distribution of the photon energy fluence in the detector. On the other hand, knowledge of the properties of the photon field from sources situated in ground is essential when applying in situ high-resolution gamma spectrometry to measure the activity of individual gamma-emitting radionuclides in the environment and estimate their contribution to the absorbed dose rate. The calculation of the photon fluence depends on the energy of the source and the atomic composition and density of the medium. For example for photon energies above 100 keV Compton scattering effects is dominant and the mass attenuation coefficients vary slowly with the atomic number of the elements. The exact elemental composition of the soil is therefore not so important. For photon energies below 100 keV, the photoelectric interaction can become important in soils containing elements of higher atomic number. Beck *et al.* (1972) calculated mass attenuation coefficient for soil with the following composition: 67,5% of SiO₂, 13,5% of Al₂O₃, 10% H₂O and 4,5% of CO₂. Changes in this composition can be important when the physics process involved in the interaction of the radiation with the medium are considered. Photoelectric effect is less important than the Compton effect at 60 keV for aluminium (Z =13), but becomes comparable for silicon (Z=14) and predominates for elements with higher atomic number.

The present report includes a summary of the principal techniques (analytical and of simulation) for converting measure data of radionuclides concentration in soil into external exposition or absorbed dose. It also introduces a new procedure based in a Monte Carlo algorithm with this end. Finally, applications of the results obtained to real situations are reported.

3. Literature review

The external exposure from residual photon emitters in soil has been studied extensively in the past. Starting from this external irradiation, the effective dose equivalent has been calculated, usually through the determination of dose-rate conversion factors. The calculations are based on the assumption that the receptor is located a 1 m above the contaminated ground.

Beck and de Planque (1968) and Beck (1972) solve the soil-air photon transport using the polynomial expansion matrix equation to calculate the exposure rate ought to gamma emitters in soil. The dose rate was calculated for a wide range of energy for radionuclides commonly found in the natural environment.

Lowder *et al.* (1964) and Beck *et al.* (1964) introduce the peak area method, a technique for converting measured data from in situ gamma spectrometry into absorbed dose. During the Chernobyl accident, the method was utilised to determine dose rate and activity from deposited fission products (Moberg (1991), Finck (1992)). Today, this method is often incorporated into emergency plans as a rapid procedure for assessing fallout from nuclear accidents. To use peak area method, it is necessary to know the relations between the primary photon fluence rate and the activity of the source per unit volume, mass or area. Even if some equations cannot be analytically solved, calculation of the primary fluence from a source with specific geometry and activity is often straightforward. However, to calculate the scattered fluence rate is a more complex problem.

Kocher and Sjoreen (1985) determine the dose-rate conversion factors. Their calculations are based on the point-kernel integration method and assume that the source concentration at any depth in soil is uniform over an infinite surface parallel to the ground plane. The dose-rate factor is applied to environmental dose assessments by means of the general equation:

$$H(t) = \chi(t) x DRF$$
(1)

where H is the external dose rate at time t, χ is the source concentration at the location of the exposed individual, and DRF is the dose-rate factor. DRF depends on the height of the receptor location above ground (this height is usually assumed as 1 m). However for photon emitter in soil, DRF depends on the depth of the source in soil but usually is

insensitive to the height of the receptor location above ground for heights of about 10 m or less.

Dose-rate factors in air at a height of 1 m above ground are tabulated for discrete photon energies between 0.01 and 10 MeV and for source depths in soil between 0 and 300 cm. These factors were determined for sources distributed in a slab of finite thickness and sources which are exponentially distributed with depth. For example Velasco *et al.* (1993) used DRF to evaluate the dose rate in air ought to ¹³⁷Cs derived of the Chernobyl accident deposited in Italian soils and distributed exponentially with depth.

The methods used to calculate the dose-rate factors involve idealized assumptions concerning vertical and lateral distributions of sources in soil and the extent of shielding provided by the air above ground. Undoubtedly these assumptions are not strictly valid for most realistic exposure situations.

Finck (1994) describes the theory needed to obtain the primary photon fluence for some typical source distribution in the ground. He considers radionuclides deposited on agricultural land which can be mixed into the top layer of the soil by cultivation procedures. This could produce uniform slab sources with thickness depending on the depth of ploughing. Exponentially decreasing source are also contemplated in this analysis.



Figure 1. Geometry used in deriving the photon fluence from radionuclides in the ground. The contribution of primary photons at height h above ground surface from a volume element $dV=dr.dR.d\rho$ at depth z is calculated using eqn. 2.

The geometry used by Finck for calculations of primary photon fluence is based on two semi-infinite volumes of soil and air separated by an infinite plane soil surface (Fig. 2). A photon source element is contained in a volume element $dV = dr dR d\rho$ of soil at depth z below the soil surface. If the photon source is represented by $S(z,r,\eta)$ photons emitted per unit volume of soil as a function of depth z, lateral distance r and azimuthal angle η , then the primary photon fluence at the position of a hypothetical detector at height h above ground and at distance R from this volume element is:

$$d\Phi = \frac{S(z,r,\eta)dV \exp(-\mu_s (R-h \sec\theta) - \mu_a (h \sec\theta))}{4\pi R^2}$$
(2)

where θ is the angle between the direction to the volume element and the normal to the airsoil interface, μ_s and μ_a are the mass attenuation coefficients for soil and air respectively. μ_s is usually calculated for soil of composition: 67.5 % SiO₂, 13.5 % Al₂O₃, 10.0 % H₂O, 4.5 % Fe₂O₃ and 4.5 % CO₂ (Beck *et al.* 1972). Figure 3 shows the soil mass attenuation as function of the source energy.



Figure 2. Mass attenuation coefficient of soil as function of the source energy

The total primary photon fluence can be obtained integrating eqn. (2) over entire photon source in the soil:

$$\Phi = \int_0^{\pi/2} \int_{h\sec\theta}^{\infty} \int_0^{2\pi} \frac{S(z,r,\eta)r\exp(-\mu_s(R-h\sec\theta) - \mu_a(h\sec\theta))}{4\pi R} d\eta \, dR \, d\theta$$
(3)

When the source distribution in soil is simple (as uniform or exponentially decrease with depth), the solution of the integral (3) can be easily calculated, but for complicated source distribution, this integral cannot be solved analytically.

For example if we assume a uniform source distribution S with infinite depth and infinite lateral extend in the ground, the primary photon fluence is

$$\Phi_{uniform} = \frac{S/\rho_s}{2\mu_s} E_2(\mu_a h)$$
(4)

where $E_2(\mu_a h)$ is the exponential integral which is defined, in the general case, as:

$$E_{n}(x) = x^{n-1} \int_{x}^{\infty} \frac{e^{-t}}{t} dt$$
(5)

In Figure 3, the photon fluence rate in air at one metre above ground from uniformly distributed source in the ground is plotted as a function of primary photon energy. The emission rate is taken as one photon per kg s. There is a distinct rise in the primary fluence with increasing energy, due mainly to the decreasing attenuation coefficient of the soil.



Figure 3. Photon fluence rate in air as a function of primary photon energy from soil sources of infinite lateral extend and different mass thicknesses below the ground surface. Curves correspond to an emission rate of one photon per kg s (from Finck (1994)).

Figure 4 shows the primary photon fluence in air at heights between 0.5 and 500 m from a photon source distributed uniformly in the soil. The photon fluence at the ground surface is $S/2\mu_s$ and declines with increasing height due to attenuation in the air. The energy dependence of the primary fluence is more accentuated at greater heights. For example, at 100 m, 35 % of the 2615 keV primary photon fluence is still present but only 6 % of the 130 keV fluence.



Relative primary photon fluence

Figure 4. Height dependence of the primary photon fluence in air above a photon source distributed uniformely in the soil. The fluence is normalised to the photon fluence one metre above the infinite plane air-soil interface. The air density is 1.24 kg/m^3 (from Finck (1994)).

In the particular case of an superficial infinite plane source, the total primary photon fluence at height h above ground can be written as following:

$$\Phi_{surface} = \frac{S}{2} E_1(\mu_a h) \tag{6}$$

In the last equation E_1 is the exponential integral of first order. The primary photon fluence rate in air one metre above an infinite plane source for an emission rate of one photon per m² s is shown graphically in Fig. 3 (bottom curve). The energy dependence of the primary fluence at one metre is small in comparison with sources uniformly distributed with depth.

From eqn. (4) is possible derive an expression for the primary fluence in air at height h above a slab source (thickness z) with uniform concentration, in the top layer of the ground (this is a typical distribution of radionuclides in agricultural land where the top layer of the soil is mixed by cultivation procedures). This fluence can be written as the difference between the fluences of two uniform sources, one starting at zero depth and the other at depth z below the soil surface:

$$\Phi_{slab} = \frac{S}{2\mu_s} \left[E_2(\mu_a h) - E_2(\mu_s z + \mu_a h) \right]$$
(7)

In Figure 4 the primary fluence rate in air one metre above ground is shown as a function of photon energy for different thicknesses of the slab source.

An exponential decreasing function has been often used to describe the vertical distribution of radionuclides in soil (Velasco *et al.* (1993), (1997)). In this case the source concentration as a function of depth in soil in a determined time is given by:

$$S(z) = S(0) \exp(-\alpha z) \tag{8}$$

where S(0) is the concentration at the ground surface, α (the alpha-factor of the distribution) is the reciprocal of the relaxation length and it depends on the radionuclide, the soil type and time after deposition.

Exponential depth distribution can be thought as an infinite number of plane source distributions with exponentially decreasing activity per unit area. The photon fluence rate from one of these plane distribution at depth z can be obtained from eqn. (6):

$$\Phi_{plane} = \frac{S(z)}{2} E_1(\mu_a h + \mu_s z)$$
(9)

The total primary photon fluence is obtained by integration over all the plane sources:

$$\Phi_{\exp} = \frac{1}{2} \int_0^\infty S(0) \exp(-\alpha z) E_1(\mu_a h + \mu_s z) dz$$
(10)

Operating adequately it is possible to obtain the following expression for the total primary photon fluence:

$$\Phi_{\exp} = \frac{S(0) / \rho_s}{2 \mu_s / \rho_s} \exp(\frac{(\alpha / \rho_s) \mu_s h}{\mu_s / \rho_s}) \left[F_1(\infty, \frac{-\alpha / \rho_s}{\mu_s / \rho_s}) - F_1(\mu_s h, \frac{-\alpha / \rho_s}{\mu_s / \rho_s}) \right]$$
(11)

In this equation the depth has been expressed in mass per unit area ($\rho_s z$) and the function $F_n(t,\alpha)$ is defined as:

$$F_{n}(t,\alpha) = \int_{0}^{t} \exp(\alpha y) E_{n}(y) dy$$
(12)

In eqn.(11) F_1 can be obtained from curves in textbooks on shielding. Figure 5 shows the energy dependence of the primary photon fluence rate in air one metre above ground from soil sources distributed exponentially with depth.



Figure 5. Primary photon fluence rate in air as function of source energy from radionuclides in soil distributed exponentially with depth. Curves have been obtained for different values of the alpha-factor of the distribution (from Finck (1994)).

Simultaneously, other investigators have used Monte Carlo simulations to calculate effective dose equivalent (O'Brien and Sanna (1976), O'Brien (1978), Jacob *et al.*(1986),

Chen (1991)). In these studies the photon transport in the soil/air interface was simulated and the resultant organ doses to an anthropomorphic phantom was determined. The calculations of Jacob *et al.* (1986) were based on discrete photon source energies ranging from 0.015 to 6 MeV. The results obtained differed significantly from those of Kocher and Sjoreen (1985). These discrepancies are attributed to the assumption of the isotropic beams in Kocher's calculation. While the simple source geometry used by Kocher and Sjoreen (1985) and other authors can often be used with reasonable accuracy, however in some cases, additional complicating factors, such as the effects of ground roughness, uneven source distribution and varying soil composition will introduce additional uncertainties into the calculus of dose rate.

Huddleston *et al.* (1965) have developed a detailed analysis of the ground roughness effects on the energy and angular distribution ought to gamma emitter from fallout. They investigated three type of terrains: 1) a flat dry-lake bed, 2) a ploughed field with a known degree of roughness and 3) a typical wild desert. To confront theoretical predictions and experimental determinations, dose vs. height measurements were made up to a height of 40 ft.

A Monte Carlo algorithm was developed by Chen (1991) to perform the photon transport calculation for the soil/air configuration, in which the soil constituents were assumed to be similar to those of the earth's crust. The calculations are based on the ICRP's concept of effective dose equivalent and the conversion factors published by the ICRP (International Commission on Radiation Protection). Using these calculations is more advantageous for two reasons. First, the ICRP's suggested conversion factors are used for the effective dose equivalent. These conversion factors were not available when the calculations of Jacob *et al.* (1986) and Kocher and Sjoreen (1985) were formulated. Second, a Monte Carlo algorithm was tailored to calculate the effective dose equivalent for the soil/air configuration without having to include an anthropomorphic phantom.

In the model proposed by Chen (1991), the photon source is assumed to be distributed uniformly in the soil from the ground surface to a contamination depth, beyond which the soil is uncontaminated (to a total depth). For practical purposes, depths of source up to five mean-free-path lengths of the source photon in the soil and depths of air up to 500 m are considered. To calculate the effective dose-equivalent responses, a Monte Carlo algorithm was developed to track the transport of photons in the soil/air medium as illustrated in Figure 6. Source photons were randomly selected from the contaminated soil

zone and their subsequent interactions determined by the probability of occurrence via photoelectric, Compton scattering, and pair-production processes.



Ts =Total Soil Thickness (5mfp)

Figure 6. Source/receptor configuration for calculation of dose responses from distributed photon sources in soil.

Clouvas *et al.* (2000) calculate the dose rate conversion factors (absorbed dose rate in air per unit activity per unit of soil mass) 1 m above ground for photon emitters of natural radionuclides uniformly distributed in the soil. In this study are confronted three Monte Carlo codes: the MNCP of Los Alamos (Briesmeister, 1993), the GEANT developed by CERN (GEANT, 1993) and the MC (Clouvas, 1998). In Figure 7 is showed a comparison of the primary photon fluence in air at 1 m above ground, ought to source uniformly distributed in the ground, obtained by the three Monte Carlo Codes and the numerical solutions of eqn. (4) (Beck *et al.* 1972). The comparison between the results indicates that all Monte Carlo codes calculate very well the unscattered radiation.



Figure 7. Primary photon fluence in air from source uniformly distributed in ground as function of the source energy. Results obtained by the Monte Carlo codes and numerical solution

The dose rate conversion factor, defined as the absorbed dose rate in air per unit activity per unit of soil mass, for a photon emitter of energy E_0 uniformly distributed in the ground can be calculated as (Clouvas *et al.* 2000):

$$\dot{D} = \sum_{i=1}^{n} \frac{\mu_a}{\rho} E_i \Phi_i(E_i) \tag{13}$$

with E_i is the average energy of band i, $\Phi_i(E_i)$ is the photon fluence per unit activity per unit of soil mass in energy band i and μ_a/ρ is the mass absorption coefficient for air at energy band i. The summation starts at the energy band i = 1 (10-20 keV) proceeds with a step of 10 keV and ends at the energy band i =n containing the photon energy of E₀. Figure 8 shows the dose rate conversion factor calculated by three Monte Carlo codes for different photon energies. In the same graphic are shown the results obtained from the work of Kocher and Sjoreen (1985) and Chen (1991).



Figure 8. Dose rate conversion factors obtained by the Monte Carlo codes and those deduced from Kocher and Chen as function of the source energy

4. Development of DAGES Code, a Monte Carlo Algorithm to evaluate dose rate in air from gamma emitters in soil

A Monte Carlo algorithm was used in order to study the behaviour of subsets of photons, starting from the initial emission from the source, up to the end of its trajectory. Every one of the possible interactions of these photons with soil was computed. In this preliminary study the source is ¹³⁷Cs.

Due to energy intensity of ¹³⁷Cs emissions, only two interaction processes of radiation with matters were accounted. The photoelectric effect, by which gamma rays deliver all their energy to an orbital electron and the Compton effect, by which only part of gamma energy is delivered when interception with an electron and pairs of electron-positron are generated when the gamma ray is absorbed on the vicinity of an atom nucleus. In case the photon verifies a photoelectric interaction, it stops its trajectory. However, when a Compton scattering occurs, the new trajectory and energy of the photon should be calculated.

To determine which type of interaction will take place it is necessary to compare the probabilities of photoelectric and Compton interaction.

The procedure was developed to obtain the external dose in a detector situated 1 m over contaminated ground. Two different types of distribution for the source in soil were considered: a) Profiles, which are uniformly, distributed with depth, and b) Profiles which are exponentially distributed with depth. Figure 2 shows a flow diagram of the used algorithm, called DAGES code.

The steps followed in the simulation are the following:

a) Emission

The photon initial position (source position) is given for his coordinates (z_0 , φ_0 , θ_0), where z_0 is the soil depth (z = 0 is the soil upper surface), φ_0 and θ_0 are the azimuthal and zenithal angles respectively. The depth z_0 was randomly selected from the contaminated soil zone. φ_0 and θ_0 were randomly selected from the interval [0, 2π] the first and [0, π] the second.



Figure 9. Flow diagram of DAGES code

b) Photon track length to the subsequent interaction

The length of the path following by the photon to the next interaction with the medium was determined using the relation (Shultis (1996)):

$$l = -\frac{ln\delta}{\mu} \tag{14}$$

where:

l (cm): track length.

 δ : random variable in the interval [0,1].

 μ (cm⁻¹): energy linear attenuation coefficient.

c) Photon-medium interaction form

The form of the interaction of the photon with the medium is selected having in account the probability of occurrence via photoelectric or Compton scattering. Pair production processes were discarded because of the source energy considered. Angles of scattering by Compton interactions were obtained from the angular distribution described by the Klein-Nishina formula (Evans (1955)).

d) Calculus of the absorbed dose in the receptor

If the photon reaches to the receptor, the absorbed energy by the air mass content in the receptor is calculated according to the procedure proposed by Thomas (1983). The dose absorbed results from the sum of individual contribution of each photon arriving to the receptor, according to the following equation:

$$D = \sum_{i=1}^{N} \Phi(E_i) \cdot \left(\frac{\mu_{ab}(E_i)}{\rho}\right)_{air} \cdot E_i$$
(15)

where

D(Gy): is the absorbed dose in air $\Phi(E_i)$ (Joule m⁻²): Photon fluence with energy E_i $\left(\frac{\mu_{ab}}{\rho}\right)_{air}$ (m² gr⁻¹): Energy absorption coefficient in air for energy E_i

Comparison of DAGES model with previous studies

Figure 10 shows the results obtained from DAGES Code for the dose rate in air due to a slab of finite thickness in soil with a uniform ¹³⁷Cs concentration. The values obtained from the analytical procedure proposed by Kocher (1985) and the simulation carried out by Chen (1991) are comparatively shown.



Figure 10. Dose rate in air due to a uniform distribution of the source into ground

5. Application of DAGES Model

A first application of DAGES model was carried out using data of radiocesium soil concentration collected within the framework of Project ECP-5: "Behaviour of radionuclides in natural and semi-natural environments", supported by European Commission, with the participation of research groups from CIS countries (Belarus, the Russian Federation and Ukraine) and from Western Europe, (Belli and Tikhomirov, 1996). Samples of soil were taken both inside and outside the 30 km Chernobyl zone. These sites include the most typical types of meadows in the CIS countries. The profile of the radionuclides in soil has been fitted by an exponential function (Velasco et al. (1997)).

Table 1 shows the site sampling, the ¹³⁷Cs deposition, the alpha-factor of the distribution and the dose rate *in situ* measured a calculated using DAGES model.

Sampling	¹³⁷ Cs Dep.	Alpha-factor	Measured	Simulated	Difference
Site	(Bq cm ⁻²)	(cm ⁻¹)	Dose Rate	Dose Rate	(%)
			(Gy/h)	Gy/h)	
Zapolje – 0	48	0.595	6,0 x 10 ⁻⁷	7.2 x 10 ⁻⁷	20
Zapolje – 1	58	0.481	6.6 x 10 ⁻⁷	6.0 x10 ⁻⁷	10
Zapolje – 2	53	0.553	6.2 x 10 ⁻⁷	8.0 x10 ⁻⁷	30
Zapolje – 3	54	0.552	6.2 x 10 ⁻⁷	8.1 x10 ⁻⁷	31
Zapolje – 4	49	0.590	6.0 x 10 ⁻⁷	8.0 x10 ⁻⁷	33

 Table 1: Dose rates in air from 137Cs in different sampling sites

Dose rate has also been calculated for two experimental sites located in Tarvisio (Italy). Soil samples were taken to a depth of 40 cm and 32 cm respectively, dividing the monolith in horizontal soil layer with different thicknesses. Dose rates were performed using DAGES model considering slab with uniform concentration according to the measured values. Tables 2 and 3 summarised for each layer, the bulk density measured (and considered in the simulation) and the ¹³⁷Cs concentration. The value of dose rate calculated in each case is also given.

Thickness of soil	Bulk Density (g cm ⁻³)	¹³⁷ Cs concentration			
layer (cm)		Bq g ⁻¹			
0-2.5	1.73x10 ⁻¹	1.38			
2.5 - 7.5	1.69x10 ⁻¹	3.7x10 ⁻¹			
7.5 – 17.5	2.1x10 ⁻¹	5.99x10 ⁻²			
17.5 – 29	3.51x10 ⁻¹	1.68x10 ⁻²			
29-40	5.07x10 ⁻¹	4.87×10^{-3}			
Dose rate (Gy h^{-1}): (9.7 ± 1.5)x10 ⁻⁹					

Table 2: Sampling station AP of Tarvisio, Italy

Thickness of soil	Bulk Density (g cm ⁻³)	¹³⁷ Cs concentration			
layer (cm)		Bq g ⁻¹			
0-2.5	8.53x10 ⁻²	7.15x10 ⁻¹			
2.5 - 7.5	1.24×10^{-1}	7.59x10 ⁻¹			
7.5 – 17.5	2.40×10^{-1}	6.82x10 ⁻²			
17.5 – 32	2.25×10^{-1}	1.01x10 ⁻²			
Dose rate (Gy h^{-1}): (6.3 ± 1.0)x10 ⁻⁹					

Table 3: Sampling station AU of Tarvisio, Italy

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