Verification of the dispersion model by airborne carbon ^{14}C

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Abstract. This paper provides insight in the verification of the Lagrangean dispersion model for dose calculation in the environment. The verification method was based on the measurement of the airborne carbon ^{14}C concentration which can be slightly increased close to the nuclear power plant. The results proved that this method is sensitive enough and that the sensitivity analysis can be used for model verification or for identification of possible improvements of the used meteorological data.

KEYWORDS: air pollution dispersion model; airborne ^{14}C; nuclear power plant; environmental monitoring; airborne ^{14}C dose assessment.

1. Introduction

The Lagrangean model is used at Krško Nuclear Power Plant (NPP) for calculation of dispersion coefficients and dose in the environment. To show compliance with the authorized dose limits it is required to present a realistic calculation of the dose to the public. This is a numerical model designed to calculate air pollution dispersion in the area of 25km x 25km. The model uses on-line local meteorological measurements.
The same model was already verified [1] for another location around a coal- fired power plant based on emission and environmental measurements of SO_2. Krško NPP is placed near the Sava River in a semi-opened basin surrounded by several hills. The region is characterized by low winds and frequent thermal inversions.
This paper presents a verification of the short range dispersion model based on the fact that the airborne carbon ^{14}C concentration can be slightly increased close to the nuclear power plant. Other radioactive effluents are not detectable in the environment and carbon ^{14}C measurements are accurate enough to detect small deviations from natural ^{14}C levels and to compare them with the calculated concentration based on ^{14}C effluents. The most of airborne ^{14}C is released during the refuelling outage. Within the pre-selected period of ten days, increased effluents of ^{14}C in the form of CO_2 were sampled from the plant ventilation. The average atmospheric dispersion parameters were calculated for two locations in the environment where CO_2 sampling plates were installed.
Increased ^{14}C activities were detected at both locations close to the plant for this relatively short time period.

2. Production of carbon ^{14}C in a nuclear reactor

The activity of ^{14}C in the source term of nuclear reactor is not important comparable to the activities of the other sources such as noble gases. During recent years nuclear fuel elements in commercial reactors have recorded of very low leakage. Carbon ^{14}C is produced in the reactor coolant. The main contribution to the formation of ^{14}C in light water reactors is the nuclear reaction ^{17}O (n, α) ^{14}C.

It is reported that about 90% of the total ^{14}C production outside the nuclear fuel is due to ^{17}O reaction [2]. The other contributors are the reaction ^{14}N (n, p) ^{14}C and there might also be a release from ^{14}C inventories in the fuel, or the (n, γ) reaction with some ^{13}C content of the coolant impurities.

The production rate due to ^{17}O reaction is proportional to the neutron flux. The release of ^{14}C from the coolant is higher during the refuelling period when the primary system is open and the air over the pools with primary water and nuclear fuel is ventilated to the atmosphere.

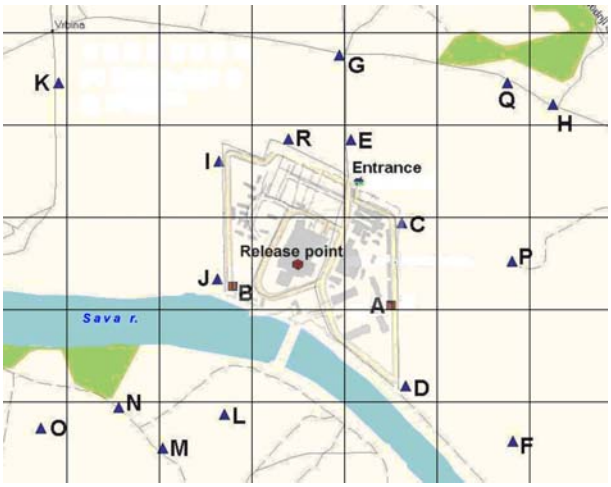
3. Collection and measurement of ^{14}C

The sample for determination of ^{14}C in the effluents is taken from the common plant vent header. Sampling is performed by distribution of gases from plant vent into two equal separated parallel streams. The flow of 10 l/h is maintained by two diaphragm pumps. The tube from the first stream is connected to the first column, where CO_2 is absorbed in NaOH solution as $NaHCO_3$. Remains of gases from the plant vent are lead through the oven, where subsistent carbo-hydrates are transformed into CO_2 at the temperature of 400 °C and with the presence of palladium as catalyst. CO_2 is then absorbed in NaOH solution (as $NaHCO_3$) in the second column. Samples are taken every two weeks. The plant presents the results usually in monthly effluents report.

In the environment, atmospheric CO_2 was collected by using saturated NaOH over a certain period of time, usually 2 months. Therefore, by this static sampling method we collected an integrated sample of atmospheric CO_2 and its ^{14}C activity corresponds to the mean atmospheric ^{14}C activity in this period. CO_2 chemically reacts with NaOH and Na_2CO_3 is formed, which is then in the laboratory hydrolyzed and the obtained CO_2 is used for benzene synthesis. Measurement is performed in the LSC Quantulus and the result of ^{14}C activity has been corrected for isotopic fractionation occurring during the sampling procedure.

Two locations were selected, marked as A and B in the Figure 1, close to the release point of the plant ventilation system, where useful results of ^{14}C were expected. Previous measurements showed that the atmospheric ^{14}C activity at the location B was always slightly higher than that at the location A. This difference corresponded to the prevailing wind direction. Based on predetermined spatial distribution of ^{14}C activities in biological samples around the plant, it was clear that important results could be obtained only within a few hundred meters distance from the plant [3]. This close distance was not very convenient for testing the dispersion model but enabled to detect higher activities in a shorter time interval.

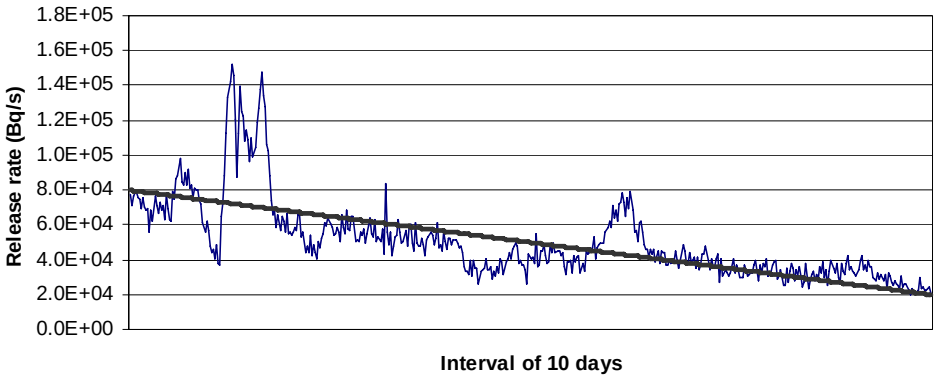
Figure 1: Sampling locations around Krško NPP for ^{14}C. Locations A and B are for atmospheric CO_2. Locations C – R for biological samples. The grid indicates cells of 250 m x 250 m.



4. Radiation monitors

Radiation monitors for noble gases were used as a reference for the gas release rate from spent fuel pool, reactor cavity and from other areas. A record history of the activity concentration and ventilation flow rates was available to enable a rough estimation of overall release rate in the period of ten days. It was assumed that CO_2 might have the same release mechanism. Figure 2 presents two assumptions of release rates: the basic one is linear and extrapolated from the measurements of ^{14}C average concentration in the effluent from the plant vent; the reference one is derived from the noble gas monitors record.

Figure 2: Release rate of ^{14}C is presented by the bold line. This release rate is derived from the average release rate of ^{14}C (50 kBq/s) in the time interval of 10 days. The estimation of the release rate of noble gases is presented by the thin line based on the effluent monitors.



5. Atmospheric dispersion calculation

The atmospheric dispersion is calculated by the modules providing an accurate view of the propagation of effluents in the atmosphere. The numerical Lagrangean particle model uses meteorological data resulting from 3D wind model. Meteorological data are taken as half an hour averaged on-line measurements from one SODAR and a 70 m meteorological tower at the plant site, and four ground level meteorological stations around the plant. They are connected to the local Environmental Information System. All the stations are within the 25 km x 25 km modelling domain. The spatial grid resolution is 250 m horizontally.

The wind module is a mass-consistent three dimensional wind field model for reconstruction of the wind field over a complex terrain using the MINERVE6 code [4]. This code is mainly based on an objective analysis scheme in terrain-following (sigma) co-ordinates, used to perform an initial interpolation of sparse ground-level and upper-air SODAR data available over the computational domain, and a final divergence-free adjustment. This method is particularly suitable for real-time applications, assuring a good compromise between the quality of the generated fields and the response time, in the presence of a well-designed measuring network. Within the MINERVE6 code, temperature fields are also computed via a 3D Cressman analysis scheme, again based on ground-level and upper-air data available from the local network.

Three dimensional wind and temperature field are then passed to the SURFPRO code, suitable for calculating turbulence scale parameters based on standard parameterizations [5], taking into account the horizontal terrain inhomogeneities through the use of a land-use horizontal field.

All the information coming from the meteorological codes is then passed to SPRAY3 stochastic Lagrangean particle dispersion model [6]. This code is based on the formulation developed by allowing simulation of the dispersion in non-stationary conditions from continuous or discontinuous sources of whatever structure, taking into account complex conditions such as the presence of complex terrain and the related meteorological inhomogeneities [7]. The entire modelling system is routinely invoked on a half hour basis using the currently available data, in order to produce a field of dilution coefficients from the power plant emission, starting from the conditions generated at the end of the previous run.

6. Dose calculation module

Dose calculation module [8] enables two different functions – a real time dose projection in case of an emergency situation and more accurate analysis of dose calculation for a longer period of very low level emissions to assess compliance to site dose constraint and to present as much as possible realistic results of atmospheric dispersion calculation.

For the dose projection in case of an accident, the radionuclide activities are derived first based on the results of the ORIGEN [9] computer code for several fuel burn-ups. In fact, instantaneous radioactivities are calculated for the selected set of radionuclides which are of the most radiological importance of the release source term. The amount of radioactivity is projected by a quick assessment of reactor core damage. The assessment module has some of the data on-line, but the assessment is accomplished by manual verification and selection based on a programmed algorithm.

The uncertainties of dose calculation related to the source term are not so important since the activity measurements and calculation are more accurate than atmospheric dispersion modelling. To get a reliable dose calculation result, it is important to have a reliable dispersion coefficient field at the local scale.

7. Verification of the dispersion model

Verification of the dispersion model was possible for only two locations, i.e. the points A and B within the site boundary. The collection of the samples of atmospheric CO_2 corresponded to the plant vent sampling interval of 10 days. Meteorological data were averaged over half an hour interval and the calculated concentration of ^{14}C in the environment was provided for each half an hour interval. The usual sampling interval is one month but in this case the shorter interval was selected since the most of ^{14}C is released during the refuelling period. Within these 10 days, the nuclear fuel was already transferred from the reactor to the spent fuel pool.

The measurement of ^{14}C in the atmospheric CO_2 gave the following result of average concentration in the time interval of 10 days:
a) Location A east of the release point: 151.1±0.9 pMC or 63.5 mBq/m^3.
b) Location B west of the release point: 275.4±1.5 pMC or 116 mBq/m^3.
It was supposed that atmospheric volumetric concentration of CO_2 was 347 ppm. The reference value was measured in Zagreb as 100.7 ±0.5 pMC.

The release rate of ^{14}C determined from concentration measured in the sample from the plant vent in the period of the ten days was 50 kBq/s. The uncertainty of this data was about 10%.

The Lagrangean model results for the two grid cells of 250mx250m, relevant for the direction of the point A and point B locations, are presented in Figures 3 to 5, as a rise of ^{14}C concentration at these locations due to ^{14}C release. Figures 3 and 4 show the result for the point A for two different release rate scenarios.

Comparison between the locations A and B is possible using Figures 4 and 5. The result for the point A is higher than for the point B which does not comply with the difference of the measured values.

Figure 3: Rise of the concentration of ^{14}C calculated for the grid cell A2 (Figure 6). Release rate is presented by Figure 2 as the ramp. One grid cell has dimension of 250 m x 250 m.

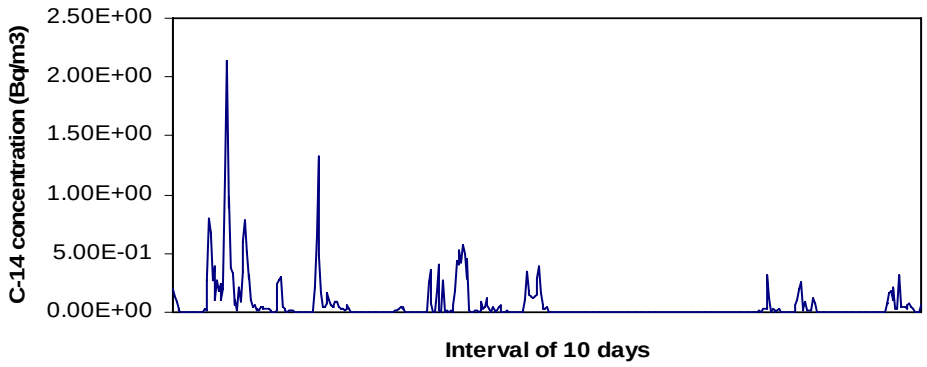


Figure 4: Rise of the concentration of ^{14}C calculated for the grid cell A2 (Figure 6). Release rate is constant during the time interval (50 kBq/s).

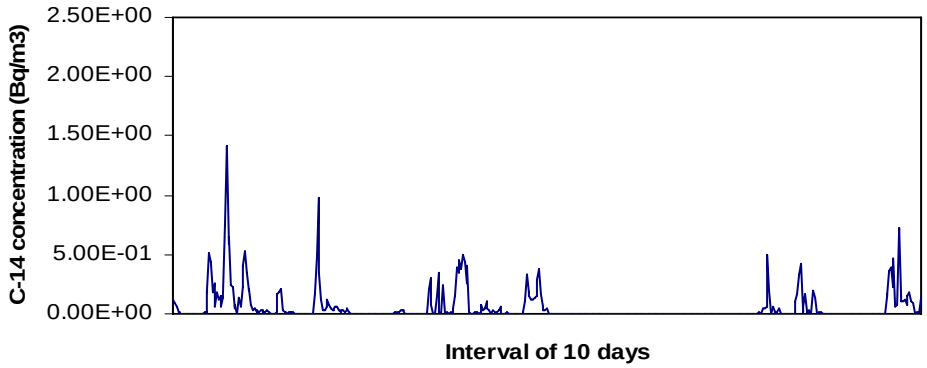
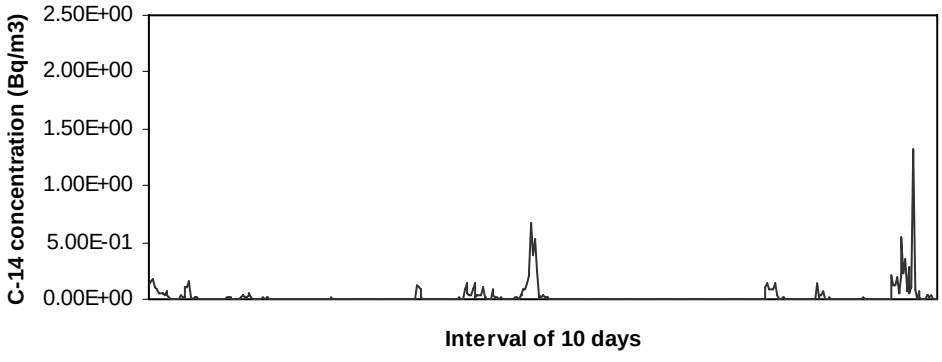


Figure 5: Rise of the concentration of ^{14}C calculated for the grid cell B (Figure 6). Release rate is constant during the time interval.



It is clear that due to the resolution of 250 m x 250 m the model gives average results for the whole cell and the measured value was only in one point. Figure 6 shows the results of the calculation for 5 cells. The concentration of ^{14}C is sensitive to direction rather than the release rate scenario.

For the three release rate scenarios, i.e. the constant release rate, the ramp and the one modulated by noble gas monitor response (Figure 2), the results vary less than 10 %. Considering direction from the release point, maximum calculated average concentration was 98 mBq/m^3 and the minimum was 65 mBq/m^3. The dispersion factor is dominant in case of this sensitivity analysis. The model was designed to give priority to wind direction measurement of sodar and not the meteorological tower. Sodar measurements were not so reliable due to higher environmental noise. Otherwise, the results of the calculated values in comparison to the measured ones are in the same range. In this case the east-west wind direction prevailed and this resulted in the higher ^{14}C concentration measured at the point B. It seems that the numerical results would comply to the measured ones only if the two points were exchanged. In the future, wind direction input to the model should be improved or the sodar replaced with a more powerful one.

Figure 6: Calculated average concentration of ^{14}C (including background 100.7 pCM) in the cell of the point B and in the cells A1 to A4 around the point A. The values with the unit (Bq/m3) show the result for constant release rate, the values without the unit is the result for ramp release rate in the time interval of 10 days (Figure 2).

Cell B 66 mBq/m3 * Point B	* Release point	Cell A1 82 mBq/m3 83 * Point A	Cell A2 98 mBq/m3 105
		70 mBq/m3 68 Cell A3	65 mBq/m3 66 Cell A4

8. Conclusion

The verification campaign of the dispersion modelling by airborne ^{14}C measurement close to the nuclear power plant proved that this method is sensitive enough and that the results are promising. The sensitivity analysis showed that wind direction input could be more accurate.

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