LA-UR-12-24856

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Title: Test of Clean Air Act Assessment Package-1988 predictions of Oxidized Tritium for Los Alamos National Laboratory

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Intended for: Report

Test of Clean Air Act Assessment Package-1988 predictions of Oxidized Tritium for Los Alamos National Laboratory

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1. Introduction

Through the Clean Air Act, the United States Environmental Protection Agency (EPA) requires emitters of anthropogenic radionuclides to calculate the equivalent dose to members of the public by using the Clean Air Act Assessment Package-1988 (CAP88) atmospheric dispersion and radiation dose assessment code. Los Alamos National Laboratory (LANL) uses CAP88 to model the dispersion and deposition of a variety of radionuclides, including oxidized tritium (HTO) for RAD-NESHAP compliance. Measurements of stack emissions of HTO are made by LANL to comply with NESHAP requirements. Concentrations of HTO are also measured at a variety of locations around the laboratory to help confirm compliance with public dose limits. CAP88 has been criticized for not accounting for turbulence associated with complex topography, such as the canyons in Los Alamos (Vauquelin 2000). Previous studies at sites other than LANL showed that the ratio of predicated concentrations to measured ambient concentrations of HTO at distant locations could be both under and over predicted depending on the wind direction, height of release point, and background tritium (Peterson 2004, Simpkins and Hamboy 1997). Other research hypothesizes that a properly used Gaussian plume model, such as CAP88, will be within a factor of 2-4 of measurements, with less accuracy for complex terrain and unsure meteorological conditions (Miller and Hivley 1987).

This project seeks to calculate the correlation between predicted values from CAP88 and measured ambient HTO concentrations from two stacks at LANL at distant locations for the year 2004 because the complex terrain has led to the postulation that CAP88 could unrealistically predict dispersion and deposition rates of HTO.

2 Calculations and Modeling

2.1Air Dispersion and Deposition Model

CAP88 predicts the diffusion of pollutants from a point source assuming a normally distributed plume and using default and site specific parameters. Values for the numerous parameters used in CAP88 were those used by LANL for NESHAP compliance. One of the important parameters in the precipitation scavenging coefficient determined by multiplying the rainfall rate (in cm/yr) by 1E-7 (yr cm^{-1} s^{-1}). The default deposition velocity of 1.8E-3 $m^{-1}sec^{-1}$ for particulates and zero for gases (CAP88-PC Version 2.1 User Guide) was also used. Site-specific meteorological information used included ambient air temperature, absolute humidity, wind speed and direction, the height of the lid, precipitation, and the amount and type of radionuclide released. Averages for Los Alamos, New Mexico include an annual precipitation of 45 cm y^{-1}, an ambient temperature of 9 degrees Celsius, an atmospheric mixing lid of 1,600 meters, and an absolute humidity of 5.5 grams per cubic meter.

The entire 2004 calendar year was split into two week periods and both stacks were modeled with 52 runs of CAP88. A wind file was created for each two week period from local meteorological data collected at four stations on LANL property (LANL 2010). The height, diameter and release rate of the two stacks were included as were the receptor distances and directions from the stack. Weekly HTO stack emissions totals in curies were determined by direct sampling. The sample is collected by an ethylene glycol bubbler and is later analyzed in a laboratory using a liquid scintillation technique.

2.2Ambient Air Measurements

Ambient air was monitored at and around LANL by AIRNET stations. The biweekly ambient HTO concentration (pCi/m^3) was determined by collecting moisture on silica gel followed by liquid scintillation analysis.

3. Results

Measured and predicted values of tritated air concentrations are shown below in Figures 1 and 2. Maximum concentrations were over-estimated by the model suggesting that CAP88 provides conservative estimates of dispersion. The program also over predicted the average concentration of HTO, however, the time profiles generally follow each other and correctly predicted the timing of spikes in the measured concentrations. To check for possible dependence of the prediction accuracy with stack emissions, the ratio of the predicted concentration to the ambient measurements were plotted as a function of total stack emissions in Figure 3. There was no significant statistical correlation between the amount of HTO emitted and the ratio of predicted concentrations to measurements; thus, showing that the accuracy of the CAP88 predictions were independent of the total emissions.

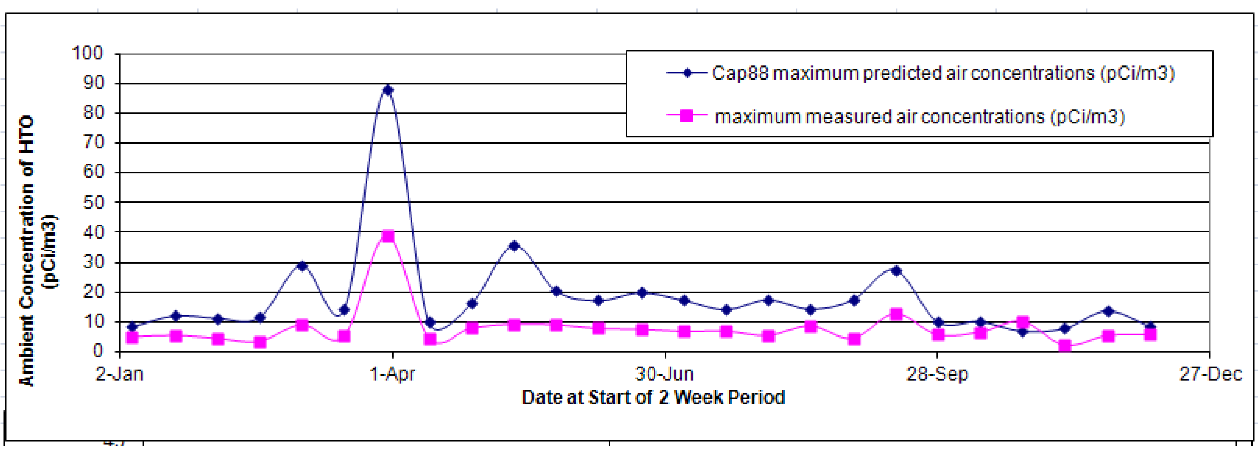

Figure 1: Maximum ambient concentrations of HTO in 2004.

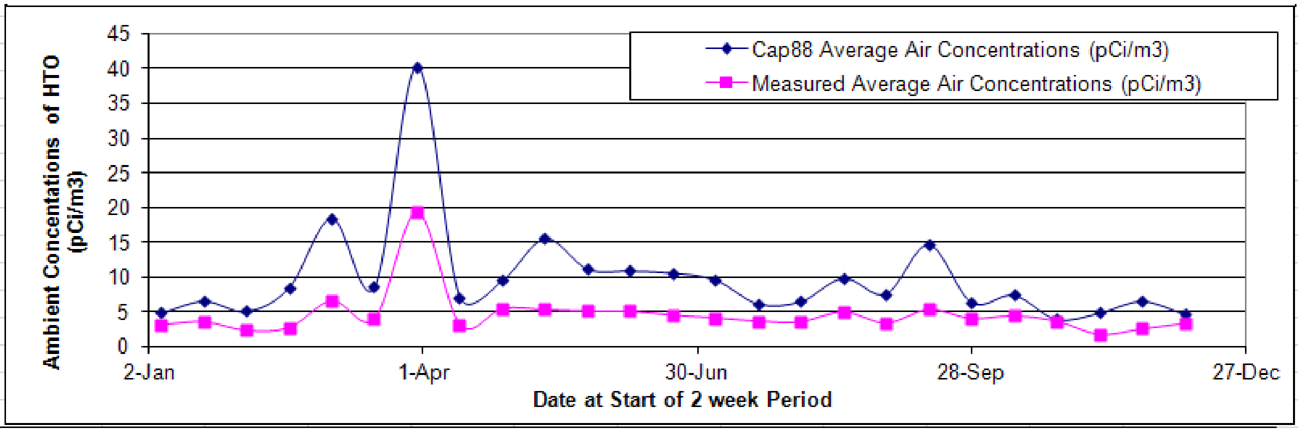

Figure 2: Average ambient concentrations of HTO in 2004.

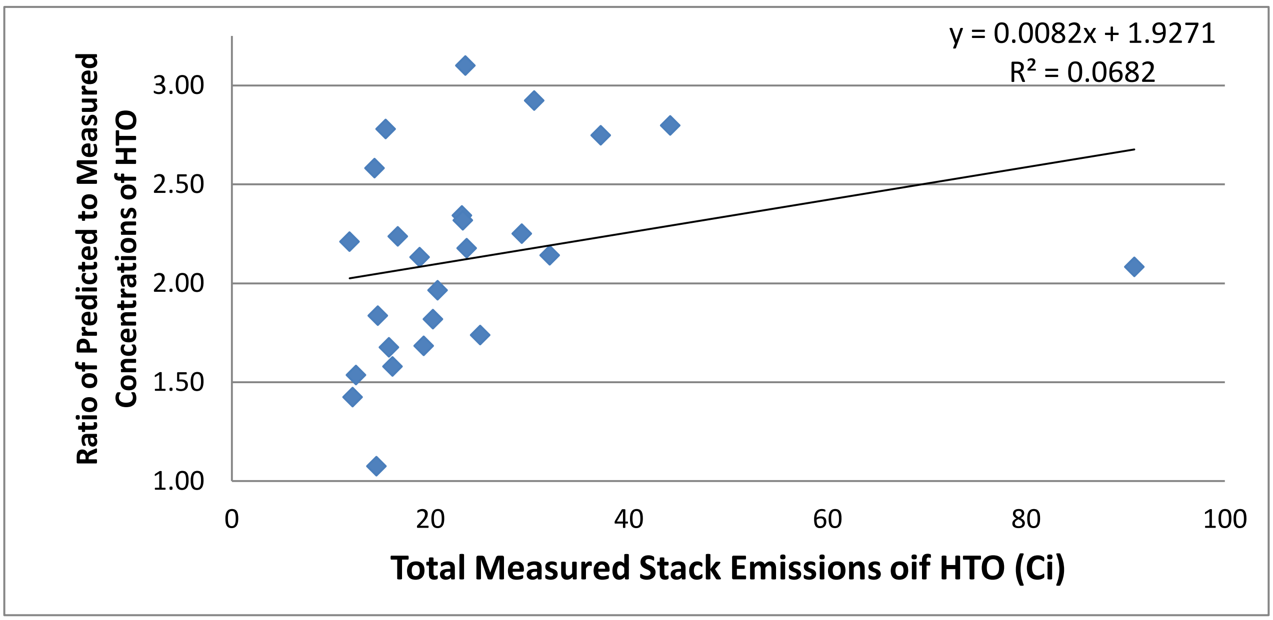

Figure 3: The ratio of predicted to measured values in relation to the amount of HTO emitted by both sources.

4. Discussion

CAP88 predicts higher average and maximum concentrations of HTO than those measured at receptor locations. Maximum concentrations are more accurately predicted than the average concentrations of HTO. Fluxuation in accuracy that may be anticipated because diurnal winds and seasonal precipitation patterns. Los Alamos has a windy spring with high winds in March, April, and May. Spring time is also associated with air mixing between the stratosphere and troposphere which can decrease horizontal dispersion of radionuclides (Roxanski *et al.* 1991). Seasonal fluxuations in precipitation have also been known to affect the dispersion of HTO in the atmosphere at other sites.

An additional factor that may contribute to overestimation of HTO is the height at which meteorological data is collected. Although the EPA standard for data height collection is 10m, Los Alamos collects data at 12m (ANSI/ANS-3.11-2005, ESR 2011). The appropriate wind speed can be calculated for the height of release to correct this problem and would be appropriate in future studies. This may be important because if wind is measured at a lower location than the release point than dispersion may be overestimated; this may be counter by the fact that light winds at lower locations result in greater plume rise (Peteson 2004).

The data does not suggest that there is a consistent ratio between measured and predicted concentrations. The ratio varies from 1 to 3.5 and is not dependent on the amount of HTO released during the period (low correlation coefficient, r^2= 0.068). If the outlier point for high stack emissions of 90 curies is removed the correlation improves only slightly to 0.3). Wind direction and precipitation variation probably account for the differences in the ratios. Previous studies have indicated the importance of accurate wind files and demonstrated the sensitivity of CAP88 to wind velocity (Peterson 2004). More accurate modeling would use absolute humidity and ambient temperature calculated for each two week period instead of using the annual average.

5. Conclusion

Results confirm those of the Tritium Working Group of the International Atomic Energy Agency's BIOMASS Co-ordinated Research Programme which suggest a ratio of 3 or less in most scenarios (IAEA 2003) . Overall CAP88 can be assumed to provide conservative estimates of the dispersion of radionuclides in air even in cases of extremely uneven terrain. Further analysis should be conducted at a variety of distances from the stack in this terrain to determine if estimates at distances greater than 2km are over or under estimated by CAP88.

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