Journal of Environmental Radioactivity xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

International challenge to model the long-range transport of radioxenon released from medical isotope production to six Comprehensive Nuclear-Test-Ban Treaty monitoring stations

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ARTICLE INFO

Keywords:
Atmospheric transport modelling
Nuclear explosion monitoring
Medical isotope production
Radioxenon background
Model inter-comparison and evaluation

ABSTRACT

After performing a first multi-model exercise in 2015 a comprehensive and technically more demanding atmospheric transport modelling challenge was organized in 2016. Release data were provided by the Australian Nuclear Science and Technology Organization radiopharmaceutical facility in Sydney (Australia) for a one month period. Measured samples for the same time frame were gathered from six International Monitoring System stations in the Southern Hemisphere with distances to the source ranging between 680 (Melbourne) and about 17,000 km (Tristan da Cunha). Participants were prompted to work with unit emissions in pre-defined emission intervals (daily, half-daily, 3-hourly and hourly emission segment lengths) and in order to perform a blind test actual emission values were not provided to them. Despite the quite different settings of the two atmospheric transport modelling challenges there is common evidence that for long-range atmospheric transport using temporally highly resolved emissions and highly space-resolved meteorological input fields has no significant advantage compared to using lower resolved ones. As well an uncertainty of up to 20% in the daily stack emission data turns out to be acceptable for the purpose of a study like this. Model performance at individual stations is quite diverse depending largely on successfully capturing boundary layer processes. No single model-meteorology combination performs best for all stations. Moreover, the stations statistics do not depend on the distance between the source and the individual stations. Finally, it became more evident how future exercises

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https://doi.org/10.1016/j.jenvrad.2018.01.030
Received 24 August 2017; Received in revised form 31 January 2018; Accepted 31 January 2018

need to be designed. Set-up parameters like the meteorological driver or the output grid resolution should be pre-scribed in order to enhance diversity as well as comparability among model runs.

1. Introduction

The Comprehensive Nuclear-Test-Ban Treaty (CTBT), an international agreement to ban all nuclear tests, has developed a global network of 321 monitoring stations and 16 laboratories for verification purposes (CTBT, 1996), the International Monitoring System (IMS). It monitors seismic, hydroacoustic, infrasound and radionuclide signatures (CTBTO, 2017).

The radionuclide component comprises measurements of aerosol-bound radioactivity at 80 locations. Half of the 80 stations shall have additional equipment to measure ambient air concentrations of four radioactive xenon isotopes (Xe-131 m, Xe-133, Xe-133 m, and Xe-135) produced in nuclear explosions. 31 noble gas stations are already in operation, and 25 of those have been certified by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO).

In 1999, the International Noble Gas Experiment (INGE) was launched to determine the feasibility of building and deploying automated systems to detect the four radioactive xenon (radioxenon) isotopes of interest (Auer et al., 2010; Bowyer et al., 2002). Commercial versions of three of the four radioxenon detection systems developed for the INGE are now deployed in the IMS: 1) The Automatic Radioanalyzer for Isotopic Xenon (ARIX), from the Khlopin Radium Institute, Russia (Dubasov et al., 2005), 2) the Swedish Automatic Unit for Noble Gas Acquisition (SAUNA, nowadays produced by Scienta Sauna Systems AB, Uppsala, Sweden), from Totalförsvarets Forskningsinstitut (FOI), Sweden (Ringbom et al., 2003), and 3) the Système de Prélèvement d' Air Automatique en Ligne avec l' Analyse radioXénons atmosphériques (SPALAX) from Departement Analyse, Surveillance, Environnement du Commissariat à l'Énergie Atomique (CEA/DASE), France (Fontaine et al., 2004).

Discrimination between radioxenon releases originating from a nuclear explosion or from civil facilities is a challenging task for the CTBTO. To our knowledge currently at least nine facilities worldwide are in operation: IRE located in Fleurus/Belgium, Mallinckrodt in Petten/the Netherlands, NIIAR in Dimitrovgrad/Russia, BaTek in Jakarta/Indonesia, NECSA in Pelindaba/South Africa, CENA in Ezeiza/Argentina, HFETR in Chengdu/China, PINSTECH PAAR-1 in Islamabad/Pakistan and ANSTO in Lucas Heights/Australia (Gueibe et al., 2017; Achim et al., 2016). Atmospheric transport modelling (ATM) combined with isotopic ratio analysis (Kalinowski et al., 2010) can be considered as the most important means for achieving this goal. A large number of studies of the release and transport of radioxenon from nuclear power plants and medical isotope production and other man-made radionuclide emission facilities have been conducted to develop an understanding of background levels (Eslinger et al., 2014; Hoffman et al., 2009; Kalinowski et al., 2008; Saey et al., 2010; Wotawa et al., 2010, 2003; Zaehringer et al., 2009; Achim et al., 2016; Schoeppner, 2017). These studies confirm that fission-based production of molybdenum-99 for medical purposes is the largest routine contributor of radioxenon (which comes as a by-product of the production process) in the atmosphere, and that related releases can be detected at large distances. The Mo-99 daughter Tc-99 m is widely used for medical purposes (Peykov and Cameron (2014), approximately 30–40 million medical procedures per year) and a future growth in demand is expected.

Radioxenon levels at IMS noble gas stations resulting from underground nuclear tests can be comparable to background levels (Ringbom et al., 2014; Saey, 2009) and are thus harder to detect in regions under the influence from medical isotope production facilities. A reduction of their radioxenon releases would therefore be useful (Bowyer et al., 2013). Nevertheless, medical isotope production facilities do meet regulatory release limits (Tinker et al., 2010; Hoffmann et al., 2001), thus their operators have little incentive for spending money on reduction measures.

Although atmospheric modelling studies using inert tracers have been conducted since the early 1980s (e.g., Ferber et al., 1986; Gudiksen et al., 1984), detailed source-term data for the simulation of the transport of radioxenon from medical isotope production facilities have not been made available until recently. A 2013 study examined the regional impact of source-term data of different time resolutions on the capability to predict IMS radioxenon detections (Schoeppner et al., 2013). The study utilized emission data down to daily time resolution from the ANSTO facility close to Sydney (Australia) and detections in Australia and New Zealand. It found increasing agreement between simulations and detections from annual down to daily time resolution of the emission data. Little influences from other sources in the Southern Hemisphere were observed. A recent international model comparison (1st ATM Challenge, Eslinger et al. (2016)) used Xe-133 stack emission data from the Institut des Radioéléments (IRE) radiopharmaceutical plant in Fleurus (Belgium) and activity concentration data collected at the IMS noble gas sampler at radionuclide station DEX33 (Schauinsland, Germany). The purpose of that exercise was to ascertain the level of agreement that can be achieved between Atmospheric Transport Models (ATMs) using stack monitoring data and xenon isotopic concentration measurements at IMS stations. One of the conclusions from that exercise was that using stack monitoring data to calculate radionuclide concentrations at a distance of about 400 km can match larger individual simulated sample concentrations (i.e., above 3 mBq m^{-3}) to within ± 40% of the measured concentrations if an optimally selected (according to the mean square error) ensemble mean of ATMs is used, and in some cases even lower deviations are achievable. Also, models using source term data in 15 min to 3 h time intervals produced similar agreement with measured concentrations as models using source term data averaged over longer intervals. In addition, even though the releases from IRE dominated the measured concentrations at DEX33, releases from other facilities such as nuclear power plants also influenced the smaller measured concentrations (see also De Meutter et al. (2016)). One of the benefits of that exercise was that it sparked many discussions on which techniques were most suitable, what knowledge and technique gaps exist, and what data fidelity is needed from stack monitors.

This current study also addresses the question of the level of agreement that can be achieved between IMS measurements and those simulated using Xe-133 stack release data and atmospheric transport modelling. Since ATM is a cornerstone of Treaty verification (Becker et al. (2007); Wotawa et al. (2003), including the discrimination between military and civil radionuclide sources) the scenario team of the challenge (made up by ZAMG, CTBTO/IDC and PNNL) sought broad participation of the respective community. The role of ATM in Treaty verification should be underpinned. Having at hand a multitude of simulations an ensemble approach pays off since this is the only way of overcoming individual ATMs' deficiencies and uncertainties and reproducing related measured samples more accurately. Reproducing measured radioxenon samples related to industrial production could be of great benefit to National Data Centers (NDCs, CTBT (1996)) which for verification purposes have to deal every month with a multitude of elevated radioxenon concentrations detected at IMS stations.

The setting of the current challenge is different in several ways compared to the previous one. Concentration data are used from six IMS radionuclide stations rather than just from one, as was used earlier.