



The global distribution of natural tritium in precipitation simulated with an Atmospheric General Circulation Model and comparison with observations



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ARTICLE INFO

Article history:

Received 30 April 2015

Received in revised form 22 June 2015

Accepted 24 June 2015

Available online 17 July 2015

Editor: B. Marty

Keywords:

tritium
hydrological cycle
GCM
stratospheric air intrusions

ABSTRACT

The description of the hydrological cycle in Atmospheric General Circulation Models (GCMs) can be validated using water isotopes as tracers. Many GCMs now simulate the movement of the stable isotopes of water, but here we present the first GCM simulations modelling the content of natural tritium in water. These simulations were obtained using a version of the LMDZ General Circulation Model enhanced by water isotopes diagnostics, LMDZ-iso. To avoid tritium generated by nuclear bomb testing, the simulations have been evaluated against a compilation of published tritium datasets dating from before 1950, or measured recently. LMDZ-iso correctly captures the observed tritium enrichment in precipitation as oceanic air moves inland (the so-called continental effect) and the observed north–south variations due to the latitudinal dependency of the cosmogenic tritium production rate. The seasonal variability, linked to the stratospheric intrusions of air masses with higher tritium content into the troposphere, is correctly reproduced for Antarctica with a maximum in winter. LMDZ-iso reproduces the spring maximum of tritium over Europe, but underestimates it and produces a peak in winter that is not apparent in the data. This implementation of tritium in a GCM promises to provide a better constraint on: (1) the intrusions and transport of air masses from the stratosphere, and (2) the dynamics of the modelled water cycle. The method complements the existing approach of using stable water isotopes.

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1. Introduction

Model–data comparison of humidity fields and water fluxes is one method of judging the skill of Atmospheric General Circulation Models (GCMs) at modelling the hydrological cycle. However, many GCMs now also model the movement of the stable isotopes of water (H_2^{16}O , H_2^{18}O , HDO, H_2^{17}O), and these well-known tracers of the past and present-day hydrological cycle can also be used to validate model performance (Hoffmann et al., 1998; Mathieu et al., 2002; Noone and Simmonds, 2002; Lee et al., 2007; Schmidt et al., 2007; Yoshimura et al., 2008; Risi et al., 2010; Werner et al., 2011).

Tritiated water (HTO) is another useful tracer. Opposite to stable isotopes of water which show variations expressed in ‰, driven by physical processes, HTO is rather a marker for reservoirs such as stratosphere, troposphere and oceans involved in the hydrolog-

ical cycle. The content of tritium in these different compartments indeed varies by orders of magnitude. The concentration of tritium in precipitation is monitored by the Global Network of Isotopes in Precipitation (GNIP), and the database is available through IAEA. A substantial dataset also exists of HTO concentration in polar ice (see Fourné et al., 2006 and the references therein). As a result, model–data comparison based on the HTO proxy is a promising way of increasing our understanding of GCM performance.

Natural tritium is mainly produced by the interaction of galactic cosmic rays (GCR) with nitrogen of the upper atmosphere, at an average rate of $\sim 2500 \text{ atoms m}^{-2} \text{ s}^{-1}$ (Craig and Lal, 1961; Masarik and Beer, 2009). It has a radioactive half-life of 4500 ± 8 days (Lucas and Unterwieser, 2000). The vast majority of this natural (cosmogenic) tritium, which amounts to a global inventory of 3.6 kg, enters the hydrological cycle in the form of tritiated water molecules (HTO). This can be compared with the $\sim 520\text{--}550$ kg of bomb tritium in the form of HTO (Michel, 1976; UNSCEAR, 2000) that was created in nuclear tests and subsequently caused a peak of bomb tritium in precipitation. Since the Nuclear Test Ban Treaty in 1963, the level of tritium measured in

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precipitation has been steadily decreasing due to radioactive decay and dilution in the world oceans. IAEA tritium long time series suggest that tritium levels in precipitation are close to their pre-nuclear test values.

It is estimated that 55% of the natural production of tritium occurs in the stratosphere (Masarik and Beer, 1999). The natural-tritium content of stratospheric water vapour has been estimated to be some $5\text{--}9 \times 10^5$ TU (Tritium Units, where 1 TU corresponds to T/H ratio of 10^{-18}) (Ehhalt et al., 2002; Fourné et al., 2006), which is several orders of magnitude greater than the natural-tritium level in precipitation (a few TU only). This difference is due to the higher production rate and the lower water content in the stratosphere. This large stratospheric reservoir also makes tritium an extremely valuable tracer for mapping the intrusion of stratospheric air masses into the troposphere, in particular in Antarctica, a region under the influence of the polar vortex (Taylor, 1968; Jouzel et al., 1979, 1982; Wagenbach et al., 1998; Fourné et al., 2006).

Koster et al. (1989) pioneered the implementation of tritium in a GCM. In the only study published so far, these authors simulated the bomb-produced tritium atmospheric distribution with the GISS GCM. They used a simple bomb-tritium input function, a low model resolution ($8^\circ \times 10^\circ$ and 9 vertical layers) and only a very short simulation period (30 July days). They did not study the latitudinal variations of tritium concentration in the tropospheric vapour and precipitation. In addition, the low model resolution and short simulation period prevented the detailed study of continental recycling. Further, the correct representation of stratospheric air inputs in a GCM requires a higher vertical resolution than could be used by Koster et al. (1989), especially above the tropopause. Simulations must also be longer than one year to study of its seasonal cycle. Finally, tritium data for the Koster et al. (1989) study were much more scarce than now, which restricted the model evaluation. With a current state-of-the-art GCM run on a modern computer, these limitations need no longer apply.

This paper describes the implementation of tritium in the Laboratoire de Météorologie Dynamique Zoom (LMDZ) Atmospheric General Circulation Model developed at LMD (Hourdin et al., 2006, 2013). Our objective is to test the simulated distribution of HTO in the atmosphere due to climate processes; we therefore focus on natural tritium and run the simulations without the massive increase of tritium due to nuclear weapon testing. This restriction allows us to study the transport of tritium and its spatial/seasonal variability under steady-state tritium inputs.

Here, the simulation of the natural-tritium content of water vapour and precipitation is validated against a compilation of published tritium datasets of pre-bomb samples dated before 1950 and of recent samples for which the bomb component can be neglected.

The outline of this paper is as follow. In Section 2, we give a description of the isotopic processes included in the model. We also describe how the model has been adapted for tritium implementation, the various simulations performed and the tritium datasets used for validation. In Section 3, we present simulated spatial variations of HTO in precipitation and water vapour and validate them against tritium data. We also evaluate the seasonal variability linked to exchanges of air masses between the stratosphere and the troposphere over two well-documented areas, Europe and Antarctica. In Section 4, we use the patterns of observed and modelled spatial variability of HTO to identify its driving mechanisms for present-day climatology and apply specific sensitivity tests on these two regions.

2. Model simulations and datasets

2.1. LMDZ-iso and isotopic processes

Risi et al. (2010) have already implemented water isotopes in the LMDZ model in the version LMDZ4 (Hourdin et al., 2006). Since then, developments improved the capabilities of the model and we have implemented tritium in version LMDZ5a (Dufresne et al., 2013), whose isotopic version is hereafter referred to as LMDZ-iso. The physical parameterisations of LMDZ5a are very similar to those used in LMDZ4, with the same Emanuel convective parameterisation (Emanuel, 1991) coupled to the Bony and Emanuel (2001) cloud scheme. The advection of water in its vapour and condensed states is calculated using the van Leer (1977) scheme. The dynamical part is based on a discrete latitude–longitude grid, at the standard resolution of $2.5^\circ \times 3.75^\circ$. To ensure a realistic description of the stratosphere and of the Brewer–Dobson circulation, the model is run with 39 layers in the vertical (against 19 in the LMDZ4 version), including 22 located above the 200 hPa pressure level (Lott et al., 2005).

Following Risi et al. (2010), LMDZ-iso uses the same physical and dynamical descriptions of the HTO molecule as for the other water isotopes. Due to mass and symmetry differences, the various isotopic forms of the water molecule (H_2^{16}O , H_2^{18}O , HDO, H_2^{17}O , HTO) have slightly different physical properties (e.g., saturation vapour pressure, molecular diffusivity), and are therefore redistributed between the vapour and condensed phases at each phase change differently, depending on atmospheric conditions (temperature, vapour saturation, etc.). The implementation of the water stable isotopes H_2^{16}O , H_2^{18}O and HDO in LMDZ-iso and of their associated isotopic processes (kinetic and equilibrium) have been extensively described by Risi et al. (2010) and Bony et al. (2008). Contrary to stable isotopes of water, the uncertainties of these isotopic effects are usually considered of minor importance because of the larger differences in tritium content in water between the reservoirs.

The saturated vapour pressure of heavier molecules (e.g., HTO) differs from that of lighter ones (e.g., H_2^{16}O) creating equilibrium fractionation during a change of phase. The temperature-dependent coefficients for equilibrium fractionation α_{eq} between vapour and liquid water or ice for HTO are given by Koster et al. (1989). Equilibrium fractionation has a significant effect on the variations of the ratio $\text{HTO}/\text{H}_2^{16}\text{O}$ in the precipitation. As an example for a temperature of 10°C , the coefficient for liquid/vapour fractionation, α_{eq} , is equal to 1.2378 for HTO/ H_2^{16}O fractionation, compared to α_{eq} values of 1.0107 and 1.098 for $\text{H}_2^{18}\text{O}/\text{H}_2^{16}\text{O}$ and HDO/ H_2^{16}O fractionations, respectively.

Isotopic fractionation is also introduced during evaporation of water at the ocean surface (with the ocean considered as an infinite reservoir). As for the water stable isotopes, the isotopic evaporation of HTO is computed following Merlivat and Jouzel (1979), including equilibrium fractionation and kinetic effects depending on the surface wind speed.

Re-evaporation and diffusive exchanges between rain and vapour as the rain falls through the atmosphere are taken into account in the model. At any level in the vertical, the isotopic composition of the precipitating water depends on several parameters: (1) the composition of the condensed water that has been converted to precipitation at upper levels, (2) the fraction of precipitation that has re-evaporated and (3) the diffusive exchanges that take place between the falling drops and the downdraft water vapour. The re-evaporation and diffusion processes are calculated depending on relative humidity following Bony et al. (2008), who extended the work of Stewart (1975). The calculation includes the kinetic effects associated with the re-evaporation of the drops in an unsaturated environment. If the relative humidity is 100%, we

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