Historical atmospheric deposition of PAHs in lakes east of the Athabasca oil sands operations

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The growth of bitumen mining activities in northeastern Alberta poses significant environmental challenges. One of the main concerns is a potential increase in emissions of harmful organic contaminants such as polycyclic aromatic hydrocarbons (PAHs). PAHs are widespread in the environment and result from incomplete organic matter combustion (kinetic process) or maturation (thermodynamic process), which makes them good markers of pyrogenic or petroleum input.

In order to understand the impact of oil sands-related mining activities to the environment it is essential to examine PAH emissions in the context of variability in natural background levels. Here we report geochronological records of PAH atmospheric deposition fluxes over the last century in two headwater lakes located approximately 50 km downwind from the main area of oil sands operations. Assessing PAH levels over the last 100 years allowed us to differentiate the deposition fluxes prior to the modern period of bitumen mining activities (i.e., pre-1970). Concentrations of the 16 EPA priority PAHs in addition to coronene, retene, perylene and 6 alkylated groups were measured in dated cores from both lakes. PAH molecular diagnostics ratios were used in order to discriminate preliminary sources and their variation over time.

Results and conclusion:

Our results showed an increase of parent and alkylated PAHs (2-fold for alkylated PAHs and 0.5-fold for 16 EPA priority PAHs) in both lakes over the past decade. Alkylated PAHs are generally related to petroleum sources. A possible explanation for this increase may be the expansion of open mine pit area resulting in greater sensitivity to wind erosion and transport. PAH molecular diagnostics ratios revealed two distinct groupings using two different sets of ratios: a pre-1980s combustion dominated type, and a post-1980s petroleum dominated one. These results point to an increasing contribution from recent oil sands mining activities, though it should be noted that overall PAH levels in these lakes are relatively low. In light of these results, and taking into consideration the future expansion plans of bitumen mining activities, it is possible that PAH fluxes in this region may continue to rise. Further insight into PAH source apportionment is expected from ongoing work involving compound-specific isotope analysis.

Meridional distribution of the atmospheric $^3He/^4He$ isotopic ratio

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Due to the release of crustal helium by natural gas and oil production and by coal mining with a low $^3He/^4He$ ratio, the atmospheric $^3He/^4He$ ratio may be decreasing with time. The direct detection of this change is problematic due to the difficulty in finding well preserved ancient air samples and has led to conflicting results. Most of the He release occur in the northern hemisphere, therefore it may create an interhemispheric gradient in the atmospheric $^3He/^4He$ ratio, as already observed for other anthropogenic gases such as CH_4, CFC or ^{85}Kr. We performed precise helium isotope measurements on air samples collected at various latitudes going from 82°30'N (Alert Station) to 78°38'S (Vostok Station). The mean helium isotope ratio (relative to our Saclay air standard) are identical for both hemisphere : 1.0004 ± 0.0005 for the northern hemisphere and 1.0004 ± 0.0009 for the southern hemisphere respectively, thus showing no detectable meridional gradient. However, as shown in Fig. 1, $^3He/^4He$ values are very homogeneous from the northermost latitudes to 20°S but the two southernmost data points show diverging trends. Therefore, additional data are needed south of 20°S.

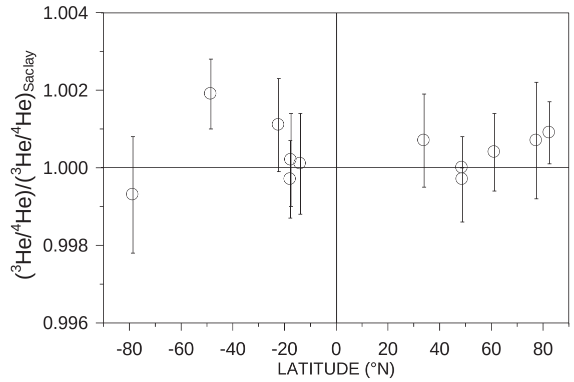

Figure 1: Meridional distribution of the helium isotopic ratio of air

The lower value for Vostok could be due either to the high elevation (3500 m) or to the relative isolation of the air masses over the Antarctic interior. If we consider this latter data point as an outlier, the mean $^3He/^4He$ value for the southern hemisphere becomes 1.0006 ± 0.0009. The two mean values, $^3He/^4He = 1.0004$ for the northern hemisphere and $^3He/^4He \leq 1.0006$ for the southern hemisphere, can be used to estimate the value of the anthropogenic helium flux using a simple two-box model of the atmosphere with an interhemispheric mixing time of about 1 year [1-2] and a anthropogenic helium input function proportional to the world release of fossil carbon since 1850. Model results indicate that the He/C molar ratio may be $\leq 2\times10^{-4}$, with a decrease of the atmospheric $^3He/^4He$ ratio $\leq 0.7\%$ over the industrial period.

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