ESTIMATION OF LONG-TERM TRENDS IN THE TROPOSPHERIC $^{14}CO_2$ ACTIVITY CONCENTRATION

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ABSTRACT. Fossil CO_2 emissions have been diluting the global $^{14}C/C$ ratio of atmospheric CO_2 (Suess effect). We estimated the $^{14}CO_2$ amount in the atmosphere (and its trend) utilizing the calculated $^{14}CO_2$ activity concentration in the atmosphere (a_{acn}, reported in mBq m^{-3}). This parameter, calculated from $\Delta^{14}CO_2$ and the CO_2 mixing ratio (reported in micromoles of CO_2 per mole of air), is connected with the $^{14}CO_2$ quantity in the volume or mass unit of air, which is not influenced by the Suess effect. This parameter can only be influenced by processes linked to $^{14}CO_2$ emissions/uptake, e.g. associated with atmosphere-biosphere or atmosphere-ocean CO_2 exchange as well as by anthropogenic emissions of $^{14}CO_2$. Results obtained from measurements at Schauinsland station, Germany, indicate a stable amount of $^{14}CO_2$ in the atmosphere since the early 1990s.

INTRODUCTION

Environmental compartments contain a mixture of 2 stable carbon isotopes (^{12}C and ^{13}C) and one radioactive isotope ^{14}C (radiocarbon). This radionuclide of global occurrence and a half-life of 5730 yr is partly of anthropogenic origin. In nature, ^{14}C is produced by nuclear reactions generated by cosmic rays in the atmosphere (Lal and Peters 1967; Burchuladze et al. 1980). The natural ^{14}C production rate is balanced by its removal from the atmosphere to other environmental compartments (e.g. biosphere, ocean). The role of direct radioactive decay in the atmosphere is negligible due to its long half-life and the short residence time of CO_2 in the atmosphere of only a few years. In the last century, nuclear weapons tests were important sources of anthropogenic ^{14}C. Consequently, $\Delta^{14}C$ in the Northern Hemisphere in 1963 was double the level of its natural production (Nydal and Lövseth 1965; Levin et al. 1995; Meijer et al. 1995). Since 1963, $\Delta^{14}CO_2$ has been decreasing exponentially due to its fast transfer to oceanic and terrestrial carbon reservoirs (Levin et al. 1980, 1995; Segl et al. 1983; Burchuladze et al. 1989; Hesshaimer et al. 1994; Levin and Kromer 1997). During the 1980s, the decrease has decelerated and from the early 1990s, the decrease can be represented by a linear trend (Levin et al. 1985, 2008, 2010; Levin and Kromer 2004).

On the planetary scale, the total ^{14}C inventory is predominantly caused by natural production (Lal and Peters 1967; Lal and Suess 1968). Both the atmosphere and biosphere contain only a small part of the total environmental ^{14}C. Radiocarbon dioxide, as the dominant chemical form of atmospheric ^{14}C, is transferred relatively promptly from the atmosphere into other environmental compartments. The proportion of anthropogenic ^{14}C is relatively small (below 2%), as seen in Table 1. ^{14}C effluents from nuclear energy facilities (nuclear power plants and spent nuclear fuel reprocessing plants) correspond to ~0.02% of the total ^{14}C inventory. The dominant part (~88%) of this anthropogenic ^{14}C is released by spent nuclear fuel reprocessing plants (UNSCEAR 2000).

Fossil CO_2, entering the atmosphere from the fuel combustion process, dilutes the $^{14}C/C$ ratio. Thus, the resulting $\Delta^{14}CO_2$ is lowered, which is known as the Suess effect (Suess 1955). The local Suess effect can be larger than $\Delta^{14}C$ ~100‰, particularly in urban areas with point CO_2 sources (fossil fuel

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Table 1 Global inventory of ^{14}C and annual ^{14}C releases (Lal and Peters 1967; Lal and Suess 1968; UNSCEAR 2000).

Source	Global inventory (PBq)	Annual production (PBq yr^{-1})	Relative share from the global inventory (%)
Natural production	12,750	1.54	98.34
Nuclear weapons testing	213	—	1.64
Nuclear energy production[a]	2.8	0.15[b]	0.022
Total	**12,966**	**1.69**	**100**

[a] Including releases from spent fuel reprocessing plants.

[b] This data covers the 1995–1997 period, recalculated according to UNSCEAR (2000).

power plants, incinerators, heating plants) (e.g. Levin et al. 1980), as well as in the vicinity of motorways (Kuc and Zimnoch 1998). Anthropogenic emissions of fossil carbon into the atmosphere have been increasing every year (CDIAC 2009). A respective trend can be seen in the atmospheric CO_2 mixing ratios. The increasing concentration of this greenhouse gas is mainly caused by anthropogenic releases (IPCC 2007).

SAMPLES AND METHODS

Monitoring of atmospheric $^{14}CO_2$ has been carried out at 2 sites in the Czech Republic (Figure 1): Prague-Bulovka (near a busy motorway; therefore, a local influence from fossil fuel combustion is observed at this site), and Košetice (a relatively clean area with only a small local load from fossil fuel combustion). In both locations, monthly integrated samples are collected using dynamic absorption in carbonate-free 0.7M NaOH solution. ^{14}C activity determinations are based on benzene preparation and measurement by a low-background liquid scintillation spectrometer Quantulus 1220™ (Svetlik et al. 2006). For calibration purposes, oxalic acid NIST SRM 4990C has been used (Schneider et al. 1995). The $^{13}C/^{12}C$ isotopic ratio in the atmospheric CO_2 was determined by a mass spectrometer using a few mL of exposed NaOH solution. Final activities, after $\delta^{13}C$ correction, are reported in per mil of $\Delta^{14}C$, following Stuiver and Polach (1977). The resulting combined uncertainties of ^{14}C activity determination have been below 7‰ (Curie 1995).

An integrating sampling system has been developed and applied for atmospheric ^{14}C measurements in Hungary (Molnár et al. 2007). One $^{14}CO_2$ sampler was installed in Debrecen, where also a high-precision atmospheric CO_2 monitoring station based on an ULTRAMAT 6F instrument was launched in September 2008 (Molnár et al. 2009). The second station was operated ~300 km from Debrecen at Hegyhátsál (as an independent background reference station), where high-precision atmospheric CO_2 mixing ratio measurements have been made since 1997. During the winter of 2008/09, the mixing ratio and ^{14}C activity of atmospheric CO_2 at Debrecen and at the reference station were measured simultaneously (2 m above ground level in Debrecen; 10 m and 115 m above ground in Hegyhátsál) (Molnár et al. 2010). Routine sample processing in the Debrecen laboratory is based on extraction of CO_2 from the samples by adding concentrated (75%) sulfuric acid into the exposed NaOH solution. The liberated CO_2 was purified over charcoal, frozen into a CO_2 trap with liquid nitrogen at –196 °C, and the remaining non-condensable components were removed by a vacuum pump. The activity of the samples was measured using the gas proportional counting method (Csongor et al. 1982; Csongor and Hertelendi 1986; Hertelendi et al. 1989; Molnár et al. 2007). The standard deviation of a single $\Delta^{14}C$ measurement was below ±5‰ (Hertelendi 1990). $\delta^{13}C$ was measured by a stable isotope mass spectrometer. $\delta^{13}C$-corrected $\Delta^{14}C$ data are reported relative to an absolute ^{14}C standard (Stuiver and Polach 1977) using NIST SRM 4990 (oxalic acid II).

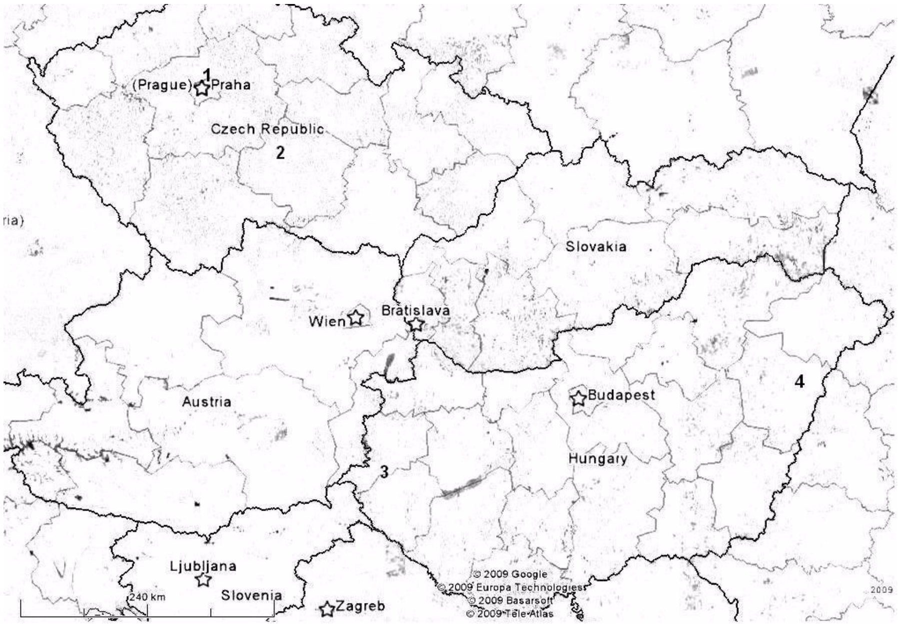

Figure 1 Location of monitoring localities in the Czech Republic (1 - Prague-Bulovka; 2 - Košetice) and Hungary (3 - Hegyhátsál; 4 - Debrecen).

Sampling station locations are shown in Figure 1. The observed levels of atmospheric $\Delta^{14}CO_2$ activity have already been reported (Svetlik et al. 2006, 2010; Molnár et al. 2007).

The atmospheric CO_2 mixing ratio has been measured in the Prague-Bulovka station since 2002 using the volumetric method. These measurements are primarily for other applications, where higher combined uncertainties (up to 3%) are acceptable. Calculations of $^{14}CO_2$ activity concentrations a_{acn} were carried out as described below. These results were associated with considerably higher uncertainties and can therefore be regarded as auxiliary data only. Activity concentrations a_{acn} (reported in mBq of ^{14}C per m^{-3} of air) of atmospheric $^{14}CO_2$, normalized to standard conditions (0 °C, 101.325 kPa), can be calculated from the values of atmospheric CO_2 mixing ratio and $\delta^{14}C$ utilizing the formula:

$$a_{acn} = a_{stdd}(1 + 0.001 \times \delta^{14}\mathrm{C})\frac{c_{CO_2}M_C}{V_{Mair}} \tag{1}$$

where c_{CO_2} is the mixing ratio of CO_2 in ppm, M_C is the molar weight of carbon (12.01 g mol^{-1}), V_{Mair} is the molar volume of air normalized to standard conditions (0 °C, 101.325 kPa, corresponding to 22.468 10^{-3} m^3 mol^{-1}), and a_{stdd} is the activity of ^{14}C standard (in Bq g^{-1} of carbon), without normalization for isotope fractionation (0.226 Bq g^{-1}).

The available published values of $\Delta^{14}C$ and CO_2 mixing ratios from the monitoring station at Schauinsland (SIL) (Levin and Kromer 2004; Levin et al. 2008; CDIAC 2009; WDCGG 2009) were compared with our results. Both the CO_2 mixing ratios and $\Delta^{14}C$ generally show large spatial variability; therefore, to calculate a_{acn}, data from the same locality are necessary. To convert the published values from Schauinsland from $\Delta^{14}C$ to $\delta^{14}C$, the estimated mean value of $\delta^{13}C$ of about –8.5‰ VPDB was applied (Levin and Kromer 1997). The time course of activity concentration a_{acn}

of atmospheric $^{14}CO_2$ in Schauinsland was calculated for the period from January 1977 to December 2003.

RESULTS AND DISCUSSION

The activity concentration a_{acn} is connected to the ^{14}C ($^{14}CO_2$) quantity in a cubic meter of air, and cannot be influenced by presence of fossil carbon (Suess effect). On local scale, the $^{14}CO_2$ activity concentration a_{acn} can be influenced only by ^{14}C releases, such as effluents from nuclear energy facilities or $^{14}CO_2$ emissions from organic substances decomposing in soils (heterotrophic respiration), as such carbon-based substances also contain ^{14}C. On the other hand, the $^{14}CO_2$ activity concentration a_{acn} can be also affected by $^{14}CO_2$ uptake by photosynthesis in biota (causing a decrease of CO_2 and $^{14}CO_2$ amount in the air).

The resulting $^{14}CO_2$ activity concentrations a_{acn} from the Prague-Bulovka station, calculated in mBq m^{-3}, have a greater combined uncertainty than those from Schauinsland (see Figure 2). Annual averages decreased from 54.4 ± 1.2 mBq m^{-3} (2002) to 51.8 ± 0.8 mBq m^{-3} (2008), although such differences are close to the limits of statistical significance due to the uncertainties of volumetric CO_2 mixing ratio determinations (~3%).

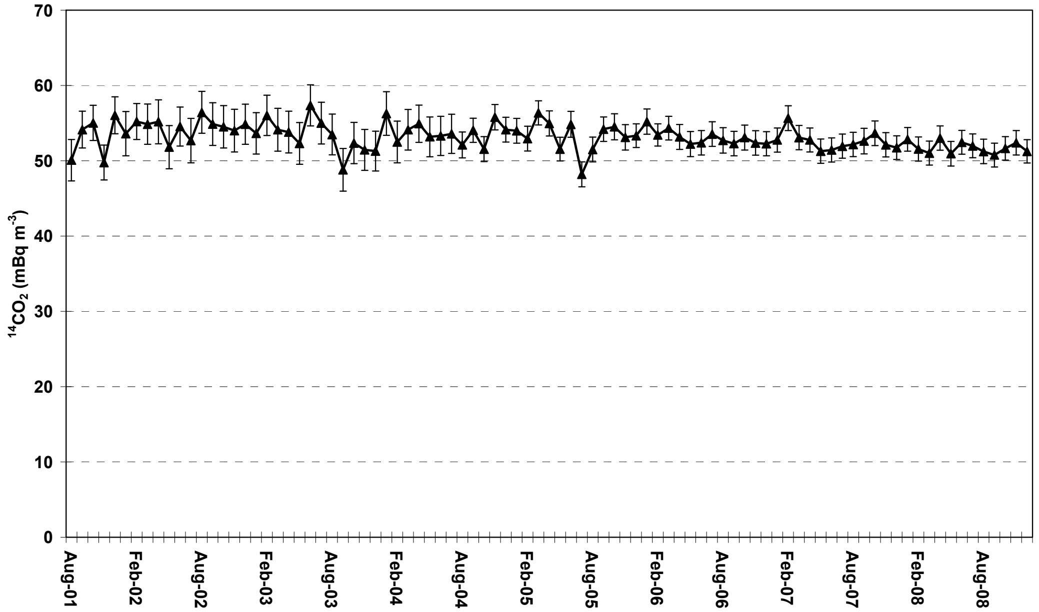

Figure 2 Time behavior of activity concentration a_{acn} (mBq m^{-3}) observed at the Prague-Bulovka station (normalized to standard conditions).

The activity concentration a_{acn} curve calculated from Schauinsland data shows seasonal variations (Figure 3). As seen from the comparison, the seasonal maxima and minima of CO_2 mixing ratio (concentration) have similar positions as the seasonal changes of $^{14}CO_2$ activity concentration. The $^{14}CO_2$ activity concentration a_{acn} curve shows a long-term decrease until about 1990. After 1992, the mean annual activity concentration of $^{14}CO_2$ a_{acn} stabilized, without a significant trend, which is even more evident from annual average values (Figure 4). This means that the number of ^{14}C atoms (or $^{14}CO_2$ molecules) in a normalized volume unit of air does not show a significant systematic decrease from 1992 to 2003 (end of the calculated data series).

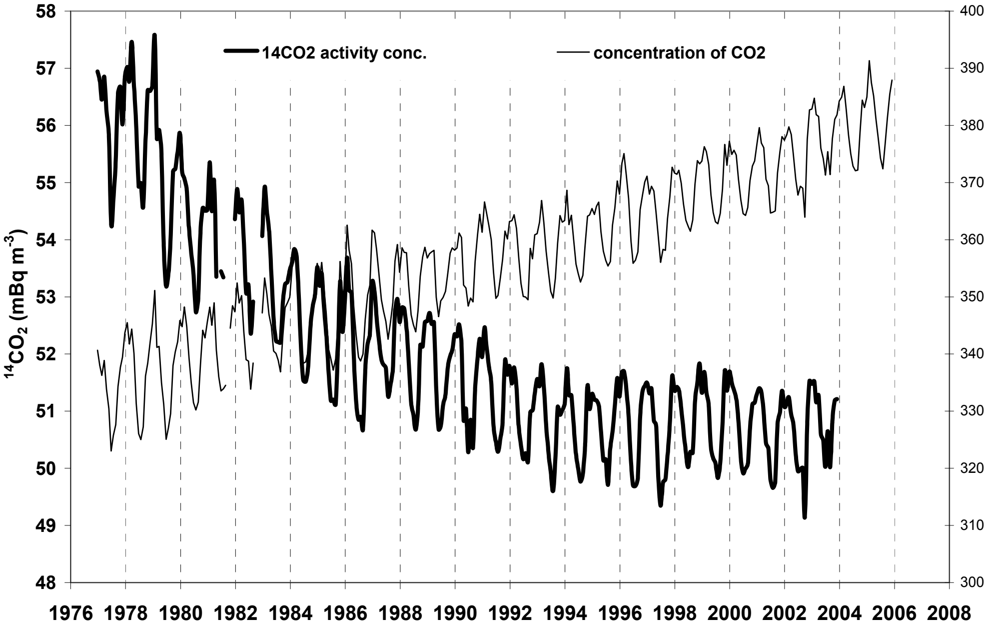

Figure 3 Activity concentration of the atmospheric $^{14}CO_2$ (a_{acn}, mBq m^{-3}), calculated from the published data from the Schauinsland monitoring station, Germany (Levin and Kromer 2004; Levin et al. 2008; CDIAC 2009; WDCGG 2009). The activity concentration was calculated from $\Delta^{14}CO_2$ and the mixing ratio of CO_2 (normalized to standard conditions).

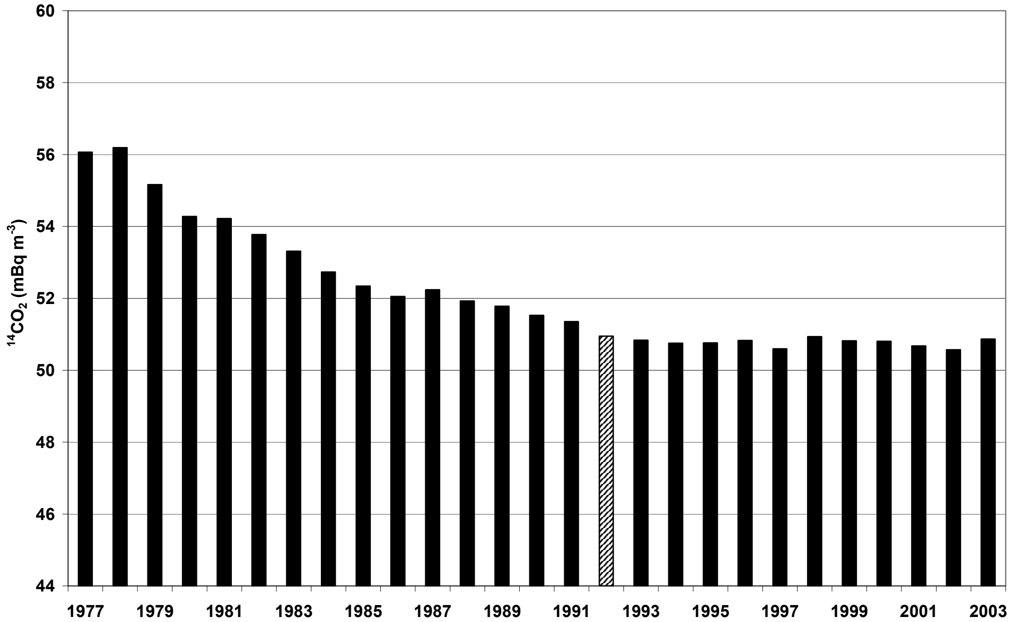

Figure 4 Annual mean values of atmospheric $^{14}CO_2$ activity concentration (a_{acn}, mBq m^{-3}) at the Schauinsland monitoring station, Germany (Levin and Kromer 2004; Levin et al. 2008; CDIAC 2009; WDCGG 2009). As evident from the diagram, a systematical interannual decreasing trend does not occur since 1992 (normalized to standard conditions).

The mean activity concentration a_{acn}, calculated from the data published for the 1992–2003 period for the Schauinsland station, amounts to 50.8 ± 0.7 mBq m^{-3}. The mean activity concentration a_{acn} calculated for the Prague-Bulovka site during 2002–2008 is 53.2 ± 1.7 mBq m^{-3}. The difference between the Prague-Bulovka and Schauinsland stations is close to the level of statistical significance. This difference may partially be due to systematic errors in the calibration and volumetric determination of the CO_2 mixing ratio at Prague-Bulovka.

The data calculated for September 2008 to April 2009 from the Hungarian monitoring sites (Molnár et al. 2010) give mean a_{acn} values of 53.2 ± 0.8 mBq m^{-3} (Debrecen), 52.8 ± 0.7 mBq m^{-3} (Hegyhátsál, 10 m above ground level), and 52.1 ± 0.8 mBq m^{-3} (Hegyhátsál, 115 m above ground level). Maxima of a_{acn} are observed during the winter, similar to the Schauinsland time series. Due to the short monitoring period, the annual mean values will most likely be smaller.

As previously mentioned, the amount of ^{14}C in the atmosphere depends on its cosmogenic production, current releases from nuclear energy facilities, plant respiration and photosynthesis, efflux of $^{14}CO_2$ from ocean to the atmosphere, and on its transfer into other environmental compartments, e.g. the biosphere and ocean (Levin et al. 2010). Therefore, if the ^{14}C content in the atmosphere is stable, this should be due to a dynamic equilibrium between the processes connected with ^{14}C inputs and outputs from the atmosphere. It seems that ^{14}C appearing in the air due to nuclear bomb tests has reached a balanced distribution in the atmosphere and associated compartments of the environment since 1992.

Such a balanced distribution of $^{14}CO_2$ in the atmosphere may have a limited duration as it can be influenced by a future $^{14}CO_2$ efflux from oceanic or biota sinks of residual bomb ^{14}C (Caldeira et al. 1998; Levin et al. 2010). Also, a stable amount of atmospheric $^{14}CO_2$ can be “quasi balanced” as a result of compensating superposition of several changing fluxes of ^{14}C, for instance: a) slowly changing emissions of anthropogenic ^{14}C from nuclear energy facilities as a result of decommissioning; and b) slowly increasing CO_2 efflux from ocean or biota with depleted or increased amounts of $^{14}CO_2$.

Supposing a stable ^{14}C activity concentration in the atmosphere during 1992–2003 (Figure 4), the observed linear decrease of $\Delta^{14}C$ since the beginning of 1990s (Levin et al. 2008; Svetlik et al. 2010) may be explained primarily by a linear increase of the CO_2 flux from fossil carbon. Anthropogenic emissions from fossil fuel combustion are a probable source of such CO_2, and have a prevailing influence on increasing CO_2 concentration in the atmosphere (IPCC 2007).

Unfortunately, the long-term trend of $^{14}CO_2$ quantity in the atmosphere can be evaluated only from a single site. Moreover, Schauinsland has significant amounts of local pollution, and there is some indication that changes in atmospheric transport in this region over the last few years have influenced the trend (Levin and Rödenbeck 2008). Therefore, data from additional sites, covering the same time period, are needed to support the presented results. To evaluate global trends in the $^{14}CO_2$ activity concentration a_{acn} in the atmosphere, data from other monitoring stations are essential. Regrettably, there are only a few monitoring localities with simultaneous long-term records of $\Delta^{14}CO_2$ and CO_2 mixing ratios.

CONCLUSIONS

New information has been obtained on ^{14}C quantity in the atmosphere using ^{14}C activity concentrations calculated from published Schauinsland data and our data. Although the annual decrease of atmospheric $\Delta^{14}CO_2$ has been ongoing since the 1990s, the mean amount of ^{14}C ($^{14}CO_2$) in the atmosphere seems to be stable since 1992. The decrease of atmospheric $\Delta^{14}CO_2$ can be explained only by increasing amount of fossil CO_2 in the atmosphere originating from fossil fuel combustion (global Suess effect).

The available long-term time series of atmospheric $^{14}CO_2$ activities and CO_2 mixing ratios observed simultaneously at the same site were available only for the Schauinsland station. In order to validate our estimation and the role of global Suess effect, it would be necessary to compare long-term trends of activity concentrations from several stations, globally distributed, located in different environmental conditions, and with different loads from local fossil fuel combustion.

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