

Greenhouse Gas Emissions at the Urban Scale



Industrial landscape with smog. © KRAJEWPIO, DREAMSTIME.COM

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Cities are responsible for more than 70% of global anthropogenic greenhouse gas emissions from buildings, transport, energy, industry, and waste-related sources. Improved urban-scale emission estimates are essential for understanding local trends and providing guidance for mitigation strategies. Current research in cities around the world is focused on establishing more robust methods for quantifying and modeling urban-scale emissions of the most abundant anthropogenic greenhouse gases: carbon dioxide (CO₂), methane (CH₄), and nitrous oxide (N₂O).

KEYWORDS: urban, cities, greenhouse gas emissions, carbon dioxide, methane, nitrous oxide

TOP-DOWN VERSUS BOTTOM-UP ESTIMATES OF GREENHOUSE GAS EMISSIONS

Total anthropogenic greenhouse gas (GHG) emissions in 2004 totaled approximately 49 Gt CO₂ equivalent (CO₂e)³, representing a 70% increase since 1970 (Rogner et al. 2007). The Intergovernmental Panel on Climate Change (IPCC) 4th Assessment Report concluded with “very high confidence” that the net effect of human activities since 1750 has resulted in “positive” radiative forcing (i.e. incoming energy greater than outgoing energy) of approximately 1.6 W m⁻² (Solomon et al. 2007). Global atmospheric concentrations of CO₂ now greatly exceed their natural range over the last 650,000 years, while atmospheric CH₄ and N₂O greatly exceed their natural ranges over the last several thousand years (Solomon et al. 2007).

More than 50% of the world’s population now lives in urban areas. Recent bottom-up⁴ estimates for approximately 100 cities across the globe (Hoornweg et al. 2011) have given

annual per capita GHG emissions ranging from more than 15 tons CO₂e (Sydney, Calgary, Stuttgart, and several major US cities) to less than half a ton (various cities in Nepal, India, and Bangladesh). Urban areas currently account for more than 70% of energy-related GHG emissions, and it is likely that more than 80% of total greenhouse gas emissions can be attributed to urban and associated suburban activities (Hoornweg et al. 2011). Therefore, improved quantification of urban-scale GHG sources and sinks is a

high priority for current research using both top-down atmospheric studies/models and bottom-up field-based measurement campaigns. Existing urban estimates are frequently based on downsizing national GHG inventories following IPCC-approved methodologies that take into account population, industrial production, and material flow models (IPCC 2006). Because these methods can have high uncertainties when applied at their intended national spatial and annual temporal scales, uncertainties are substantially increased at smaller scales. Conversely, the extrapolation of small-scale field studies to entire urban areas or regions can also introduce large errors. A summary of the main sources of world greenhouse gas emissions is shown in TABLE 1.

During the last decade, field measurement capabilities and modeling strategies yielding urban-scale data have greatly improved. This has led to the recognition that localized emissions trends are important for improved scientific understanding of sources and sinks, better integration of top-down and bottom-up measurement and modeling strategies, and more informed carbon- and nitrogen-management policy decisions. Various methods have been used to quantify urban fluxes of GHGs to the troposphere, including ground-based sampling, tower-based micrometeorological measurements, instrumentation on buildings, mobile measurements, and aircraft-based measurement strategies (Moriizumi et al. 1998; Nemitz et al. 2002; Grimmond et al. 2002; Soegaard and Møller-Jensen 2003; Vogt et al. 2006; Pataki et al. 2006; Henninger and Kuttler 2007; Mays et al. 2009). Stable isotope and radiocarbon data have been extensively used to track magnitude and sources of CO₂ emissions to the atmosphere. The carbon isotope composition of CO₂, often represented in a mixing diagram of δ¹³C versus 1/[CO₂] (referred to as a “Keeling plot”), has been used to constrain mixing proportions and the temporal variability of CO₂ sources (Newman et al. 2008). For CH₄ it can be a significant challenge to use isotopic mixing ratios to constrain local sources and

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3 CO₂e is defined as the amount of carbon dioxide (CO₂) that would cause the same extent of radiative forcing as a given GHG over a certain time period. It is given by the product of the total mass of the GHG and its “global warming potential” (GWP) factor. CH₄ and N₂O have GWP values of 25 and 298, respectively, on a 100-year time horizon (Solomon et al. 2007).

4 “Bottom-up” and “top-down” are two different approaches to the study of large, generally complex systems. Bottom-up approaches infer the characteristics of the large system based upon observation of its smaller subsystems, while top-down approaches observe the large system and infer the contribution of its subsystems. In the case of atmosphere, GHG studies, the “bottom-up” approach studies the GHG fluxes at the Earth’s surface to infer the changes in the atmosphere, and the “top-down” approach examines the atmospheric GHG concentration and changes to infer fluxes from land and ocean sources.

TABLE 1 PRINCIPAL SOURCES OF ANTHROPOGENIC GREENHOUSE GASES AND THEIR RELATIVE FLUXES IN PERCENTAGE CO₂ EQUIVALENT, CIRCA 2000

SECTOR		END USE/ACTIVITY	GHG SPECIES
Transportation	13.8%	Road	9.9%
		Air	1.6%
		Rail, ship, and other	2.3%
Electricity and heat	24.6%	Residential buildings	9.9%
		Commercial buildings	5.4%
		Unallocated fuel combustion	3.5%
		Iron and steel	3.2%
		Other fuel combustion	9.0%
		Chemicals	4.8%
		Industry	10.4%
		Cement	3.8%
		Fugitive emissions	3.9%
		Other industry	5.0%
Industrial processes	3.4%	Transmission & distribution losses	1.9%
		Oil/gas extraction, refining, and processing	6.3%
Land-use change	18.2%	Deforestation	18.3%
		Harvest/management	2.5%
		Afforestation/reforestation and other	-2.6%
Agriculture	13.5%	Agricultural soils	6.0%
		Livestock & manure	5.1%
		Rice cultivation	1.5%
Waste	3.6%	Landfills	2.0%
		Wastewater, other waste	1.6%

Data from World Resources Institute (www.wri.org). For clarity, only contributions greater than 1.5% are reported for the END USE/ACTIVITY, and therefore percentages do not necessarily sum up to the SECTOR percentages. Sector percentages are often a combination of two or more end uses/activities. Roughly 80% of global GHG emissions are from urban/suburban sources.

sinks due to the complexity of multiple local sources with differing isotopic compositions, as well as the temporal and spatial variability of microbial CH₄ oxidation rates through terrestrial soil profiles.

MEASUREMENTS AND MODELS

Continuous, real-time, in situ measurements of atmospheric H₂O, CO₂, CH₄, and N₂O concentrations and isotopic compositions at ambient conditions can now be made with commercially available, portable infrared absorption spectrometers. These instruments are yielding abundant high-precision data for improved understanding of localized GHG biogeochemical cycles.

Urban CO₂ Emissions

Carbon dioxide is the most abundant anthropogenic GHG. Its concentration in the atmosphere has increased from a preindustrial atmospheric concentration (i.e. mixing ratio) of 280 ppmv (parts per million by volume) to 394 ppmv in March 2012. It has an average atmospheric residence time that can extend to 30,000 years (Archer 2005). The current annual rate of increase is about 2 ppm as measured at Mauna Loa (www.esrl.noaa.gov/gmd/ccgg/trends/index.html#mlo). The largest fraction of anthropogenic CO₂ emissions, by far, is derived from fossil fuel combustion for electricity production, transportation, and industrial processes (US EPA 2012). Fairly accurate estimates of the fossil fuel combustion sources can be obtained from analysis of combustion technology and the production and sales records for a variety of fuel types throughout the economy, as well as from air pollution monitoring, traffic,

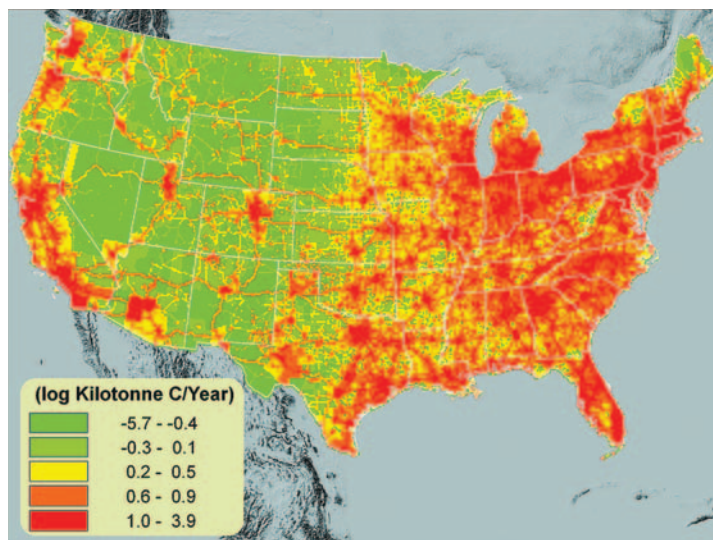


FIGURE 1 Total CO₂ emissions from fossil fuel combustion in the United States during 2002, on a 10 km × 10 km grid, in units of log kilotonne C/year. The pattern of CO₂ emissions correlates well with population density. REPRINTED WITH PERMISSION FROM GURNEY ET AL. (2009), COPYRIGHT AMERICAN CHEMICAL SOCIETY

and census data (Gurney et al. 2009). The areal distribution of annual CO₂ emissions from fossil fuel combustion during 2002 in the United States shows a strong spatial correlation with the distribution of urban centers (Fig. 1).

Urban CO₂ fluxes have been reported for a number of cities in terms of mass per unit area per unit time or annual mass per capita. Typical average values are 410 μg m⁻² s⁻¹ in southeast Mexico City (Velasco et al. 2005); 440 (nocturnal) to 1670 μg m⁻² s⁻¹ (hourly averages) in Chicago during summer (Grimmond et al. 2002); 840 μg m⁻² s⁻¹ in Indianapolis (Mays et al. 2009); and 1140 μg m⁻² s⁻¹ (or 10 kilotonnes km⁻² y⁻¹) in Edinburgh during autumn, with a range of 750 (night) to 1670 (day) μg m⁻² s⁻¹ (Nemitz et al. 2002). Interestingly, human exhalation of CO₂ was estimated to yield an average CO₂ flux of 88 μg m⁻² s⁻¹ in Edinburgh city center (Nemitz et al. 2002). There is a clear diurnal cycle reflected in every city studied to date (Fig. 2). In the center of Edinburgh, the increase in daytime CO₂ flux is from transportation activities, and nighttime flux is dominated by nontraffic sources such as residential heating.

The major sinks for atmospheric CO₂ are the biosphere, absorption by oceans, and long-term silicate weathering. The biosphere emits CO₂ by respiration at night (typically from 0 to 440 μg m⁻² s⁻¹ from grasslands and forests at temperate latitudes) but consumes CO₂ by photosynthesis during daylight hours (up to 1100 μg m⁻² s⁻¹ in actively growing forests). Although a large portion of the emitted anthropogenic CO₂ is removed by the more rapid sinks, a substantial fraction will remain in the atmosphere for many millennia, even if fossil fuel combustion ends today (Archer 2005).

Urban CH₄ Emissions

Methane (CH₄) is a potent GHG, with a 100-year global warming potential (GWP) of 25 (relative to CO₂ = 1), a current atmospheric mixing ratio of about 1.8 ppmv, an atmospheric lifetime of 12 years, and estimated emissions of 500–600 Tg CH₄ y⁻¹ (Solomon et al. 2007). Natural sources include wetlands (about 30% of the annual total) and smaller marine, geologic, and terrestrial sources (e.g. geologic leakages, termites, native ruminant animals). The many diverse anthropogenic sources of CH₄ include fossil

fuel production and transport leakages (natural gas, coal), agricultural emissions (ruminant animals and manures, rice production), biomass burning, and human waste-related emissions (landfills, wastewater).

The major sink for atmospheric CH₄ consists of reactions with tropospheric OH, while methanotrophic methane oxidation in aerobic soils and stratospheric reactions provide smaller sinks (Solomon et al. 2007). Because CH₄ has a relatively short atmospheric lifetime combined with a relatively high GWP, many countries have targeted CH₄ sources as a strategy to achieve meaningful GHG emission reductions within about a decade. A major problem with respect to CH₄ is that the relative annual contributions of individual sources and major driver(s) for observed inter-annual variability in atmospheric mixing ratios are poorly understood (Bousquet et al. 2006). In large part, this is due to multiple CH₄ sources with annual contributions of about 5% to 15% of the total global CH₄ emissions but with high annual and interannual uncertainties at smaller regional and urban scales.

Better quantification of atmospheric CH₄ sources and sinks at smaller urban and regional scales is essential for improved understanding of atmospheric CH₄ trends. A wide variety of approaches is being employed to elucidate this diverse-sourced gas, as shown in the following examples. Lamb et al. (1995) applied a tracer method using SF₆. Focusing on natural gas leakages at the urban scale, they measured a wide range of 0.0024 to 258 L CH₄ min⁻¹ from gas distribution facilities and total emissions of ~2 μg CH₄ m⁻² s⁻¹ from a small midwestern United States city. Using atmospheric isotopic mixing ratios (¹⁴C and δ¹³C), Moriizumi et al. (1998) concluded that urban CH₄ emissions in winter were dominated by natural gas leakages and auto exhaust. Annual anthropogenic CH₄ emissions from Los Angeles were estimated at ~4.2 Mt CO₂e using a top-down method based on atmospheric CH₄/CO₂ ratios (Hsu et al. 2010). London's CH₄ sources and emissions were estimated by a top-down method, relying on δ¹³C measurements and sampling during nighttime inversions at a basin site south-

west of the city (Lowry et al. 2001). The CH₄ source δ¹³C value, averaging -48.7‰, was apportioned mostly to natural gas leakages and waste-treatment emissions, and London's annual emissions for 1996 were estimated at 240–312 kt CH₄. A bottom-up static chamber method was applied by Kaye et al. (2004) to measure CH₄ emissions in the vicinity of Fort Collins, Colorado. They concluded that all systems exhibited uptake of atmospheric CH₄, with highest values for native grasslands (-33 to -67 μg CH₄ m⁻² h⁻¹), lower values with similar seasonal patterns for wheat soils, and lowest values for corn and urban lawn systems (less than -27 μg CH₄ m⁻² h⁻¹). In Los Angeles, Wunch et al. (2009) focused on atmospheric column measurements of CH₄ for the South Coast Air Basin. They found that large diurnal CH₄ variations correlated with CO₂ and CO, and they estimated the yearly emissions at approximately 0.6 Tg CH₄. Using an aircraft-based approach in which flux calculations were based on vertical concentration and wind-speed gradients, Mays et al. (2009) determined that CH₄ emissions in Indianapolis ranged between 18 and 72 μg CH₄ m⁻² s⁻¹, with major sources including combustion and noncombustion sources (natural gas distribution network, wastewater treatment facilities, landfill sites). Hecobian et al. (2011) used a back trajectory analysis method with forward plume movement estimation in a global study of CH₄ emissions from biomass burning (495 individual fire plumes), and they observed higher atmospheric CH₄ mixing ratios in plumes from urban sources. Bellucci et al. (2010) measured CH₄ emissions from wastewater treatment at three metropolitan activated-sludge water-reclamation plants in Chicago, finding negligible emissions from membrane-capped anaerobic digesters, emissions from primary tanks averaging 0.03–4.0 g CH₄ m⁻² h⁻¹, and higher emissions of about 100 g CH₄ m⁻² h⁻¹ from floating-cover anaerobic digesters.

Landfills in urban areas can emit CH₄ at rates ranging over several orders of magnitude; for example, in a recent field validation study at two California landfills for a new process-based model (Spokas et al. 2011), Bogner et al. (2011) measured seasonally variable average rates ranging from -0.04 to >100 g CH₄ m⁻² d⁻¹ depending on cover type (daily, intermediate, final) and soil microclimate-dependent CH₄ oxidation for each cover type (Spokas and Bogner 2011). As can be observed from the diversity of methods and results to date for urban CH₄ emissions and urban atmospheric CH₄ mixing ratios, a research priority is the continued development and application of multiple measurement techniques and field-validated models.

Urban N₂O Emissions

Nitrous oxide (N₂O) is both a greenhouse gas and an ozone-depleting substance. It has a 100-year GWP of 298, a current atmospheric mixing ratio of about 0.32 ppmv, a radiative forcing of 0.16 W m⁻², and an atmospheric residence time of 114 years (Solomon et al. 2007). There are no significant tropospheric sinks of N₂O, and slow photodissociation to N₂ in the stratosphere is the main mechanism of removal, resulting in its relatively long atmospheric residence time. The ozone-depleting potential of nitrous oxide is comparable to that of many hydrochlorofluorocarbons being phased out under the 1987 Montreal Protocol (Ravishankara et al. 2009), but it is not regulated by the Protocol as its full potential in a global warming perspective has become clear only in recent years.

Nitrous oxide is naturally produced by bacterial metabolism during nitrification and incomplete denitrification in many natural environments, particularly soils, oceans, estuaries, and rivers. The largest anthropogenic sources of atmospheric N₂O are fertilized agricultural soil (~69%),

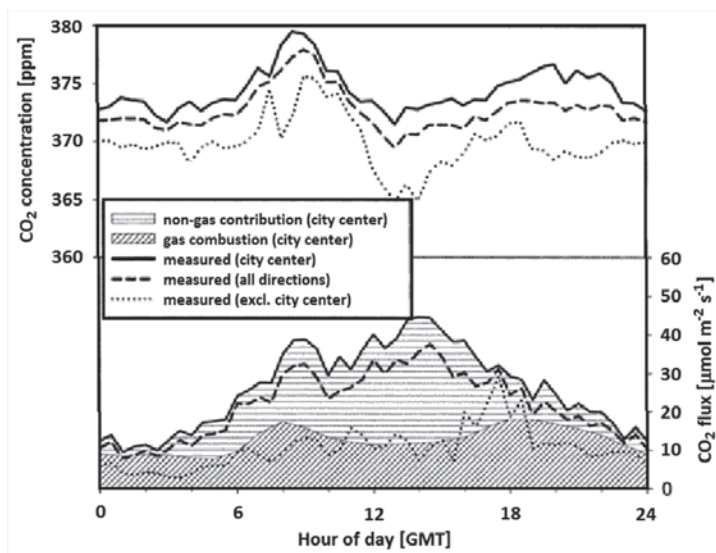


FIGURE 2 Diagram showing averaged diurnal concentrations and fluxes of CO₂ in central Edinburgh, UK, measured from a tower 65 m above street level, during the period October 28–November 30, 2000. Averages are shown from the following wind directions: city center, non-city center, and all directions. The measured city-center flux is divided into two components: combustion of natural gas (diagonally hatched area) and nongas sources including traffic, oil, and coal combustion (horizontally hatched area). REPRINTED WITH PERMISSION FROM NEMITZ ET AL. (2002), COPYRIGHT AMERICAN CHEMICAL SOCIETY

mobile and stationary combustion (~14%), manure management (~6%), nitric and adipic acid production (~6%), and wastewater treatment (~1.5%), which together account for about 310 Tg CO₂e (>96%) of the total anthropogenic N₂O emissions in the United States. This represents about 35% of the global (natural and anthropogenic) N₂O emissions (US EPA 2012). Emissions of N₂O at the urban scale are poorly constrained, as there are only a limited number of studies and fluxes have high uncertainty due to the presence of multiple temporally and spatially variable sources. Additionally, natural and anthropogenic sources are often interconnected; for example, it is not clear how much of the N₂O produced in coastal environments is due to natural nitrogen cycling and how much is contributed to natural waters by fertilizers during agricultural runoff. For these reasons, the global mass balance for N₂O is not yet clearly defined and is being adjusted as new studies become available.

In this respect, isotopic analysis is a powerful tool for tracing N₂O sources and the biological pathways responsible for its production (Stein and Yung 2003). In some instances, the interpretation of N₂O stable isotope data is challenging and can be inconclusive, since N₂O is produced via multiple coexisting biological pathways (Townsend-Small et al. 2011). However, the analysis of the isotopomeric N₂O composition (i.e. the site-specific N isotope distribution in the linear N–N–O molecule), under the right conditions (e.g. no further bacterial consumption of N₂O after its formation), can distinguish between N₂O produced by nitrification and denitrification (Sutka et al. 2006).

From an urban- and periurban-landscape perspective, N₂O emissions are nearly ubiquitous, and many point source measurements utilize static chambers for the determination of emissions and total fluxes. Urban ecosystems such as fertilized and irrigated lawns and grass fields have N₂O emissions comparable to those of highly managed and fertilized agricultural soils, and up to ten times higher emissions than unfertilized natural grasslands, wheat fields, and urban forests, based on studies from Fort Collins, Los Angeles, Phoenix, and Baltimore in the United States (Townsend-Small et al. 2011). Urban areas can contribute substantially to N₂O emissions and can be the major source around rapidly growing megacities with diminishing percentages of agricultural soils. In addition, mobile combustion accounts for ~7% of the total anthropogenic N₂O emissions in the United States and Canada, while stationary combustion accounts for another ~7%. In contrast with CO₂, combustion-related emissions of N₂O are not straightforward to estimate, as they depend on fuel properties, catalyst technology, and driving habits. Older vehicles generally show higher N₂O emissions than new vehicles; aggressive, congested, and cold-start urban driving may increase N₂O mixing ratios in the exhaust gas by up to 40 times compared to highway driving, due to lower catalyst temperatures (Graham et al. 2009).

Biological nitrification and denitrification during wastewater treatment contribute ~1.5% of the total anthropogenic N₂O emissions, according to US EPA (2012). Measured emissions from septic tank wastewater in treatment facilities are generally higher than those predicted by existing models, which generally underestimate emissions (Ahn et al. 2010). Considerable amounts of N₂O are released during wastewater treatment (FIG. 3), mainly (>85%) by partial denitrification from the aerated septic tanks (Bellucci et al. 2010). Such measurements, when properly taken into account, might dramatically increase estimates for net N₂O fluxes from wastewater treatment plants. The reduction of



FIGURE 3 Aeration basin for activated-sludge processing at a large metropolitan wastewater treatment plant, Stickney, Illinois. PHOTO: REIKA YOKOCHI

these emissions depends on correct design and operation of the biological treatment processes, as well as the chemical characteristics of the incoming wastewater.

Landfill emissions of N₂O can vary over several orders of magnitude, similar to the wide range of observed N₂O emissions from other terrestrial soil systems. In a recent landfill study in the Los Angeles area, average N₂O fluxes varied by almost two orders of magnitude; the highest values were associated with daily cover materials and were attributable to high moisture content, available N, and restricted aeration in the recently placed refuse (Bogner et al. 2011). The highest documented landfill fluxes have been observed from final cover soils amended with high-N materials, such as biosolids (Bogner et al. 2011), or from “semi-aerobic” landfills (Tsujiimoto et al. 1994). The maximum values for N₂O fluxes measured from composting operations (approximately 1–8 g m⁻² d⁻¹; Andersen et al. 2010) are generally higher than N₂O emissions from landfill cover materials.

OUTLOOK

The growing concern about levels of atmospheric GHGs and the realization that most of the anthropogenic GHG flux is from urban areas have led to enhanced global efforts to improve the quantification of urban GHG emissions. New measurement tools, from portable field spectrometers (for precise concentrations and isotope ratios) to satellite-based instrument platforms (for total column and profile measurements), are providing a wealth of new real-time data with high precision and spatial-temporal resolution. Predictive models for urban-scale GHG emissions are improving via advances in modeling combined with field validation at appropriate urban scales. Once the relative magnitudes of anthropogenic GHG emissions from different urban sources are sufficiently well known, more appropriate and cost-effective remedial strategies can be put into action.

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The position is a joint appointment: 50% at the University of Bern and 50% as head of the Laboratory for Waste Management (LES) at PSI. The anticipated starting date at PSI is January 2014. Employment at PSI will be 100% until the position at the University of Bern opens in August, 2015.

At the University of Bern the professorship carries responsibilities for leading a research group, for managing the Institute's mineralogy laboratories, for teaching mineralogy at BSc and MSc levels (BSc in German and MSc in English), and for selected administrative duties within the Institute of Geological Sciences and the Faculty of Science.

At PSI the position of Laboratory Head carries responsibility for managing the science and funding of the Laboratory for Waste Management, a research laboratory focused on physico-chemical processes in mineral systems suited for hosting waste repositories (in particular clays) with currently 24 scientists and technicians and access to a uniquely large concentration on site of state-of-the-art research infrastructure (X-ray synchrotron, neutron source, hot laboratory, etc.).

An established scientist is sought with an Earth Science background, a reputation for creativity in research, talent in teaching and experience in laboratory management. The ideal candidate will combine the most modern laboratory, analytical and theoretical approaches to address fundamental issues in clay mineralogy and its applications to geological waste disposal. A record of obtaining external research funds and a willingness to collaborate with other research groups in both Institutes are required.

Applications including a CV, a publication list, copies of the 5 most important publications and a statement of past and future research interests should be sent by email as single PDF file to: The Dean's Office, Faculty of Science, University of Bern. Email: andrea.perez@natdek.unibe.ch.

Deadline for the receipt of applications is February 15, 2013.

For informal enquiries please contact Prof. Dr. Laryn Diamond (diamond@geo.unibe.ch) at the University of Bern or Dr. Jean-Marc Cavedon (jean-marc.cavedon@psi.ch) at the Paul Scherrer Institute.

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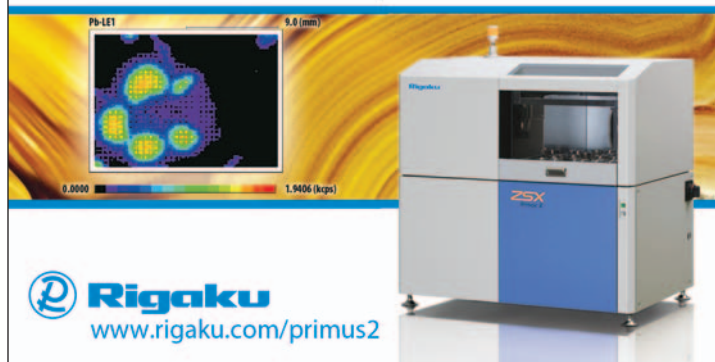


Red beryl from the Violet Claims, Beaver Co., Utah. Image by J. Scovill from the Photographic Guide to Mineral Species CD, available exclusively from Excalibur Mineral Corp.

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